NUMERICAL MODELLING OF A FCC REGENERATOR

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Abstract. One of the most import oil refining process is the Fluid Catalytic Cracking. This process consists in the conversion of a gasoil stream, through its vaporisation inside a vertical tube (the riser), into dearer products. Gasoline, liquefied petroleum gas, nafta, and other hydrocarbons fractions are the result of the gasoil catalytic cracking process. Concurrently to the cracking, a layer of coke grows over the catalyst surface and the catalyst deactivates. The spent catalyst is thus sent to the regenerator where occurs the burning of the coke. After that, the regenerated catalyst is sent back to the riser and the craking begins again. This work aims the numerical modeling and analysis of a FCCU (Fluid Catalytic Cracking Unit) regenerator. In the modeling, the regeneration stages are divided into two regions: dilute and dense. The latter comprises a bubble and an emulsion phase. The dense region, both the bubble and the emulsion phases, is model as a CSTR (Continuous Stirred Tank Reactor) in which ordinary differential equations represent conservation equations of mass, energy and species. In the dilute region, the conservation principles are written as partial differential equations to describe the change in space and time of the variables. The partial differential equations of the dilute region are discretized in space. The resulting set of ordinary differential equations (ODE) together with the ODE of the dense region are solved by the 4th order Runge-Kutta method. The results of a case study show the transient behaviour of some cracking parameters such as, temperature, gas and coke concentration. This model is adequate for control purposes.

***Keywords**. Regenerator, coke combustion , catalyst, mathematical modeling*

1. Introduction

A Fluid Catalytic Cracking Unit (FCCU) receives residues from different parts of a refinery (atmospheric and vacuum distillation units). The purpose of a FCCU is the cracking of heavy molecules of the residues. This is an endothermic process and takes place in a vertical tube reactor with ascending flow (riser). The feedstock charge gets in contact with a catalyst at a high temperature (500 to 700^{o}C) and it is cracked in dearer products, such as gasoline and LPG. After that, the products are separated from the catalyst to stop the cracking and to avoid loss of performance of the process. The reactions also produce, light gasoil, fuel gas and coke that is deposited on the surface of the catalyst. The coke deposition reduces the available area for the reactants and the catalyst is deactivated. In order to recover its activity, the catalyst is sent to a regenerator. Air is injected in the regenerator and forms a non-homogeneous fluidized bed with the catalyst. The coke on the catalyst surface is thus burnt in this bed. The burning of the coke has two essential functions: a-) the regeneration of the catalyst, restoring its catalytic activity and, b-) the increase of the catalyst temperature to provide energy to the cracking when it returns to the riser.

The economic relevance of the fluidized catalytic cracking is the main reason to study this process. Small gains on the performance of the process may signify savings of millions of dollars. The optimization of the performance requires a deep knowledge of the process and modeling helps on the problem understanding.

Several works have been conducted in order to model the regeneration process. Han and Chung (2001) developed a dynamic model of a FCC in which the regenerator was divided in two regions. Santos (2000) presented different models for the regenerator. Ali and Rohani (1997) published a dynamic model of a FCC converter and later, it was modified by Malay et al. (1999). Based on the ESSO's model IV, McFarlane et al. (1990) developed a simulation model of the converter and its auxiliary equipment. Lansarin (1997) developed a mathematical model to simulate the steady state behavior of a UOP Stacked converter.

Other authors published more specific studies of the regenerator. Among others, Guigon and Large's (1994) work applies a two-phase model to a various stage regenerator. Lee et al. (1987) developed three different models for a typical regenerator and compared the models with experimental results of an industrial plant. Maciel's et al. (1996) model explored the influence of the air jet at the feed grill. A dynamic modeling of fluidized bed with a rigorous hydrodynamics description of Geldart's A and B type particles is presented by Falsi-Saravalou and Vasalos (1991). The authors describe the dense bed as two phases: bubble and emulsion. The dilute bed above the dense one is modeled as one-dimensional flow reactor.

Based on previous works, the current paper presents a dynamic model to simulate the regeneration process. This model intends to be applied to control algorithm. The results of a case study show the model potential.

2. Mathematics Modeling

The current model separates the regenerator in two distinct regions: dense and dilute. The dense region is composed of two phases: bubble and emulsion, as shown in Fig. 1. According to Lansarin (1997), this hypothesis is acceptable for simulation and control purposes. The model has the following characteristics: i-) the bed is on the minimum fluidization condition for solids in the emulsion phase; ii-) the bubble-phase does not contain solids; iii-) heat and mass are exchanged between bubble and emulsion phases; iv-) the major part of the combustion takes place in the emulsion phase; v-) the effluent gas of the regenerator consists of CO, CO_2, O_2, H_2O, N_2; vi-) the burning of hydrogen is considered instantaneous; vii-) the hydrogen-carbon ratio is constant during the coke burning; viii-) the cyclones have an efficiency of 100%; ix-) the specific heat of the catalyst is assumed constant for the considered temperature range; x-) the specific heat of the gases is a function of the temperature; xi-) the temperature is considered uniform through the dense phase and a function of the height in the dilute phase; xii-) solid and gas are in thermal equilibrium within the emulsion phase.

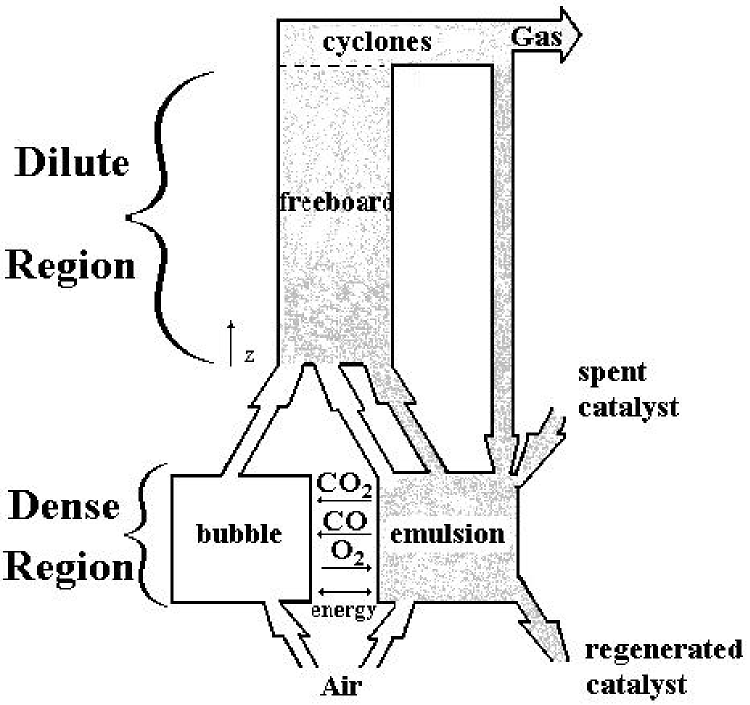


Figure 1. Schematic diagram of the regenerator model.

2.1 Dense region

The dense region comprises: i-) the emulsion phase that is composed of gases and catalyst in the minimum fluidized condition and; ii-) the bubble phase, that is constituted by the air amount that exceeds the minimum fluidization.

2.1.1 Emulsion Phase

This phase is considered as a continuous stirred tank reactor (CSTR), composed of solids (catalyst and coke), air (O_2, N_2) and gases formed in the combustion (CO, CO_2, H_2O). The void fraction of the minimum fluidized bed was determined by a correlation similar to that employed by Santos (2000). The dimensionless Ergun´s equation, as presented by Levenspiel (1996), is used to compute the minimum fluidization speed.

Conservation of solids: the mass balance applied to the catalyst in the dense region considers the streams shown in Fig. 2. The storage of the catalyst in the emulsion phase is written as:

$$\frac{dm_{c,E}}{dt} = \dot{m}_{scLCVm} + \dot{m}_{c,CL} - \dot{m}_{TCV} - \dot{m}_{c,F} \tag{1}$$

where $\dot{m}_{c,E}$ is the mass of the catalyst in the emulsion phase, $\dot{m}_{scLCVm}$ is the flow rate of the spent catalyst, $\dot{m}_{TCV}$ is the flow rate of the regenerated catalyst, $\dot{m}_{c,CL}$ is the catalyst flow rate that returns to the dense region from cyclones and $\dot{m}_{c,F}$ is the catalyst flow rate discharged to the dilute region. A mass balance of the coke can be written as:

$$m_{c,E}\frac{dw_{cq,E}}{dt}=(w_{cq,LCV}-w_{cq,E})\dot{m}_{scLCVm}+(w_{cq,E}-w_{cq,CL})\dot{m}_{c,CL}+\dot{R}_{cq,E} \tag{2}$$

where the terms $w_{cq,E}$, $w_{cq,LCV}$ and $w_{cq,CL}$ indicate, respectively, the coke on the catalyst within the emulsion phase, in the spent catalyst and in the circulating catalyst (from the cyclones). $\dot{R}_{cq,E}$ is the burning rate of coke in the emulsion phase.

Mass conservation of gases: the modeling of the emulsion phase as a CSTR (Continuos Stirred Tank Reactor) provides similar balances to the gas species. The conservation of each gas component can be written as:

$$\frac{dC_{i,E}}{dt}=\frac{v_{g,E}}{L_E}(C_{i0,E}-C_{i,E})+D_{BE}(C_{i,B}-C_{i,E})+\frac{\dot{R}_{i,E}}{\mathring{a}_{mf}} \tag{3}$$

where $C_{i,E}$ is the *i* (= CO, O_2, CO_2, H_2O, N_2) component concentration within in the emulsion phase. $C_{i0,E}$ is the *i* component concentration at the inlet of the air, $C_{i,B}$ is the *i* component concentration in the bubble phase, $v_{g,E}$ is the gas speed in the phase, D_{BE} is the mass diffusion coefficient between the bubble and emulsion phases, $\dot{R}_{i,E}$ is the rate of consumption or generation of each *i* component, L_E is the dense region length.

Energy conservation: the application of the energy balance to the emulsion phase can be express as:

$$\left[m_{c,E}c_{pc}+m_{g,E}c_{pg,E}\right]\frac{dT_E}{dt}=\dot{m}_{ar,E}\Delta h_{ar,E}+\dot{m}_{LCV}c_{pc}(T_{LCV}-T_E)+\dot{m}_{c,CL}c_{pc}(T_{CL}-T_E)+\Delta\dot{Q}_{reac,E}-\Delta\dot{Q}_{BE}-\Delta\dot{Q}_{los,E} \tag{4}$$

where T_E is the emulsion phase temperature, T_{LCV} is the spent catalyst temperature, T_{CL} is the temperature of the catalyst that returns from the cyclones, c_{pc} is the catalyst specific heat, m_{gE} is the mass of the gas in the emulsion phase, $c_{pg,E}$, is the specific heat of the gases in the emulsion phase, Δh_{ar} is the enthalpy of the air at the inlet. $\Delta\dot{Q}_{reac,E}$ is the energy released by the reactions. $\Delta\dot{Q}_{BE}$ refers to the heat exchanged with the bubble phase, and $\Delta\dot{Q}_{los,E}$ is the heat lost to the ambient.

2.1.2 Bubble Phase

The bubble phase is composed of only gases. The oxidation of CO into CO_2 is considered to take place in the bubble phase. The transient mass balance of each species is given by:

$$\frac{dC_{i,B}}{dt}=\frac{v_{g,B}}{L_E}(C_{i0,B}-C_{i,B})\ \text{-}D_{BE}(C_{i,B}-C_{i,E})+\dot{R}_{i,B} \tag{5}$$

where $C_{i,B}$ indicates the *i* component (CO, O_2, CO_2, H_2O, N_2) concentration in the bubble phase, $C_{i0,B}$ is the *i* component concentration of the at inlet of the air and $v_{g,B}$ is the speed of the bubbles. $\dot{R}_{i,B}$ accounts for the oxidation of CO into CO_2. The energy conservation can be written as:

$$m_{g,B}\frac{dT_B}{dt}=\dot{m}_{ar,B}c_{p,ar}(T_{ar}-T_B)+\Delta\dot{Q}_{reac,B}+\Delta\dot{Q}_{BE} \tag{6}$$

where T_B is the bubble phase temperature, T_{ar} is the temperature of the air that enters the bubble phase, $c_{p,ar}$ is the specific heat of the air at the inlet, $\Delta\dot{Q}_{reac,B}$ is the energy released by the reactions.

2.2. Dilute Region

The dilute region is modeled as an one-dimensional flow reactor. The variation of each component concentration and of the temperature of the dilute region are written as a function of the height. The model predicts the amout of coke on the catalyst surface which is dragged to the cyclones and also the oxidation of CO into CO_2 in the gaseous phase. The amount of catalyst in the dilute region decays exponentially with height and its fraction of volume is calculated by the Kunii and Levenspiel's (1996) empirical relation.

Conservation of mass: The coke conservation equation takes into account changes with height. The storate of coke in the dilute region is given by:

$$\frac{\partial(w_{cq,F})}{\partial t}+\frac{v_{cF}}{e_{cF}}\frac{\partial(w_{cq,F})}{\partial z}=\dot{R}_{cq,F} \tag{7}$$

where $w_{cq,F}$ is the coke fraction of the catalyst in the dilute region, e_{cF} is the solid fraction (catalyst and coke) of dilute region, v_{cF} is the speed of the catalyst and $\dot{R}_{cq,F}$, the rate of coke consumption. The conservation of the gas species assumes the form:

$$\frac{\partial(C_{i,F})}{\partial t}+\frac{v_{g,F}}{e_{gF}}\frac{\partial(C_{i,F})}{\partial z}+\frac{C_{i,F}}{e_{gF}}\frac{\partial(v_{g,F})}{\partial z}=\dot{R}_{i,F} \tag{8}$$

where $C_{i,F}$ is the concentration of the component i (O_2, CO, CO_2, H_2O, N_2) in the dilute region, $v_{g,F}$ is the speed of the gas, e_{gF} is the void fraction of the dilute region and $\dot{R}_{i,F}$ is the rate of consumption or formation of the i component.

Conservation of energy: The energy conservation equation in partial differential form is:

$$(r_c c_{pc} e_{c,F}+r_{g,F} c_{pg,F} e_{g,F})\frac{\partial T_F}{\partial t}=-(v_{c,F} r_c c_{pc} e_{c,F}+v_{g,F} r_{g,F} c_{pg,F} e_{g,F})\frac{\partial T_F}{\partial z}+\dot{Q}_{reac,F}+\dot{Q}_{los,F} \tag{9}$$

where T_F is the temperature of the dilute phase, $r_{g,F}$ is the specific mass of the gases, $c_{pg,F}$ is the specific heat of the gases, $\Delta\dot{Q}_{reac,F}$ is the energy released by the reactions, $\Delta\dot{Q}_{los,F}$ is the energy lost to the ambient.

2.3. Overall Mass Balance

The total mass of catalyst in the regenerator is the sum of the dense and dilute region mass:

$$m_{c,RG}=m_{c,E}+m_{c,F} \tag{10}$$

where $m_{c,RG}$ is the mass within the regenerator, $m_{c,F}$ is the mass of catalyst in the dilute region.

The rate of change of the mass of gases in the regenerator is given as a function of the inlet air flow, of the amount of gases formed during the burning of coke burning and of the exhaust gas flow:

$$\frac{dm_{g\,rg}}{dt}=\dot{m}_{ar}-\dot{m}_{g,CL}+R_{i,j} \tag{11}$$

2.4 Pressure in the Regenerator

The pressure in the regenerator is calculated by the ideal gas law:

$$P_{rg}=\frac{r_{g,rg}RT_E}{M_{g,rg}}Z_g \tag{12}$$

where $r_{g,rg}$ is the specific mass of the gases, obtained from the overall mass balance in the regenerator, $M_{g,rg}$ is the molecular weight of the gases in the regenerator, and, Z_g, the compressibility factor of the gases.

2.5 Equation of the Flowrate Control Valve

The gas flowrate at the regenerator outlet is given as a function of the difference of pressure through the valve:

$$\dot{m}_g=\frac{k_v X_v}{\sqrt{a_v+(1-a_v)X_v^2}}\sqrt{P_{rg}-P_{ex}} \tag{13}$$

where k_V is the constant of the valve, X_v is the opening position of the valve, a_v is the ratio of maximum and minimum positions of the valve and P_{ex} is the pressure of the gases within the regenerator.

2.6. Kinetic Model

Usually, coke is composed of different chemical components (C, H, N, S), but mainly of carbon and hydrogen. Therefore, in this model, coke consists in the hydrocarbon CHx. The kinetic model considers the formed coke has a constant ratio of carbon and hydrogen (x = constant), the reaction of hydrogen is complete and instantaneous, and the combustion of the coke is the slowest in the reaction (bottleneck). The combustion of coke is modeled according to the Arbel's et al. (1996) model that considers two reactions: one forming carbon monoxide (Eq. (13)) and another carbon dioxide (Eq. (14)). Beyond the combustion reactions, oxidation of CO to produce CO_2 (Eq. (15) and (16)) also disputes the available oxygen in the regenerator. The considered reactions are:

$$CH_x + (0.5 + x/4)O_2 \xrightarrow{k_1} CO + x/2\,H_2O \tag{13}$$

$$CH_x + (1 + x/4)O_2 \xrightarrow{k_2} CO_2 + x/2\,H_2O \tag{14}$$

$$CO + 1/2\,O_2 \xrightarrow{k_{3c}} CO_2 \tag{15}$$

$$CO + 1/2\,O_2 \xrightarrow{k_{4h}} CO_2 \tag{16}$$

where x is the ratio of hydrogen and carbon, k_1, k_2 and k_{3c} are, respectively, the constants of the heterogeneous reaction rates and k_{4h} is the constant of the homogeneous reaction rate. In the combustion, the reaction rate is of second order and depends on the oxygen and coke concentrations:

$$r_1 = (1 - e)\, r_c k_1 w_{cq} y_{O_2} P \tag{17}$$

$$r_2 = (1 - e)\, r_c k_2 w_{cq} y_{O_2} P \tag{18}$$

where r_c is the specific mass of the catalyst particle, w_{cq} is the coke fraction of the catalyst, y_{O_2} is the molar fraction of the oxygen and P is the pressure. The constants k_1 and k_2 are defined as:

$$k_1 = \frac{b_c k_c}{b_c + 1} \tag{19}$$

$$k_2 = \frac{k_c}{b_c + 1} \tag{20}$$

where k_c and b_c are, respectively, the overall rate of coke combustion and the CO/CO_2 ratio on the surface of the catalyst (Arbel et al., 1996). The oxidation of CO is dependent on the type of the catalyst (composites of vanadium, nickel and copper contribute to the total combustion) and it can be homogeneous and/or heterogeneous. The rate of CO oxidation (Eq. (15) and (16)) is given by:

$$r_3 = (1 - e)\, r_c k_{3c} y_{O_2} y_{CO} P^2 \tag{21}$$

$$r_4 = e k_{4h} y_{O_2} y_{CO} P^2 \tag{22}$$

where e is the void fraction. All the constants (k_c, k_{3c} and k_{4h}) of reaction depend on the temperature and are calculated by the following model of Arhenius.

3. Method of Solution

In the model presented in the previous section, the dense region is described by ordinary differential equations solved by the 4th order Runge-Kutta's method. The partial differential equations of the dilute regions were discretized along space by employing the finite difference approach and the 4th order Runge-Kutta's method was used to integrate the equations along time. The time-step and the Δz which provides a grid independent solution is in the order of 0,01s and 0,2m, respectively. In this case, each calculation minute corresponds to a 6 minutes simulation in a Pentium III computer.

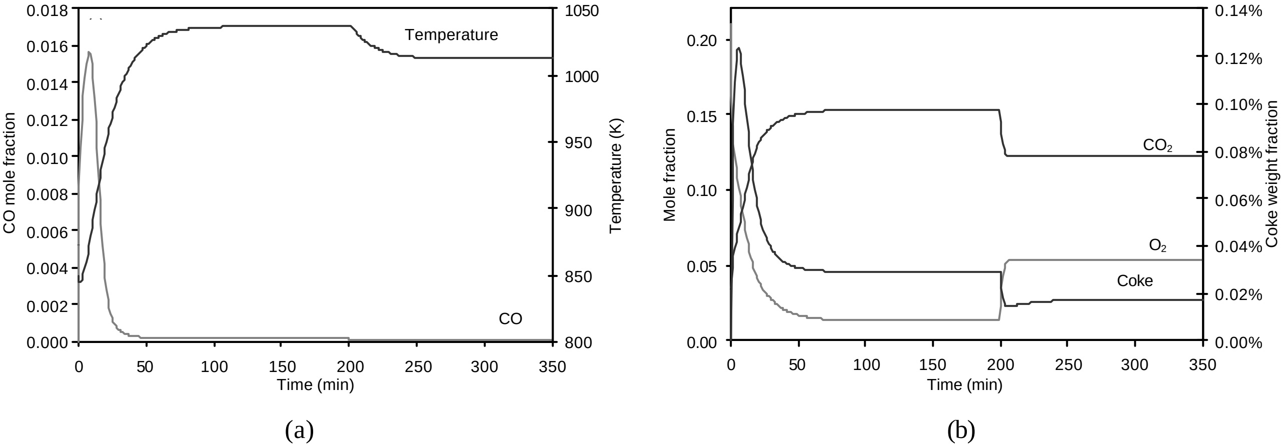
4. Results

The model was used to simulate a regenerator of the FCC pilot plant of SIX[#]. In the simulations, the inlet spent catalyst and air flow rates are considered constant and the regenerated catalyst flow rate is made equal to the inlet catalyst flow rate. The gas flow rate controls the pressure within the regenerator and it depends on the difference of pressure through the valve. A sensibility analysis regarding the inlet air flow rate and the coke fraction in the spent catalyst was conducted. These variables were chosen because of their direct influence on the rates of reactions. The variation of air flow was carried out to observe its influence on the combustion condition (partial or total) of the regenerator and the step change of coke fraction to verify the dependence of the regenerator transient behavior on the step change.

Some simulations were carried out with different air flow rates. For a 0.070 kg/s air flow rate, the transient results are shown in Fig. (2a) and (2b). At this condition, the regenerator operates in the limit between the partial and total combustion. This limit was found out by the steady state analysis of the composition of the gases for different air flows. The numerical stability was tested for severe operating conditions. As shown in Fig. (2a) and (2b), the curves present high gradients before stabilization and this implies the use of small time steps.

Once the catalyst is free of coke in the beginning of the simulation, there is a lack of energy generation and the temperature of the regenerator reduces (Fig. (2a). In this initial period, the amount of coke within the regenerator increases (Fig. 2(b)) and reaches its maximum value (0.123%) in 6 minutes. Then, it reduces to 0.029% after 100 minutes reaching the steady state.

The period that the maximum coke fraction happens coincides with the time of maximum concentration of CO (Fig. (2a)). This is a function of the low temperature of the reactions what favors the partial combustion. In the steady state, the temperature is higher (1036K) and the CO mole fraction falls to a very small value that can be considered zero. On the other hand, the CO_2 level increases with the time (Fig. (2b)) and its molar fraction stabilizes at 0.15. Fig. (2b) shows the concentration of O_2 always decreases. This happens because the constant consumption of oxygen and the temperature elevation that increases the speed of the reaction and favors the complete combustion.



(a) (b)

Figure 2. – Initial transient results and response to a 10% step change of spent catalyst coke fraction after stabilization.
(a) Temperature and CO mole fraction and (b) CO_2 and O_2 mole fractions and coke fraction in the regenerated catalyst.

After 200 minutes of simulation (steady state was reached after 100 minutes), a 10% step change in the spent catalyst coke fraction was established. Fig. (2a) and (2b) show the behavior of some variables after the coke fraction has been reduced to 90% of its initial value. After a new steady state has been reached, the molar fraction reduced 25% of its previous value, the temperature diminished 24K and the concentration of O_2 increased once CO_2 formation has decreased. As can be seen, the temperature fall did not affect the level of CO formation.

In order to evaluate the influence of the inlet air flow on the performance of the model, the simulation was run for 6-hour period for different operating conditions. The results indicate the inlet air flow rate has a direct influence on the

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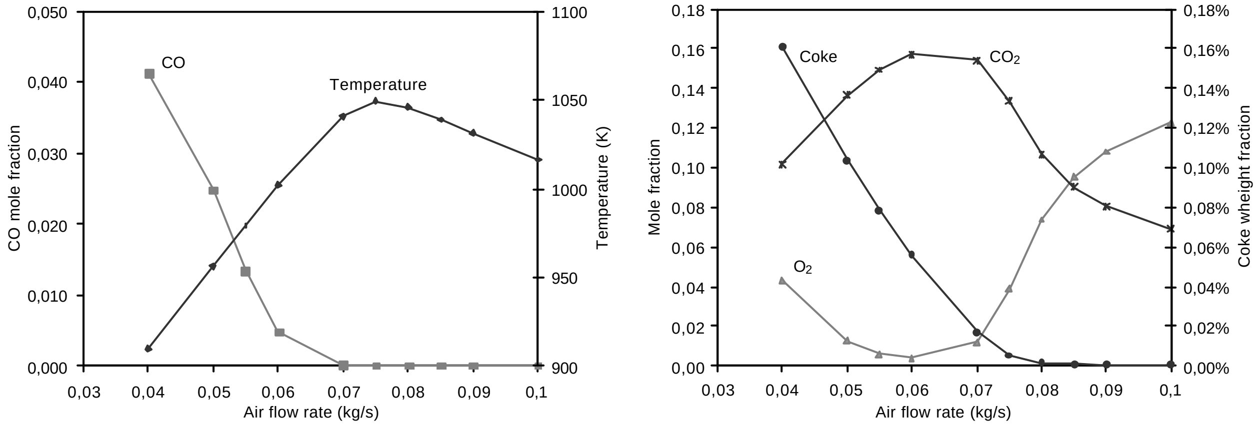
[#] Refinery of PETROBRAS located in São Mateus do Sul – PR, Brazil.

combustion, affecting the temperature, species concentration and coke amount. Figures (3a) and (3b) show the different steady state for distinct values of inlet air flow rates. As seen in Fig. (3a), the temperature increases to a maximum value and the CO concentration reduces to zero as the air flow rate raises. At the maximum temperature, the regenerator operation has reached its total combustion regime. Analyzing the composition of the gases, one can see the transition from partial to total combustion takes place above an air flow rate of 0.07kg/s. After that, the excess of air causes the reduction of temperature diminishing the efficiency of the regeneration.

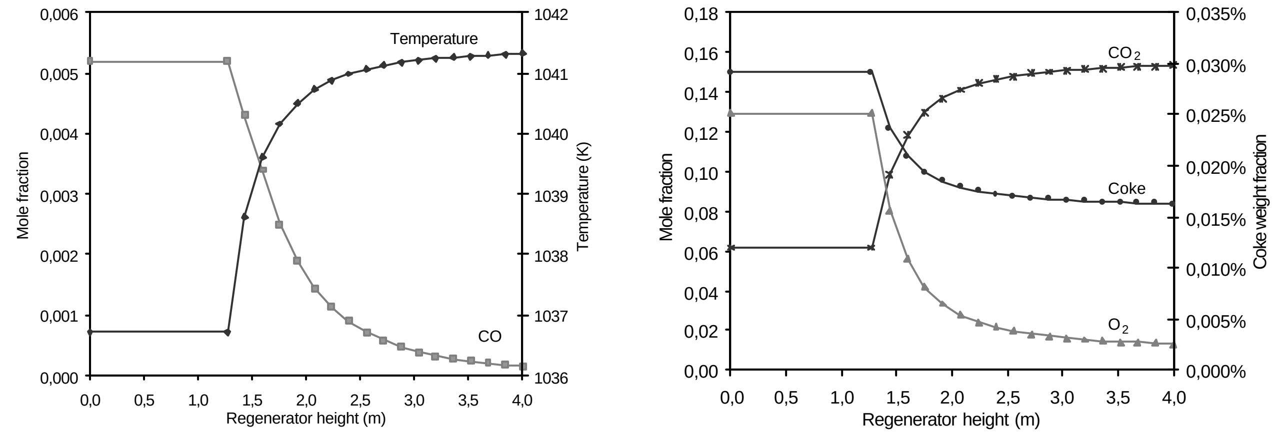


(a) (b)

Figure 3. (a) Effect of the air flow rate on the regenerated catalyst temperature and on the CO mole fraction of gases. (b) Effect of the air flow rate on the O_2 and CO_2 mole fraction and the remaining coke on the regenerated catalyst.

Figures (4a) and (4b) present the temperature and concentration profiles of the dense and dilute phases for the steady state condition (0,070 kg/s air flow rate). The constant temperature and/or concentrations values represent the dense region which is approximately 1,3m high. The dilute region is above it. Composition of gases and coke contents are considerably changed in dense region and the catalyst with a smaller fraction of coke returns to the dense bed reducing the coke fraction of that region.

The dilute region reactions take place because the rupture of the bubbles at the height of the dense region. The oxygen/coke ratio increases and the coke continues to be burnt along the dilute region, raising the temperature to 1041,3K at the top of the regenerator (Fig. (4a)). The temperature increases with the coke combustion and CO oxidation. One can note the mole fraction of O_2 is reduced significantly as coke and CO consume more oxygen to produce CO_2 (Fig. (4b)).



(a) (b)

Figure 4. (a) Steady-state temperature and CO mole fraction profiles in the regenerator. (b) Steady-state mole fraction and coke faction on the catalyst.

5. Conclusions

A FCCU (Fluid Catalytic Cracking Unit) regenerator is modeled in this paper. The approach divides the regenerator into two regions: dilute and dense. The latter comprises a bubble and an emulsion phase. Conservation equations of mass, energy and species are employed to model both dense and dilute regions. The dense region, both the bubble and the emulsion phases, is model as a CSTR (Continuous Stirred Tank Reactor) and the flow is considered one-dimensional in the dilute region. The partial differential equations of the dilute region are discretized in space. The resulting set of ordinary differential equations (ODE) together with the ODE of the dense region are solved by the 4th order Runge-Kutta method. The results of a case study show the transient behaviour of some cracking parameters such as, temperature, and gas and coke concentration.

Some concluding remarks:

- The results show the operation start-up is very sensitive to the initial values of the variables. Considering the plant control system, this should be a matter of concern.
- A step change reduction of 10% in the coke concentration of spent catalyst was performed and the regenerator temperature and the coke concentration in the regenerated catalyst diminish.
- The existence of the dilute region has a great influence on the coke fraction of the dense region because of the returning catalyst flow from the cyclones.
- The regenerator temperature and the CO concentration are very sensitive to air flow rate changes. The complete combustion can be identified when the temperature reaches its maximum value and when the CO concentration is zero.
- The developed model has a great potential for control purpose as the equations are rapidly solved. For each minute of computation, six minutes of simulation is performed in Pentium III computer.
- The results show coherence with models found in the literature.

Other solution methods, such as finite difference, will be investigated in order to reduce the solution time. The solution time is quite important for control purposes.

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