

Outlet Temperature Correlation and Prediction of Transfer Line Exchanger in an Industrial Steam Ethylene Cracking Process^{*}

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Abstract Predicting the best shutdown time of a steam ethylene cracking furnace in industrial practice remains a challenge due to the complex coking process. As well known, the shutdown time of a furnace is mainly determined by coking condition of the transfer line exchangers (TLE) when naphtha or other heavy hydrocarbon feedstocks are cracked. In practice, it is difficult to measure the coke thickness in TLE through experimental method in the complex industrial situation. However, the outlet temperature of TLE (TLEOT) can indirectly characterize the coking situation in TLE since the coke accumulation in TLE has great influence on TLEOT. Thus, the TLEOT could be a critical factor in deciding when to shut down the furnace to decoke. To predict the TLEOT, a parametric model was proposed in this work, based on theoretical analysis, mathematic reduction, and parameters estimation. The feasibility of the proposed model was further checked through industrial data and good agreements between model prediction and industrial data with maximum deviation 2% were observed.

Keywords transfer line exchanger, outlet temperature, parametric model, steam ethylene cracking

1 INTRODUCTION

Scheduling of decoking of the steam ethylene cracking furnaces has attracted intensive attention in last decades [1–5]. Many scheduling models of Mixed Integer Linear Programming (MILP) or Mixed Integer Non-Linear Programming (MINLP) have been proposed, in which the scheduling models must be combined with a reliable simulation of the process [6] to give a practical guideline on the scheduling and planning of decoking. However, the transfer line exchangers (TLE) in the process have not been considered in the MINLP due to the difficulty of coking prediction in TLE. In addition, the outlet temperature of TLE (TLEOT) that changes with the run length of cracking process is a key parameter in practice. Thus, it is necessary to make prediction of TLEOT for the practical scheduling and planning models. As well known, the maximum run length of a cracking furnace is mainly determined by the coking situation in both the radiant coils and TLE. With the coke accumulated in the radiant coils, the heat transfer capacity decreases, and more fuel gas has to be burnt to compensate for the increased heat resistance and keep the coil outlet temperature (COT) constant. While with the coke increasing on inner wall of TLE, the high level of heat energy recovery from the cracked gas/steam mixture at high temperature will decrease and the TLEOT of cracking gas/steam mixture will increase due to the increasing heat resistance. However, the amount of fuel gas cannot be provided and the TLEOT cannot be allowed to increase unlimitedly in practice. Thus, the

furnace has to be shut to decoke when some upper limitations are reached.

The mechanisms of coke formation in TLE have received lots of attention, and several coke mechanisms have been proposed in literatures. Chen *et al.* [7] proposed a mathematic model by considering the droplets condensation of high boiling components in the cracked gas/steam mixture and water-gas reaction for the coking in TLE. The coking mechanism of mass transfer and condensation-surface poly-condensation reaction in TLE was proposed by Zou *et al.* [8]. They found the condensation process could be the controlling step of coking process when the surface temperature was higher than the transition point, otherwise, surface-poly-condensation reaction would be the controlling step. A comprehensive model in a transfer line exchanger tube incorporating a semi-empirical coke formation equation was proposed by Manafzadeh *et al.* [9] to predict the coke deposition rate both chemically and physically and the effect of this coke on the operational time of TLE. Van Geem *et al.* [10] performed an experiment on the coke formation under TLE conditions in an electro-balance setup, and found that the TLE material and its pretreatment had a strong influence on the coking rate, and the rate of coke deposition on the 15Mo3 cylinder was practically independent of the concentration of mono- and di-olefins and mono- and di-aromatics, but depended mainly on the temperature and water-to-hydrogen ratio. Other many efforts [11–16] to understand the coking problem have been made by academics and industrial researchers as well. However, the theory based models cannot be

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directly used to calculate and predict the changes of TLEOT in a complete run length.

Although some researchers [7, 8, 17] have presented some methods to predict the TLEOT, they didn't consider the effect of the flow rate change of feedstock on TLEOT. In order to be consistent with the industry reality of feed flow rate change, a correlation and prediction model of TLEOT in industrial steam ethylene cracking processes was proposed by the combination of theoretical analysis, mathematic reduction and parameters estimation. The feasibility of the proposed model was validated by the industrial data and it gave a good prediction for industrial data with a maximum deviation of 2%.

2 THEORETICAL ANALYSES AND MODEL REDUCTION

The steam cracking process transfers hydrocarbons such as ethane, liquefied petroleum gas (LPG), light naphtha, naphtha and hydrocracking tail oil (HVGO) into small hydrocarbons: ethylene, propylene and other products. Because of the complexity of the system, there are many factors affecting the production process. For the module of TLE, they are connected with the boiler drum, *i.e.* two to four TLEs coping with one boiler drum.

The cracked gas/steam mixture comes out from the radiant tubes at about 800 °C–860 °C and the temperature is usually kept constant for a certain type of feedstock. If the temperature is controlled well by advanced control strategy [18–20], the fluctuation of the temperature could be simply neglected.

Lack of advanced control to keep dilution steam/hydrocarbon ratio (SHR) constant, it may make the SHR fluctuate, and further lead to the change of products distribution, especially when the hydrocarbon feed flow rate changes. This could change the coking precursor concentration and the coking rate, so the real coking rate doesn't follow a regular principle.

When the coke accumulates to the limit inside the radiant tubes or the TLE tubes, the furnace has to be regularly switched off from the normal process to decoke. For a tubular cracking furnace, it has to be switched off to decoking when the tube maximum temperature (TMT), the pressure drop along the radiant tubes or the TLEOT rises to their tolerable upper limits. There are mainly three decoking methods such as steam decoking, steam-air decoking and air decoking to transform the coke into gas off the tube. However, the coke on the inside wall of the transfer line exchangers could only be removed partially. Due to the hardly-removed coke accumulated in the inside wall of TLE, it needs mechanical or hydraulic decoking to clear the TLE tubes periodically. Thus, the initial TLEOT will differ from each other in consecutive periods.

Based on the complex situation of TLE, the model reduction is necessary to obtain a parametric empirical model. The model parameters are then estimated by a robust nonlinear least-square method

based on the trust-region algorithm.

2.1 Heat transfer mechanism and reduction

The TLEs are always used to quench the high temperature cracked gas/steam mixture to stop the secondary reactions. The TLE usually includes the components of tube side and shell side, and it is connected to the boiler drum which provides the special processing water to recover heat from the cracked mixture at high temperature, as shown in Fig. 1.

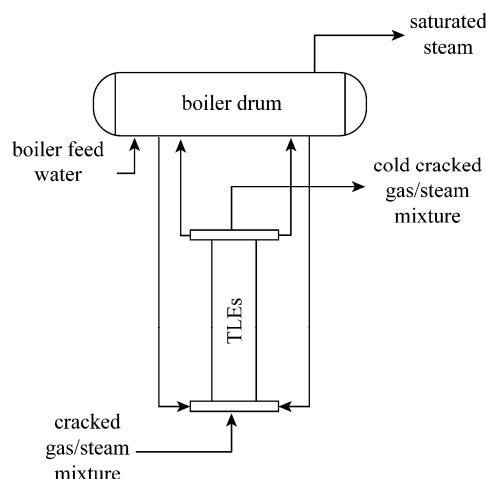


Figure 1 Diagram of TLEs connecting with the boiler drum

The overall heat transfer process in TLE could be expressed by the energy balance in Eqs. (1)–(3) [21]:

$$CF(T_i - T_o) = KA\Delta t_m \quad (1)$$

with

$$\Delta t_m = \frac{T_i - T_o}{\ln \left(\frac{T_i - T_s}{T_o - T_s} \right)} \quad (2)$$

$$\frac{1}{K} = \frac{D_o}{D_i} R_i + \frac{D_o}{D_i} \frac{1}{\alpha_i} + \frac{D_o}{D_c} \frac{\delta_c}{\lambda_c} + \frac{D_o}{D_w} \frac{\delta_w}{\lambda_w} + \frac{1}{\alpha_o} + R_o \quad (3)$$

where C is the heat capacity of cracked gas/steam mixture, F is the feed flow rate, T_i is the inlet temperature of the TLE (which equals the coil outlet temperature of cracked gas), T_o is the outlet temperature of the TLE, K is the overall heat transfer coefficient, A is the total heat transfer area, Δt_m is the logarithmic mean temperature difference, T_s is the boiling temperature of tube outside water. In Eq. (3) R_i and R_o are the inside and outside fouling heat resistance of TLE, respectively; α_i is the convective heat transfer coefficient of tube inside; δ_c and δ_w are the thickness of the coke and tube wall, respectively; λ_c and λ_w are the heat conductivity of the coke and tube wall, respectively; α_o is the tube outside wall boiling heat transfer coefficient, which is much larger than α_i ; D_o , D_i , D_c and D_w are the outside diameter of the tube, inside diameter of

the coke layer, arithmetic mean of inner and out diameters for the coke layer, and arithmetic mean of inner and out diameters for the tube, respectively. The overall heat transfer resistance in the TLE tube constitutes of six parts, as shown in Fig. 2.

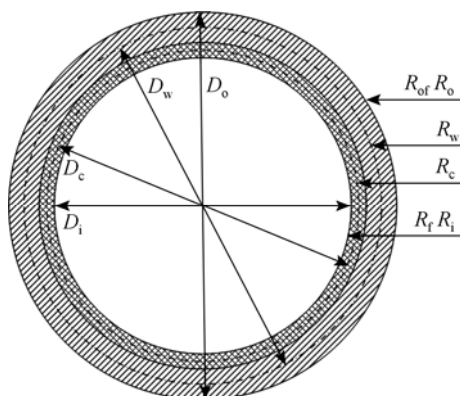


Figure 2 Diagram of TLE tube heat transfer resistance

Because of the predominance of the inside convective heat transfer resistance in the overall heat transfer, a proper correlation has to be used to calculate α_i . The well-known Dittus-Bolter correlation:

$$Nu = B \cdot Re^a \cdot Pr^b \cdot Gr^c \quad (4)$$

is selected because of its popularity [21], and B is the coefficient of Dittus-Bolter correlation. Here $Nu = \alpha d / \lambda$ shows the influence of the convection on the heat transfer; $Re = \rho d u / \mu$ the influence of the fluid flow state; $Pr = c_p \mu / \lambda$ the influence of the fluid physical properties, and for gas the value of Pr is about 1, while Pr for liquid is much larger than 1; $Gr = \beta g \Delta t l^3 \rho / \mu^2$ the influence of the natural convection, where the β and g are the volume expansion coefficient for the liquid and the gravitational acceleration, respectively.

Here, the following assumptions are made:

(1) $Re > 10000$, namely the flow is fully turbulent;
(2) $0.7 < Pr < 160$, and it holds for the general fluids;

(3) The fluid has low viscosity;

(4) $l/d > 30-40$, namely the fluid flow in the tube is fully developed, $B = 0.023$, $a = 0.8$, and if the fluid is heated, $b = 0.4$, if the fluid is cooled, $b = 0.3$.

Therefore, Eq. (4) can be reduced to

$$\alpha = 0.023 \frac{\rho^{0.8} c_p^{0.4} \lambda^{0.6} u^{0.8}}{\mu^{0.4} d^{0.2}} \quad (5)$$

To reduce the complexity of the heat transfer calculation in the TLE, the reference physical properties of the gas/steam mixture can be those at the average temperature of the inlet and outlet ones of the TLE. Considering the facts that the inlet temperature of the TLE is usually to keep constant, and the increase of 60°C between the start and end of the operation of TLE, the difference of the average of the inlet and

outlet temperatures of TLE between the start and end of the operation was about 30°C , which has small influence on the physical properties. Thus, the reference physical properties can be considered as constant. So the inside convection heat coefficient of the TLE can be approximately proportional to 0.8th power of the feed flow rate.

To reduce the calculation in Eq. (3), the heat transfer resistances from inside fouling, outside fouling, tube wall can be properly assumed as constant. Because the outside boiling heat coefficient is much larger than the inside convection coefficient, the heat transfer resistance from the outside boiling heat transfer could be neglected. Thus the total heat transfer resistance in Eq. (3) could be reduced to

$$\frac{1}{K} = R'_{cc} + \frac{R'_{ff}}{u^{0.8}} \quad (6)$$

where R'_{cc} is the heat transfer resistance from constant part and gradually increasing part of the coking, R'_{ff} the constant parameter related to the inside convection heat transfer, u the fluid flow rate.

Combination of Eqs. (1), (2) and (6), Eqs. (7)–(9) are obtained as follows:

$$\begin{aligned} \frac{CF}{KA} &= \frac{C}{A} \left(R'_{cc} F + F \frac{R'_{ff}}{\left(\frac{F}{\rho S} \right)^{0.8}} \right) \\ &= \left(\frac{C}{A} R'_{cc} \right) F + \left(\frac{C}{A} R'_{ff} \rho S^{0.8} \right) F^{0.2} \\ &= R'_{cc} F + R'_{ff} F^{0.2} \end{aligned} \quad (7)$$

$$\frac{1}{\ln \left(\frac{T_i - T_s}{T_o - T_s} \right)} = R'_{cc} F + R'_{ff} F^{0.2} \quad (8)$$

$$T_o = \exp(\ln(T_i - T_s)) - \frac{1}{(R'_{cc} F + R'_{ff} F^{0.2})} + T_s \quad (9)$$

where S is the total cross sectional area of tubes with feed flow rate F .

2.2 Coking mechanism

Many factors could affect the coke deposition rate and the type of coke that deposits in TLE, such as the temperature distribution in TLE, tube material, steam hydrocarbon ratio, coke precursors.

The formation of coke in TLE could involve three different reaction mechanisms [7, 10, 14]. The first one consists of metal surface catalyzed reaction between pyrolysis-formed carbon and the metallic tube wall, and the filamentous coke containing metal particles was formed. The second one is that the bulk gas polymerization of the pyrolysis products goes through undesirable secondary reactions with the active sites of coke surface in the high temperature zone of TLE. And the third mechanism is that the

heavy components with high boiling points such as aromatics, C_6^+ , etc., condensate on the tube wall with low temperature through condensation polymerization and dehydrogenation. Meanwhile, the coke in TLE can also be removed through water gas reaction. So the net coking rate is determined by the difference between the forming and removing rates of coke in TLE.

Kern and Seaton [22] proposed an empirical fouling model as shown below:

$$\frac{dR_f(t)}{dt} = \phi_d - \phi_r \quad (10)$$

$$R_f(t) = R_{f,c} [\ln(t) / \ln(t_{p,c})] \quad (11)$$

Here, the rate of fouling deposition ϕ_d depends on the type of fouling mechanism, while the rate of fouling removal ϕ_r depends on the hardness or adhesive strength of the deposit and the shear stress due to the flow velocity, as well as the system configuration. Although many investigators have proposed some different forms of empirical rates for the deposition and removal in different areas (depending on the type and mechanism of fouling), the time-dependent behavior of fouling process can be mainly characterized by four types of models, that is, linear, power-law, falling-rate and asymptotic fouling models [22–24]. Combined with the studies [7–17, 25] of the coking deposition in the TLE and coking removal, the falling-rate fouling model was chosen as the fundamental type of empirical time-dependent coking fouling model, and the probabilistic falling-rate fouling model can be reduced in to a deterministic one for simplification, and the reduced model is Eq. (11), where the $R_{f,c}$, $t_{p,c}$ are constant parameters. In our work, it was difficult to get the real beginning time of the coking fouling in the industrial case, so another parameter was added. Then, the coking fouling empirical model is represented by

$$R_f(t) = R_{f,c} [\ln(t + p_b) / \ln(t_{p,c})] \quad (12)$$

Replacing $R_{f,c} / \ln(t_{p,c})$ by p_a leads to Eq. (13):

$$R_f(t) = p_a \ln(t + p_b) \quad (13)$$

The coking fouling $R_f(t)$ in Eq. (13) corresponds with the coke related parameter R_{cc} in Eqs. (7)–(9), so R_{cc} can be expressed in a parametric model in Eqs. (14) as follows:

$$R_{cc} = p_a \ln(t + p_b) \quad (14)$$

where p_a , p_b are the constant parameters.

2.3 Model formulation

The outlet temperature of the transfer line exchanger (TLEOT, T_{tleot}) could be modeled by a parametric model in Eqs. (15)–(16).

$$\frac{1}{\ln\left(\frac{T_{cot} - T_s}{T_{tleot} - T_s}\right)} = p_a F \ln(t + p_b) + R_{ff} F \quad (15)$$

$$T_{tleot} = \exp\left\{\ln(T_{cot} - T_s) - \left[p_a \ln(t + p_b) F + R_{ff} F^{0.2}\right]^{-1}\right\} + T_s \quad (16)$$

The parameters p_a , p_b and R_{ff} in Eq. (15) can be determined by the industry data containing the coil outlet temperature (COT , T_{cot}), T_{tleot} and saturation vapor temperature (T_s), respectively. The parametric model considers the effects of the flow rate of feedstock and the run length on T_{tleot} , and the reduction model is proper for the industrial practice.

2.4 Model parameters estimation

Once the TLEOT mathematic expression are determined through theoretical analysis, the model parameters can be directly obtained by fitting the expression with the industrial data by nonlinear least squares based on bisquare weights for robust and trust-region for the optimization algorithm.

The least square method is to minimize the sum of the squares of errors between the theoretical and real data. Due to some reasons, errors would arise onto data points, and they usually follow a normal distribution. These situations and other uncertainties make the conventional nonlinear least squares optimization disappointing. Besides, the conventional least-squares fitting has its high sensitivity to the abnormal data points, which have great influence on the fitting results because of squaring magnifies their effects. So the robust nonlinear least squares optimization procedure with trust-region optimization algorithm [26–29] was employed for the parameter estimation in the current work. The robust nonlinear least squares method minimizes a weighted sum of squares. Here the nonlinear least squares fitting based on bisquare weights for robust and trust-region for the optimization algorithm was used to implement the parameters estimation procedure.

3 RESULTS AND DISCUSSION

The online decoking method in the plant can only remove the coke in the inner tube of TLE partly, resulting in different initial coke amount for different run lengths. To reduce this impact on the model, the industrial data of T_{tleot} for cracking naphtha feedstock in one run length, which were just decoked by mechanical or hydraulic decoking method, were used. The obtained parameters for cracking naphtha are shown in Table 1. The fitting results show goodness of root-mean square error (RMSE) at 0.01937 and R-square at 0.96, it was demonstrated that the assumption of the model was reasonable in certain extent.

Table 1 The obtained parameters and the goodness of the result

Parameter	p_a	p_b	R_{ff}	RMSE	R-square
value	0.00564	1.81	0.253	0.0194	0.960

Figure 3 shows the industrial data and the correlation obtained from Eq. (16), and it can be seen that the absolute errors are all less than 10 °C. And it can also be found that if the flow was at the range of 22–25 t·h⁻¹, smaller errors can be observed; otherwise the errors would increase. This indicates the feasible range of the flow in the proposed model in Eq. (16). Fig. 4 shows the theoretical T_{leot} for different flow rates calculated by the proposed models. It can also be seen that the TLEOTs increase fast at initial several days and then gradually increase slowly, and larger flow results in higher TLEOT. Those results are well coincident with the industrial practice. The coke related parameter R_{cc} in Fig. 5 presents that the coke increases fast at initial and then slowly, and dR_{cc}/dt shows that the coke deposition rate is large at initial and then decreases with time. These results are well coincident with the reported results in literatures [7–16] and practice.

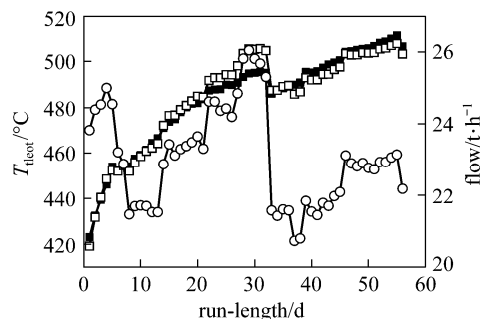


Figure 3 Comparison between the theoretical and industrial results for TLEOT

○ Flow rate; ■ industrial data; □ model predicted

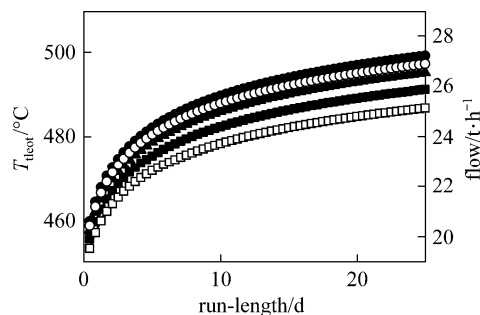


Figure 4 Theoretical T_{leot} for different flow rates

flow rate/t·h⁻¹: □ 21; ■ 22; ▲ 23; ○ 24; ● 25

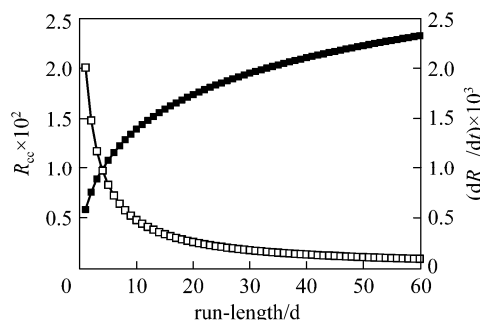


Figure 5 Coke related R_{cc} and dR_{cc}/dt

□ dR_{cc}/dt ; ■ R_{cc}

4 MODEL VALIDATIONS

The proposed correlation model would have no meaning in the real process industry if it was not validated soundly with more sets of industrial data. Once the initial states of the furnace and transfer line exchanger in a runlength are known, such as the feed flow rate F and T_{leot} , the future T_{leot} in the runlength could be predicted by the proposed model by the addition of the correction bias (D_{bias}) which takes into account the different initial coke amount (resulted from hardly-removed coke remaining after online decoking) for the different run length. So the proposed prediction model turns into

$$T_{leot} = \exp \left\{ \ln(T_{cot} - T_s) - \left[p_a \ln(t + D_{bias} + p_b)F + R_f F^{0.2} \right]^{-1} \right\} + T_s \quad (17)$$

Several other runlengths of industrial data of naphtha cracking have been used to validate the developed prediction model for T_{leot} as shown in Figs. 6–8. D_{bias} for each runlength was well tuned by the initial (first day) states of the prediction runlength. So the more coke amount at the initial stage for the prediction of T_{leot} results in larger D_{bias} . D_{bias} is about 5 in Fig. 6. For another runlength of about 52 days in Fig. 7, D_{bias} is about 3.5, and the maximum prediction error is 10 °C. This occurred at low feed flow rates, but smaller prediction errors were observed at feed flow rates of

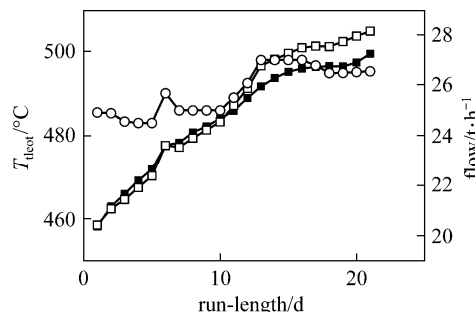


Figure 6 Comparison of TLEOT between theoretical and industrial results for one case with $D_{bias}=5$

○ flow rate; ■ industrial data; □ model predicted

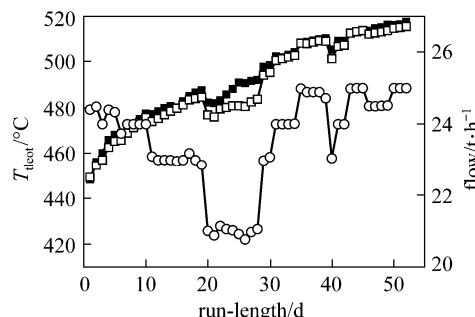


Figure 7 Comparison of TLEOT between theoretical and industrial results for one case with $D_{bias}=3.5$

○ flow rate; ■ industrial data; □ model predicted

23 t·h⁻¹–25 t·h⁻¹. While D_{bias} is about 400 in Fig. 8, which is so large, and this is due to both initial hardly-removed coke and that produced by heavy hydrocarbon HVGO in process for the first 17 days of the run-length. As for HVGO cracked in the furnace, the coke amount deposition in the TLE is much more than that from naphtha, and in the first 17 days the coke deposition amount in the TLE is more serious than that by cracking naphtha process, so a larger D_{bias} was used to tune the model for naphtha in Fig. 8. Meanwhile, the big D_{bias} means that the furnace needs to decoke by mechanical or hydraulic decoking method. After all, it can be found that the maximum absolute deviations of the T_{tleot} are mostly less than 10 °C, limited to 2%. Considering industry reality, the prediction of the proposed model is reasonable.

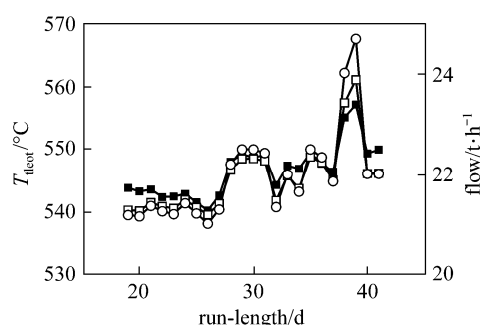


Figure 8 Comparison of TLEOT between theoretical and industrial results for one case with $D_{\text{bias}}=400$

○ flow rate; ■ industrial data; □ model predicted

5 CONCLUSIONS

Taking into account the effects of feed flow rate and coke formation in TLE on the outlet temperature of TLE, a prediction model of outlet temperature of TLE in industrial steam ethylene cracking furnace was developed through theoretical analysis, mathematic reduction and parameters estimation. The proposed model was validated with industrial data from different run-lengths, and gave good correlation and prediction with the maximum deviation 2%. The proposed model is proved to be reasonable and could give a helpful guideline to the operators and the dispatchers in industrial practice.

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NOMENCLATURE

A	total heat exchange area, m ²
a	exponents for the dimensionless numbers of Re
b	exponents for the dimensionless numbers of Pr

C	heat capacity of the fluid inside of the tube, J·kg ⁻¹ ·°C ⁻¹
c	exponents for the dimensionless number of Gr
c_p	heat capacity, J·kg ⁻¹ ·°C ⁻¹
D_{bias}	correction bias considering the initial coke amount in the TLE
D_c	arithmetic mean of inner and out diameters for the coke layer, m
D_i	inside diameter, m
D_o	outside diameter, m
D_w	arithmetic mean of inner and out diameters for the tube, m
d	diameter, m
F	feed flow rate of the furnace connected with three transfer line exchangers, kg·s ⁻¹
Gr	Grash of number
K	overall heat transfer coefficient in transfer line exchanger
Nu	Nusselt number
p_a	parameter for the proposed parametric model
p_b	parameter for the proposed parametric model
Pr	Prandtl number
R_{if}	parameter for the proposed parametric model related with the convection heat transfer
R_{cc}	coke related variable in TLE
R_{if}	inside fouling heat transfer resistance of the transfer line exchanger
R_{of}	outside fouling heat transfer resistance of the transfer line exchanger
Re	Reynolds number
T_{cot}	coil outlet temperature for the radiant tube in the furnace, °C
T_{tleot}	outlet temperature of the considered transfer line exchangers, °C
T_i	inlet temperature of the transfer line exchanger, °C
T_o	outlet temperature of the transfer line exchanger, °C
T_s	saturation vapor temperature, °C
t	time in run-length, s
ΔT_m	logarithmic mean temperature difference, °C
u	flow rate, m·s ⁻¹
α_i	tube wall inside convective heat transfer coefficient, W·m ⁻² ·°C ⁻¹
α_o	tube wall outside boiling heat coefficient, W·m ⁻² ·°C ⁻¹
δ_c	thickness of the coke layer, m
δ_w	thickness of the tube wall, m
λ	heat conductivity, W·m ⁻¹ ·°C ⁻¹
λ_c	the heat conductivity of the coke, W·m ⁻¹ ·°C ⁻¹
λ_w	heat conductivity of the tube wall, W·m ⁻¹ ·°C ⁻¹
μ	viscosity, kg·m ⁻¹ ·s ⁻¹
ρ	density, kg·m ⁻³

Subscripts

c	coke
i	inside
o	outside
w	wall

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