Chem. Bull. "POLITEHNICA" Univ. (Timişoara)

Kinetic Models Adaptation of Catalytic Cracking Unit

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Abstract: The catalytic cracking unit represents a vital element for any refinery specialized in the motor fuel production. The numerical modeling and simulation is the first important step in a process control system development. The model of the catalytic cracking process is characterized by a relative big number of the parameters. This paper presents the research results obtained by modeling, adaptation and simulation of the catalytic cracking process. The authors have identified four components associated to the process adaptation of the catalytic cracking model's. The first component represents the reactor constructive data. The second components refers to the adaptation of the model catalyst the phisical – chemical properties. The third components attend the estimation of the phisical – chemical properties of the raw material from experimenatal data. The last component contains the methodology of the kinetic model's parameters adaptation.

1. Introduction

The catalytic cracking process reprezents an essential point in the rafineries structure. The process and implicit the catalytic cracking unit are caracterized by a high degree of complexity and an economical profitability. The catalyts preformance and the advanced control of the process contribute to increase the profit and the gasoline production. One of the omponents of the advanced control and the performances of this system are directly determined by the process mathematical model.

The mathematical process model of FCC is treated under various aspects in numerous works Some of the works deal with the kinetic models of the catalytic process [1, 2, 3]. Another category of works is focused on the reactor modelling, in a steady-state or dynamic regime, using a certain kinetic model of the catalytic cracking process [4, 5, 6].

One delicate probleme of modelling is reprezented by model adaptation. The result of this operation is numerical values set associated to the parameter of the mathematical model. For catalytic cracking unit, mathemathical models have a big numbers of parameters, reaching few zecimal parameters.

The determination of this parameters reprezente an operation of identification in steady – state regime, and depend by a large meassured experimental data. Because of the economic importance of the catalytic cracking unit, the access to experimental data and the publishing of this data is restricted and the results obtaind in model adaptation operation is alomost impossible.

This paper try to reduce this gap, and the authors present the research results accomplish in the last few years in modelling, adaptation and simulation of the catalytic cracking process.

The start point of this paper is Weekman kinetic model, the model adaptation by authors is proper for advance control. [7]

Base on this kinetic model, the authors have elaborated the mathemathical model of the reactor associate catalytic cracking process. An important part of this paper is dedicated to adaptation of the process's kinetic model and mathematical model of the reactor. The authors have identified four componets for adoptation to the catalytic cracking process and the unit: constructive data of the reactor, the phisical- chemical properties of the catalyst, phisical properties of the raw material and estimation of the kinetic parameteres associated models.

2. The kinetic model of the catalytic cracking process

The kinetic model used in control system of the process must be relative simple and describethe global phenomenal that has occur in the riser. Based on this criterions was selected the Weekman kinetic model. This model is characterized by three chemical components, that are made by distillate limit in : base material (diesel oil) - A, gasoline -B, gas and coke - C, figure 1.

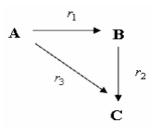


Figure 1. Kinetic diagram Weekman 's.

The rates of the three reactions from kinetic diagram are definition on these relations from table 1.

TABLE I. The kinetic equations

Reaction	Kinetic equation
1	$r_1 = -k_1 * Y_A^2$
2	$r_2 = -k_2 * Y_B$
3	$r_3 = -k_3 * Y_A^2$

 k_1, k_2, k_3 are dependent of the raw material quality, riser temperature, activity equilibrium catalyst and operating condition [8]. For determination of this constant is used relation

$$k_{j} = k_{j}^{0} \cdot f_{j}(MAT) \cdot \theta_{j}(t_{c}) \cdot \gamma_{j}(c_{cocs}) \cdot e^{\left(\frac{E_{j}}{R}\right)\left(\frac{1}{T_{0}} - \frac{1}{T}\right)},$$

where k_j^0 represents reaction rate constant at 482°C; $f_j(MAT)$ - attenuation function, that takes in consideration the effect of the equilibrium catalyst's activity; $\theta_j(t_c)$ - diminishing function, that takes in consideration the effect of the coke catalyst's deactivation; $\gamma_i(c_{cocs})$ - diminishing function, that takes in consideration the effect of the remanent coke; E_j activation energy of the chemical reaction *j*; *T*, *T_{ref}* - riser and reference temperature.

The *rate constants* k_j^{θ} are dependint on the base material structure and quality, and is characterized on aromatic carbon and naftenic carbon ration, C_A/C_N :

$$k_j^0 = e^{l_{0j} * \ln\left(\frac{C_A}{C_N}\right) + l_{1j}}, \quad j = 0, 1;$$
 (2)

$$k_2^0 = 5.55 \times 10^{-2}$$
 [h⁻¹]; (3)

$$k_3^0 = k_0^0 - k_1^0 \,. \tag{4}$$

The activity of the equilibrium catalyst is experimental determined using a standard method [9], and the effect of equilibrium catalyst activity's is inclusion in diminishing functions [10]:

$$f_1 = \ln \frac{MAT}{1 - MAT} \tag{5}$$

$$f_j = a_{0j} + a_{1j} \cdot MAT + a_{2j} \cdot MAT^2, \quad j = 2,3.$$
 (6)

The *catalyst's deactivation* due to coke formations in reactor is taken in consideration by a diminution's function in relation to the contacting time between the catalyst and the raw material. The structure function $\theta_j(t_c)$ is exponential type

$$\theta_j(t_c) = e^{\left(-a_{dec} \cdot t_c\right)},\tag{7}$$

where a_{dez} is rate deactivation constant

$$a_{dez} = 43.5 - 7.56 * (C_A / C_N)^{-1}.$$
 (8)

The efficiency of the catalyst regeneration is expressed by the function $\gamma_j(c_{cocs})$, the form of the attenuation functions being as follows [11]:

$$\gamma_1 = 1 + 0.151 * c_{cocs}; \tag{9}$$

$$\gamma_2 = 1 - 3,125 * c_{cocs} ; \tag{10}$$

$$\gamma_3 = 1/(1+3,53*c_{cocs}). \tag{11}$$

Remanent coke content is expressed by the dependence of the regenerator's temperature T_{reg} .

$$c_{cocs} = 4.31 * 10^8 \ e^{-(0.568 + 0.03195 T_{reg})} \ . \tag{12}$$

3. The mathematical model of the reactor

The catalytic cracking reactor is compose by the interfusion nod subsystem and the riser subsystem.

The interfusion node. The model of the interfusion node is represented by a heat balance in the steady state regime, and the feed sctock is complety vaporized on the base of the riser. [12] The temperature of the interfusion node T_{in} is calculated with the relation

$$T_{in} = \frac{Q_{cat1} * C_{pcat} * T_{reg} + Q_{mp} * C_{pmp} * T_{mp} - \Delta H_{vap} * Q_{mp}}{Q_{cat1} * C_{pcat} + Q_{mp} * C_{pmp}}.$$
(13)

The risre subsystem. The riser is considered a tube reactor with a total displacement operated in an adiabat regime. The riser mathematical model contains theree material balance equations on component and one heat balance equation.

The material balance equations systems have the form

$$\begin{cases} \frac{dY_A}{dz} = -\frac{1}{U_v} * (k_1 + k_3) * Y_A^2 \\ \frac{dY_B}{dz} = \frac{1}{U_v} * (k_1 * Y_A^2 - k_2 * Y_B) \\ \frac{dY_C}{dz} = \frac{1}{U_v} * (k_2 * Y_B + k_3 * Y_A^2) \end{cases}$$
(14)

The heat balance lengthways riser is represented by differential equation

$$\frac{dT}{dz} = \left(-\frac{dY_A}{dz}\right) * \frac{\left(-\Delta H_{r1}\right)}{\left(Y_A C_{pA} + R_{abur} C_{pabur} + R_{cat} C_{pcat}\right)},$$
(15)

and initial condition from the differential ecuation system are

$$\begin{cases}
Y_A(0) = 1 \\
Y_B(0) = 0 \\
Y_C(0) = 0 \\
T(0) = T_{in}
\end{cases}$$
(16)

The variables are:

Y- mass fraction for pseudo-component of reaction;

z- space coordinate in the riser;

 U_V – vapor's rate in riser;

 ΔH_{r1} - enthalpy of reaction 1;

 C_p – heat capacity;

 R_{abur} – ratia de abur de stripare;

 R_{cat} – the raport catalyst/ fresh feed flow;

 T_{in} – the temperature of the interfusin nod.

Base on the mathematical model was elaboreted an simulation program for reactor associate the catalytic cracking process.

4. The adaptation of the models

The authors identified four components associated to the adaptation of the mathematical models of the catalytic cracking process:

- the reactor constructive data;
- the thermodynamic properties of the catalyst;
- the estimation of some phisical properties of the material flows;
- the adaptation of some kinetic parameters of the kinetic model.

4.1. Reactor constructive data

Within the first component of the model adaptation phase, there are included the geometrical specifications of the reactor, table 2.

TABLE 2. Reactor constructive data

Parameter	Variable	Value	Fundamental units
Reactor area	A_r	1.32	m ²
Height riser	H_{riser}	35	m

4.2. Catalyst phisical properties

The catalyst physical properties are determinated experimental or are take it from liteterature. In table 3 are presented the values of the catalyts properties used in catalytic cracking unit.

Parameter	Variable	Value	Fundamental units
Heat capacity of catalyst	$C_{p,cat}$	0.28	kcal kg ⁻¹ grd ⁻¹
Catalyst dezactivation factor	MAT	0.68	-
Mass ration of dipersion steam into riser	Rabur	0.0015	-

4.3. Phisical properties of the material flows

The thermodynamic properties of the feed stock can be determined experimentally, calculated with empirical relations or estimated based on the data from literature. In table 4 are preseted the expressions that are used in determination of the phisical –chemical properties of the raw material.

The research activity of the authors has been als focusd to obtain operate industrial date. Thus, was collected data from procces for a autochthonous catalytic cracking unit, table 5. Based on data that are characterized the raw material (density, sulphur and averange bolling point temperature) were calculed the next phisical properties: characterization factor, heat capacity, mass molar averge, molar average boiling point, aromatic, parafinic, naphtenic carbon concentration. In the table 6 are preseted the valus of this phisical properties of the raw material.

4.4. The adaptation of the kinetic parameters

The success of the mathematical model application for the catalytic cracking unit reaction block depends in a grathdeal on the accuracy of the kinetic parameters. The authors have identified the next step of the kinetic model adaptation:

- specification the energy activation for each chemical reaction;
- numerical estimation of the coefficients of the kinetic model.

Parameter	Variable	Relation	Formula	Fundamental units
Characterization factor	K	(17)	$K = \frac{\sqrt[3]{1,216(T_{mv} + 273)}}{d}$	-
Heat capacity	$C_{p,mp}$	(18)	$C_{p,mp} = \frac{4-d}{6450} * (1.8 T_{mv} + 702) * (0.0935K - 0.111)$	kcal kg ⁻¹ grd ⁻¹
Liquid enthalpy	h	(19)	$h = (2,964 - 1,332 d)T + (0,003074 - 0,002254 d^{2})T^{2}$	kJ kg ⁻¹
Vapour enthalpy	Н	(20)	$H = -304,23 - 210,61 d + (6,0083 - 0,45638 d) T + (0,0023447 - 0,00059037 d) T^{2}$	kJ kg ⁻¹
Molar average boiling point	T _{mm}	(21)	$T_{mm} = T_{mv} - \left[\frac{T_{90} * T_{10}}{170 + 0.075T_{mv}} + 1.5\right]^{1/3}$	°C
Mass molar average	M_{mol}	(22)	$M_{mol} = 0.013679 \times e^{-0.09776 \ln d} \times e^{1.6071 \ln Tmv}$	kg kmol ⁻¹
Aromatic, parafinic, naphtenic carbon concentration	$C_A \\ C_N$	(23)	$w = d - 0.851 - 1.11 (n - 1.475)$ $v = 2.51 (n - 1.475) - d + 0.851$ $C_R = \begin{cases} 820w - 3S + \frac{10000}{M}, & w \ge 0\\ 1440w - 3S + \frac{10000}{M}, & w < 0 \end{cases}$ $C_A = \begin{cases} 430v + \frac{3660}{M}, & v \ge 0\\ 670v + \frac{3660}{M}, & v < 0 \end{cases}$ $C_N = C_R - C_A$	%

TABLE 5. The feed properties and the operating data [Brazi]

		Feed p	roperties			(Operating data	1	
Day	Density	Sulphur [% weight]	Boiling point temperature at 10% volume [°C]	Averange boiling point temperature [°C]	Feed temperature [°C]	Feed flow rate [kg/h]	Regenerator temperature [°C]	Catalyst ratio	Reactor temperature [°C]
1	0.912	0.40	299	378.0	203	176292	711	6.0	531
2	0.914	0.40	286	370.0	200	177083	710	6.0	531
3	0.900	0.36	289	356.0	202	524250	711	5.5	531
4	0.912	0.34	302	374.0	207	154333	709	6.0	530
5	0.905	0.36	275	368.0	205	154792	706	6.2	530
6	0.907	0.35	263	364.0	203	149042	706	6.2	530
7	0.907	0.32	262	365.0	196	155667	708	6.2	530
8	0.907	0.30	286	368.0	195	478330	709	5.7	531
9	0.904	0.31	283	367.0	193	162292	709	6.3	531
10	0.908	0.37	284	367.0	194	157792	709	6.2	531
11	0.911	0.35	275	367.0	189	161750	710	6.2	531
12	0.912	0.15	300	370.0	191	501458	711	6.2	531
13	0.910	0.39	287	372.0	191	171000	710	5.6	530
14	0.909	0.37	298	370.0	190	167083	712	6.2	531
15	0.907	0.32	281	366.0	191	169542	711	6.1	532
16	0.907	0.36	288	371.0	200	512292	710	6.1	531
17	0.906	0.33	282	370.0	198	170375	710	5.6	531
18	0.906	0.34	280	363.0	205	173167	710	6.1	531
19	0.910	0.36	287	368.0	205	174292	712	6.0	531
20	0.909	0.35	286	365.0	205	167333	712	5.9	531

TABLE 6. Averange measurated	l and	estimated	oil	feed	properties
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Type of propertie	Properties	Averange value	Fundamental units
	Density	0.9080	-
Measurated	Sulphur	0.34	% weight
	Boiling point medium temperature	367.4	°C
	Molar boiling point temperature	358.7	°C
	Molar mass	311.5	kg kmol ⁻¹
	Characterization factor	10.13	-
Estimated	Heat capacity	0.546	kcal/kg grd
	Refraction indice	1.50	-
	Aromatic carbon	23.83	%
	Naphtenic carbon	23.17	%

The first step of the kinetic model adaptation is determination of the energies activation for chemical reactions. In case of the Weekman kinetic model, the values of the energy activation values are taking from literature, table 7, [13].

TABLE 7. The energy activation values

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Reaction	Energy activation [kcal kmol ⁻¹]
1	10,000
2	18,000
3	10,000

The second step of the operation adaptation is determination the coefficients of the Weekman kinetic model. The kinetic model can be write under matrix forms

$$Y = H(X), \tag{24}$$

Where *Y* represent the output variables vector, $Y^{T} = [T_{R}, Q_{B}]; \quad X - \text{input variables vector,}$ $X^{T} = [d, s, T_{mv}, T_{mp}, T_{reg}, R_{cat}, Q_{mp}].$

The variables are:

d – density of the raw material;

- s the sulphur contain in the raw material;
- T_{mv} the boiling point medium temperature of the raw material;
- T_{mp} raw material temperature after the prehiting furnace;
- T_{reg} the catalyst temperature from regenerator;
- R_{cat} the raport catalyst/raw material;
- Q_{mp} raw material flow;
- T_R ouput temperature from reactor;
- Q_B gasoline flow.

Taking into account the experimental data that are preseted in table 5, these included input and output of the model (24) and it is thought that industrial measurement of the temperatures is in good conditions, can be genereted the next relation

$$F_{ob} = \sum_{j=1}^{m} \left[\left(T_{R,j}^{\exp} - T_{R,j}^{\text{mod}} \right)^2 \right],$$
(25)

where $T_{R,j}^{exp}$ represented the experimantal temperature of the reactor for *j* day ; $T_{R,j}^{mod}$ - the estimation temperature whit (16) model for input data of the *j* day .

In the proposed kinetic model, there were defined the following approximation functions:

 - k_j⁰, the approximation function of the reaction rates constants at 432°C temperature

$$k_{j}^{0} = e^{l_{0j} * \ln\left(\frac{C_{A}}{C_{N}}\right) + l_{1j}}, \quad j = 0, 1;$$
 (26)

- $f_j(MAT)$, the attenuation function that takes into account the effect of the equilibrium catalyst activity

$$f_{2,3} = a_{0j} + a_{1j} \cdot MAT + a_{2j} \cdot MAT^2 .$$
 (27)

As part of the four aproximations functions are 10 kinetic parametres. Using the variable

$$\boldsymbol{Z}^{T} = \begin{bmatrix} a_{02}, a_{12}, a_{22}, a_{03}, a_{13}, a_{23}, l_{00}, l_{10}, l_{01}, l_{11} \end{bmatrix},$$
(28)

the expression (25) is convertied in a multivariable function, line

$$F_{ob}(\boldsymbol{Z}) = \sum_{j=1}^{m} \left[\left(T_{R,j}^{exp} - H_j(\boldsymbol{Z}) \right)^2 \right].$$
(29)

For the minimization of the objective function (29) was used the cyclicial exploration algorithm with contant step [14]. The algorithm deteminte the minimum of the nelinear, multivariable objective function without restrictions and without used the derivation of the function. The point $\mathbf{Z}^{(0)}$, the initial solution, was define by Mateos, and the values are presented in table 8. Because the initial solution has values by unit order, the exploration step selected is 0.01. In table 8 are presented comparative results obtained for different initial conditions of the $\mathbf{Z}^{(0)}$ variable. To start up with initial solution, obtain the an optimal solution $\mathbf{Z}^{(1)}$, for that only $z_4^{(1)}$ variable has easy change values in report whit $\mathbf{Z}^{(0)}$.

It is ntroduction the optimal value $z_4^{(0)} = 3$ in the vector $Z^{(0)}$ and it is execution again the optimization program. The optimal solution $Z^{(2)}$ prezent the change in report whit $Z^{(0)}$ for $z_2^{(2)} = 1.17$ and $z_9^{(2)} = 0.13$ variables. It is iterated the optimization operation introducing in the initial solution $Z^{(0)}$ the values $z_4^{(0)} = 3$ and $z_9^{(2)} = 0.1$. It is observed that the optimal solution is stabilize for the 1, 2, 4, 5, 6, 7, 8, and 10 variables. It is introduced in the initial solution $Z^{(0)}$, the $z_9^{(0)} = 0.4$ value, and the stabilization of the optimal solution was obtained for 1, 3, 4, 5, 6, 7, 8, 9 and 10 variables. The last variant presented in table 8, it start up by initial solution $Z^{(0)}$ and the changes $z_4^{(0)} = 3, z_7^{(0)} = -0.3$ si $z_9^{(0)} = 0.4$.

Taking in account the stability of the induced solution from the minimization of the objectiv function (29), the authors consider that the variant four of the optimal solution preseted in table 8, can be considered the optimal solution. The table 9 contains the optimal solution of optimization problem and the correspondent of the variables associated to the objectiv function (29) whit kinetic model parameters.

Because the optimization algoritm is dedicated to the minimization of the multivariable objective functions without restrictions, it is neccery a verification of the thermodynamic consistence of the optimal solution. This consist in calculation of the rate constants for each raw material and to check up the condition

$$k_0^0 > k_1^0 > 0. (30)$$

The calculation executed for each data experimantal set using the optimal value of the kinetic parameters $l_{00}, l_{01}, l_{10}, l_{11}$, it was confirmed the values of the constants rates reactions at 482°C that fulfil the resctriction function (30).

TABLE 8. Optimal solution of the objectiv function (27)

2		Optimal solution for							
Variable number	Initial solution	Initial solution Z⁽⁰⁾	Initial solution $Z^{(0)}$; $z_4^{(0)} = 3$	Initial solution $Z^{(0)}$; $z_4^{(0)} = 3$; $z_9^{(0)} = 0.1$	Initial solution $Z^{(0)};$ $z_4^{(0)} = 3;$ $z_9^{(0)} = 0.4$	Initial solution $Z^{(0)}$; $z_4^{(0)} = 3$; $z_7^{(0)} = -0.3$; $z_9^{(0)} = 0.4$			
1	1	1	3.55	3.55	3.55	3.55			
2	-0.1	-0.1	1.17	1.17	0.53	1.17			
3	1.8	1.8	2.43	1.8	2.11	2.11			
4	1.0	2.27	3.1	3.13	3.12	3.04			
5	-0.8	-0.73	-0.78	-0.79	-0.76	-0.79			
6	-0.8	-0.79	-0.8	-0.8	-0.8	-0.8			
7	-0.4	-0.4	-0.36	-0.32	-0.33	-0.27			
8	3.2	3.2	3.2	3.2	3.2	3.2			
9	-0.5	-0.5	0.13	0.41	0.47	0.71			
10	3.0	3.039	3.08	3.1	3.12	3.07			
	al objectiv ction value	38.45	20.28	19.95	20.00	20.38			
Optir func	nal objectiv ction value	11.90	15.02	14.8	14.57	15.02			
	umber of tion function	75	197	141	194	162			

TABLE 9. Kinetical optimal values

Variable z_i	Kinetical parameter	Value
1	<i>a</i> ₀₂	3.55
2	<i>a</i> ₁₂	1.17
3	a ₂₂	2.11
4	<i>a</i> ₀₃	3.04
5	<i>a</i> ₁₃	-0.79
6	a ₂₃	-0.8
7	l ₀₀	-0.27
8	l ₁₀	3.2
9	<i>l</i> ₀₁	0.71
10	l ₁₁	3.07

Therefore, for the catalytic cracking unit from which derive the experimantal data presented in table 5, the kinetic model have the particular form

$$k_0^0 = e^{-0.27 \star ln \left(\frac{C_A}{C_N}\right) + 3.2}; \qquad (31)$$

$$k_1^0 = e^{(C_N)^{+1.07}}$$
(32)

Using the values from the table 9, the approximation functions (27) associated to the catalyts used in the catalytic cracking unit have the relations:

$$f_2 = 3.55 + 1.17 * MAT + 2.11 * MAT^2;$$
(33)

$$f_3 = 3.04 - 0.79 * MAT - 0.8 * MAT^2.$$
(34)

5. Conclusion

The elaboration of the control system for the catalytic cracking unit require to cover the modelling and numerical simulation step. The authors have elaborating a matemathical model and simulation program of the catalytic cracking process. The simulation process involve the adaptation of the process model. The reseach realized by authors have identified four componets associated the adaptation operation of the catalytic cracking process modeles.

Reactor constructive data. From available industrial data was determined the constructive elementes of the reactor, that is neccery for the mathematical model. The phisical properties of the raw material. The raw material is characterized by some measurable phisical properties in laborator. The authors have identified and tested an empirical relation set which permited the estimation of the phisical properties, that is necceray for the mathematical model of the catalytic craking reactor.

The methodology of the kinetic model's parameters adaptation. The model of the catalytic cracking is characterized by a big relativ number of kinetic parameters. If the approximation function structure of this parameters is known in general, the coefficients values form the functions structure depend directly by phisical properties of the raw material and the catalyst.

The central point of this paper is represented by the special calcul methodology dedicat the adaptation of the kinetic parameter model. It was elaborated an multivariable objective function, used for tuning the mathematical model at experimentale date of the unit.

For the minimzation of the objective function was selected the cyclicial exploration algorithm with contant step. It was elaborated a complex program for the minimization of the objective function.

For numerical determinated of the coefficients values associed the approximation function of the kinetic parameters, the authors was testing the simulation program for different initial solution. The optimal solution was determinated base on independent criterion in raport with initial solution. Because the optimization algorithm is fated the multivariable function without restrictions and the kinetic model involve the restriction by function type, was assesses the resctriction functions for all experimental date set.

The results obtain allowed the kinetic parameter validation and implicit the model of the catalytic cracking model for the typical experimental date of the unit.

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