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OPTIMAL OPERATION PROFIT OF A PILOT ROTARY KILN FOR CHARCOAL ACTIVATION

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Abstract— **This work presents an optimization study of a pilot rotary kiln steady state operation, used to manufacture activated carbon (AC) from eucalyptus wood and, a sensitivity analysis on the optimal solution. The main goal is the maximization of a process operating profit function, considering constraints on the optimization variables as: residence time, solid feed, activation steam and heating gas flow rates, for maintaining product quality and maximum production yield. Bounds on optimization variables have been previously defined by a sensitivity analysis of the kiln performance carried out via simulation of the steady state mathematical model. Due to the kind of equality constraints, the optimization problem was solved coupling the optimization code with a differential equation solver in a Matlab^{TM} framework. Results attained allow getting valuable information for design, operation policy, regulation and reactor scale-up in order to develop a local technology for activated carbon production using a regional raw material.**

Keywords– **activated carbon, rotary kiln, optimization, operating profit.**

I. INTRODUCTION

Activated carbon (AC) is an adsorbent material widely used in different industrial and purification processes. The physical activation procedure of charcoal with steam is the core of production plants of activated carbon from cellulose materials. The transformation process is strongly endothermic and involves the gasification reaction of carbon with steam within the particle at temperatures between 1073 and 1373 K (Yehaskel, 1978). The direct-heated rotary kiln is broadly used for physical activation. The pilot rotary kiln is a cylinder that rotates around its longitudinal axis and essentially operates as a heat exchanger (Ortiz *et al.*, 2003a). It has a diameter of 0.30 m with 3.70 m length and is covered with 0.15 m of insulation material. The cylinder is lightly inclined (*i.e.*, slope about 2-6%) to help the axial displacement of the solid bed, which moves towards the discharge end as the hot gases circulate in counter-current mode. The rotational rate is 0.5-2.5 rpm. Figure 1 shows a scheme of the rotary kiln. The solid feed is carbonized matter obtained from a variety of raw materials such as eucalyptus wood.

The hot gases originated by the combustion of natural gas, which arise from a central burner, supply the necessary energy for the activation reaction. Water vapor is injected as the activation agent in co-current mode. Most of the raw materials are relatively pure solids, with moisture contents around 5 to 10%. Usually, the content of impurities in the carbonized solid is negligible. It is supposed that the solid bed moves as a pseudo fluid with axial displacement and without retro - mixing, and it rolls or slides in transverse direction as the cylinder rotates. Table 1 shows the main dimensions and nominal operating conditions of the pilot rotary kiln. The mathematical model and steady state simulation of the kiln operation has been previously reported by Ortiz *et al.* (2003a).

Optimization studies for different kind of rotary kilns have been reported in the open literature. The production yield optimization of a rotary kiln used in the coke calcination process has been carried out by means of simulation of a steady state mathematical model (Bui *et al.*, 1993). Stylianides (1998) has reported a model which allows developing an optimal plan to increase the clinker production capacity. The net present value (NPV) of the cash flow generated in the installation of new kilns has been used as objective function. An optimization model for geometry design of a laboratory-scale rotary kiln pyrolyser of municipal solid wastes (MSW) and the optimum solutions have also been approached by Li *et al.* (2002). In that work, the objective function is the minimal space occupied by the kiln and constraints are imposed by the practical operating variables (rotational speed and kiln slope). The bounds on manipulated variables come from: desired material volumetric flow (MVF), complete pyrolysis time, design of the kiln (exit dam and internal structures), heat transfer and the experimental limitation on the relation length/radius of the kiln.

This single objective constrained nonlinear optimization problem was solved by means of sequential quadratic programming (SQP) in Matlab™ Toolbox (MathWorks, 2002).

In a previous work (Ortiz *et al.*, 2003c), the operation of the pilot rotary kiln under study has been optimized using the production yield as objective function and a simplified model for the constraints which assures the product quality. On the other hand, a complete study on the dynamic behavior has been accomplished by means of Simulink™ – Matlab™ in Ortiz *et al.* (2005).

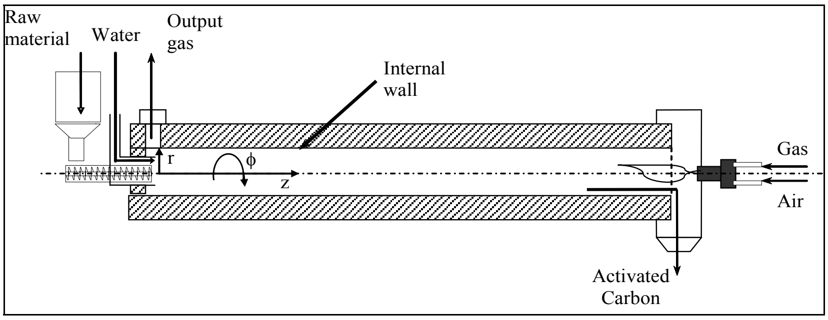

Fig. 1. Pilot rotary kiln scheme

Table 1. Rotary kiln parameters and nominal operation conditions

Length	3.7 m	Reaction temperature	1073 – 1173 K
Internal diameter	0.30 m	Rotary kiln pressure	Atmospheric
External diameter	0.60 m	Rotation speed	0.5 to 2.5 rpm
Raw material	Solid carbonaceous material	Rotary kiln slope	2 to 6 %
Solids flow rate	$\leq 4.5 \cdot 10^{-3}$ kg s^{-1}	Residence time	≤ 7200 s
Moisture content	5 – 10 %	Solid input temperature	Room temperature
Particle size	0.002 m (-6+20) # ASTM	Gas input temperature	≤ 1600 K
Activation gas	Water vapor	Heating	Natural gas combustion
Water vapor flow rate	$\leq 11 \cdot 10^{-3}$ kg s^{-1}	Gas input flow rate	≥ 4 · [solid input flow rate]

This paper aims to present an optimization study of this reactor in order to find the best operational conditions for the maximum operating profit. Such goal has been achieved maintaining a good quality product and maximum production yield. The optimization variables have been previously defined by means of a sensitivity analysis of a pilot rotary kiln performance (Ortiz *et al.*, 2003b). The objective function represents the operating profit, and includes the incomes by sales, and operating costs. The constraints of the problem are constituted by a set of ordinary differential equations (steady state mathematical model), hence, the optimization problem was solved coupling the optimization code with a differential equation solver in Matlab framework. Outcomes attained allow getting valuable information for design, operation policy, regulation and reactor scale-up in order to develop a local technology for activated carbon production using a regional raw material.

The paper is organized as follow:

1) The benefit of carrying out an optimization study is presented and the objective function to be optimized with the corresponding independent variables is analyzed.

2) The equality constraints constituted by the steady state model of the process are exposed.

3) The operational restrictions which define the upper and lower bounds of the optimization problem are discussed.

4) The problem to be solved is formulated and a suitable algorithm for its solution is proposed.

5) The sensitivity of the optimal solution to changes in different parameters of the problem is analyzed.

6) Preliminary conclusions about the optimization study carried out are presented.

II. OPTIMIZATION PROBLEM

The pilot rotary kiln operation previously described is a very complex task because of a number of process constraints must be considered in order to produce an activated carbon with suitable physical properties and, at the same time, to achieve a profitable process.

The chemical reaction carried out in the physical activation process requires an extreme caution on the followings variables: production rate, burn off, and activation temperature. Such variables considerably influence the rotary kiln performance (Ortiz *et al.*, 2003b). The burn off and activation temperature can be correlated with different quality indexes, such as mechanical strength, pore volume, and micro-porous to meso-porous ratio (Tancredi *et al.*, 1996). Laboratory tests have also shown that activation temperature, residence time, flow rate and composition of gas phase, and the presence of alkaline additives determine such a performance (Wigmans, 1989). On the other hand, there is a trade-off between a high production rate with low adsorption area and a low to moderate production rate with high adsorption area. Finally, due to the physical activation in a direct heated rotary kiln is an intensive heat consume process, the total energy of the plant must be correctly integrated in order to obtain a reasonable profitability.

In this first contribution the main goal of the optimization study is to select the main operating conditions in order to achieve the maximum process operating profit producing a commercially acceptable product. Since the plant has not still been integrated energetically, this problem will be addressed in a next work.

A. Objective Function

The sensitivity analysis of the rotary kiln performance carried out by Ortiz *et al.* (2003b) showed that the manipulated variables, which have large influence on the equipment performance, are the following ones: rotational speed of the cylinder (N), solid feed flow rate (Q_{s0}), heating gas flow rate (Q_{gL}) and the water vapor flow rate (Q_v). All this variables can substantially modify the solid temperature profile which is considered as a key feature to obtain a product with good quality. Therefore, an adequate control of the activation temperature (T_a) allows us determining the activated carbon quality. T_a must be maintained between 1073 and 1173 K in order to avoid the charcoal combustion (Wigmans, 1989).

Keeping in mind such operational constraints, a "supervisory control" optimization problem has been formulated. The objective function represents the operating profit, and takes into account the product sales income and the operating costs for an annual operating time of 7500 hours.

Therefore, the objective function has been defined in terms of activated carbon value minus raw material, water vapor and heating gas costs per year as follows:

$$F(x) = [a.x_2 - b.x_1 - c.x_4 - d.x_3]Y.3600 \qquad (1)$$

Where $F(x)$ [U\$S/year] is the Objective Function; x, the vector of variables and,

$x_1 = Q_{s0}$ = raw material mass flow rate [kg/s]

$x_2 = Q_{sL}$ = product mass flow rate [kg/s]

$x_3 = Q_{gL}$ = input heating gas mass flow rate [kg/s]

$x_4 = Q_v$ = input water vapor mass flow rate [kg/s]

The variable x_2 represents the functionality

$$x_2 = f(x_1, x_3, x_4, x_5) \qquad (2)$$

with x_5=N = rotation speed [rpm]

The coefficients in Eq. 1 are: [U\$S/kg],

a = product selling price,

b = raw material cost,

c = heating gas cost

d = water vapor cost

Y = Number of operating hours per year [hr/year]

The unit costs in Eq. (1) have been estimated from the following sources:

Product selling price. An average price of 2.00 U\$S/kg, between granular and powdered activated carbon has been considered (ChemProfiles, 2001).

Raw material cost. This unit cost has been averaged from different suppliers that sell charcoal from eucalyptus wood in the regional market (2003). A price of 0.0832 U\$S/kg has been used.

Heating gas cost. It has been calculated with the utility cost estimation from Ulrich (1984) and updated to 2004 with data from the US Energy Information Administration (2006). Supposing that natural gas is used as fuel and the capital cost is neglected, the unit cost of heating gas is 0.0623 U\$S/kg.

Water vapor cost. From the utility cost estimation in Ulrich (1984), by using coefficients, as the Chemical Engineering Plant Cost Index (2006) and the average unit price for natural gas from the US Energy Information Administration, updated to 2004. For saturated steam of 106 Pa, the unit cost is 0.1402 U\$S/kg.

The solid product mass flow rate Q_{sL}, (variable x_2), is a dependent variable. Its value is obtained from the numerical solution of the differential algebraic equations system (DAE), which constitutes the reactor mathematical model (Ortiz *et al.*, 2003a).

B. Optimization problem statement

Equality constraints: The steady state operation of the pilot rotary kiln has been described by a differential algebraic equations system in Ortiz *et al.* (2003a). The model essentially represents the axial variation of mass flow rate and temperature for the solid and gas phase inside the reactor. In respect of the radial dimension, it is assumed the uniformity of conditions (*i.e.*, flow rates and temperature) based on: the rolling bed operation mode, the average particle size of 2 mm and a gas flow regime in the transition (Ortiz *et al.*, 2005), and also due to the small size of the reactor like is detailed in Ortiz *et al.* (2003a). On the other hand, the steady state model has been validated with the experimental data from a typical endothermic reaction in a similar rotary kiln in Ortiz *et al.* (2003a). Therefore, the equality constrains are constituted by a set of ordinary differential and algebraic equations. In each optimization program iteration the model must be solved by means of a solver using the corresponding boundary conditions. The main equations of the model are constituted by the energy and mass balances for the two phases:

$$\frac{dQ_s}{dz} = f_1(x, \mathrm{P}_1) \qquad \frac{dQ_g}{dz} = f_2(x, \mathrm{P}_2) \qquad (3)$$
$$\frac{dT_s}{dz} = f_3(x, \mathrm{P}_3) \qquad \frac{dT_g}{dz} = f_4(x, \mathrm{P}_4)$$

The boundary conditions are:

$$\begin{aligned} Q_S\ (z=0) &= Q_{S0} \\ Q_g\ (z=L) &= Q_{gL} \\ T_S(z=0) &= T_{S0} \\ T_g\ (z=L) &= T_{gL} \end{aligned} \qquad (4)$$

The model is completed with relations for the solid moisture, wall temperature, kiln geometry and the corresponding correlations for the heat transfer coefficients. P_k, k = 1, …,4, represents the model parameters. A complete model description can be seen in Ortiz *et al.* (2003a)

Inequality constraints: From a previous study (Ortiz *et al.*, 2003b) on the pilot rotary kiln performance, different ranges for varying the manipulated variables have been determined. Such calculation was carried out by changing one manipulated variable and keeping the others constant. The mentioned ranges were established in accordance with the possible values for activation temperature, which is the critical operating variable. As result of such study, upper and lower bounds on the manipulated variables were set in order to assure that the specified activation temperature level to be constant.

The variable N, kiln rotation speed, influences inversely on the solid bed residence time (Perry and Green, 1984). Therefore, the operating limits on N have been obtained analyzing the incidence of residence time on the rotary kiln performance (Ortiz *et al*., 2003b).

Finally, a constraint on the maximum production yield (η) has been imposed. In fact, the output solid mass flow rate, Q_{sL}, is subject to the equality constraint of the reactor model, and to the limits imposed to the production yield in order to assure a commercially acceptable product quality. With regard to η, Encinar *et al.* (2001) has reported that to achieve a good size and distribution pore in the activated carbon; the production yield must not be greater than 65–70 %. Of course, such values depend on the commercial product type. From a previous optimization work using a simplified model (Ortiz *et al.*, 2003c), the upper bound for η has been determined. The lower bound for η has been imposed from bibliography data (Rodríguez Reinoso *et al.*, 1995) taking in account the used raw material. The variation range for the input mass flow rate Q_{s0} has been obtained from the curve that relates the average activation temperature with Q_{s0}.

The mathematical definition for η is shown below and a complete formulation of the optimization problem is presented in Table 2.

$$\eta = [Q_{sL} / Q_{s0}] \cdot 100 = \text{Production yield [\%]} \qquad (5)$$

C. Problem feature and solution method

The optimization problem presented in Table 2 is a Multi-variable Non Linear Programming with equality constrains constituted by a set of differential algebraic equations and feasible operating ranges for the main manipulated variables, as inequality constrains. A similar problem has been studied by Murase *et al*., (1970) for the optimal thermal design of an auto-thermal ammonia synthesis reactor.

Due to the set of differential algebraic equations must be solved numerically to update the objective function value in each iteration search, a two-stage procedure showed in Fig. 2 has been adopted (Edgar and Himmelblau, 1988).

The two-stage procedure has been implemented in Matlab^{TM}, using the DAE solver previously developed by Ortiz *et al.* (2003a) and an optimization code based on the solver "fmincon", which finds the minimum of a constrained nonlinear multivariable function and uses a Sequential Quadratic Programming (SQP) method.

The algorithm works as follow: with an initial guess for x, the optimization code evaluate the objective function and then send the data to the DAE solver, which use the boundary conditions to integrate numerically the DAE system. The output variables are returned to the optimization code in order to update the objective function value and determine if an optimal point has been achieved. Basically, the DAE solver allows calculating the solid mass flow rate (Q_{sL}) at the output end of the rotary kiln by means of solving the coupled gas phase mass and energy balances in the kiln (Ortiz *et al.*, 2003a.)

The optimum operating conditions for the maximum production yield obtained by Ortiz *et al.* (2003c) have been used as an initial guess for x. Table 3 shows the solution of problem with the corresponding value for the maximum operating profit and the optimal operating conditions with its lower and upper bounds.

Table 2. Optimization problem.

Objective Function:	$\min F(x) = -[2.0\,Q_{sL} - 0.0832\,Q_{s0} - 0.1402\,Q_v - 0.0623\,Q_{gL}]\,Y\,.\,3600$
Constraints:	
Steady state model (Ortiz *et al.*, 2003a)	Boundary conditions
$\frac{dQ_s}{dz} = f_1(x, P_1)$ $\quad \frac{dQ_g}{dz} = f_2(x, P_2)$ $\frac{dT_s}{dz} = f_3(x, P_3)$ $\quad \frac{dT_g}{dz} = f_4(x, P_4)$	$Q_{s\,(z=0)} = Q_{s0}$ $Q_{g\,(z=L)} = Q_{gL}$ $T_{s\,(z=0)} = T_{s0}$ $T_{g\,(z=L)} = T_{gL}$
Operation constraints	
$0.66 < N < 1.172$	$0.01714 < Q_{gL} < 0.019$
$0.0030 < Q_{s0} < 0.0038$	$45.00 < \eta < 65.86$
$0.011 < Q_v < 0.020$	

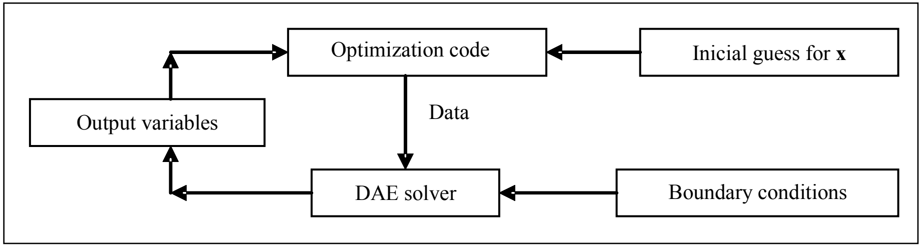

Fig. 2. Two-stage procedure to solve the optimization problem

D. Results and discussion

Both, the objective function and constraints are continuous functions and, the relationship between the production yield and the manipulated variables is:

$$\begin{aligned}\eta = {} & 24.71\times N^{-2} - 52.02\times N^{-1} - 4.10^{5}\times Q_{s0}^{2} \\ & + 36736\times Q_{s0} \times 45232\times Q_{v}^{2} - 2393.4\times Q_{v} \\ & + 55913\times Q_{gL}^{2} - 3920.7\times Q_{gL} + 73.8\end{aligned} \quad . \quad (6)$$

Equation (6) has been achieved by fitting and averaging the curves of η vs. N, Q_v, Q_{gL} and Q_{s0} respectively by using a quadratic regression. The before mentioned curves were obtained by simulation with the steady state mathematical model in Ortiz *et al.* (2003b).

The optimal values of the manipulated variables, which maximize the operation profit, are shown in Table 3; besides another initialization points (x_0) has been tested. The obtained results with different combinations for x_0 have produced optimal profits higher than the value depicted in Table 3, but with production yields higher than the upper bound. Furthermore, for this case the activation temperature profile presents values near to its lower bound. Such operation mode can produce a low quality activated carbon with reduced adsorbent properties.

Keeping in mind the high non-linear form and the difficulty of the optimization problem, a quasi-optimal solution, which assures the maximum production yield with acceptable quality, has been adopted. On the other hand, such a solution holds an actual physical sense for the pilot plant. This solution has been verified by means of simulation in Matlab^{TM}, solving the DAE system with the “ode15s” solver.

From Table 3, it can be concluded that for an optimal operation the water vapor/solid ratio must be kept on 2.9 and, the heating gas/solid on 4.55. On the other hand, as it can be seen on next section the water vapor flow rate is, certainly, the most sensible of the manipulated variables for the optimal kiln operation. Thus, in order to assure an optimal operating profit and a high quality product, the control on Q_{v0} and Q_{gL} must be tight.

E. Sensitivity analysis

The influence of the main physicochemical parameters in the steady state model and, the economical (costs and price) coefficients on the optimum have been studied. Also, the impact of the optimal operation variables and the economical parameters on the objective function was analyzed.

Physicochemical parameters influence: The model that describes the steady state operation of kiln as a function of variables x and the parameters P_k is represented by Eq. (3). By means of parameters sensitivity analysis, it can be determined that the parameters which variation has the greatest influence on the optimal operating point of objective function are: reaction enthalpy, viscosity and thermal conductivity of heating gas. The solid thermal conductivity and other model parameters (not shown here) do not produce a significant impact on the optimal solution.

The most significant relative impacts on the optimal solution by varying each model parameter are shown in Fig. 3. As it can be seen, the main effects on the optimum take place when the percentage of change in the parameters exceeds 20%, particularly in the negative sense. This means that special attention must be focused to mass flow rates and temperatures inside the kiln and, agree with the necessity of a constant temperature.

Economical coefficients influence: In order to evaluate its influence on F(x), each price and cost coefficient P_i (for i=1,…,4) in the objective function was varied since 10% to 40%, maintaining the others constant.

Figures 4 and 5 show the corresponding impacts on F(x) when the coefficients are increased and decreased respectively. Figure 6 shows the accumulated variation in the cost coefficients versus the variation in the price coefficients. As can be seen from Figs. 4 and 5, the impact ranking on profit is as follow: product price, water vapor cost, heating gas cost and raw material cost. This means that although the product price is the most important economical factor (see Fig. 6), particular attention must be taken with the water vapor cost and heating gas cost.

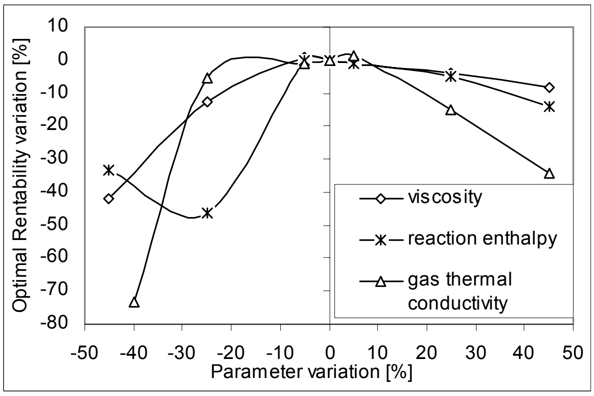

Fig. 3. Relative impact of the main model parameters on the optimal solution point

Table 3. Solution of optimization problem.

	N [rpm]	Qs0 [kg/s]	Qv [kg/s]	QgL [kg/s]	η [%]	Profit [$/year]
Initial point	1.172	0.0038	0.011	0.01714		55796
Solution point	1.172 (tr = 6000s)	0.0038	0.011	0.01731	65.82	
Lower bound	0.66	0.0030	0.011	0.01714	45.00	
Upper bound	1.172	0.0038	0.020	0.019	65.86	

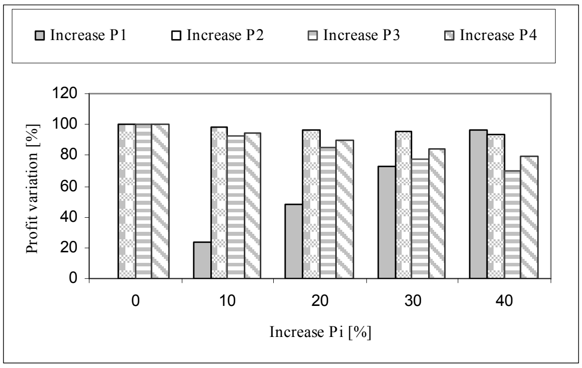

Fig. 4. Impact of economical coefficients on F(x) due to increase in each one

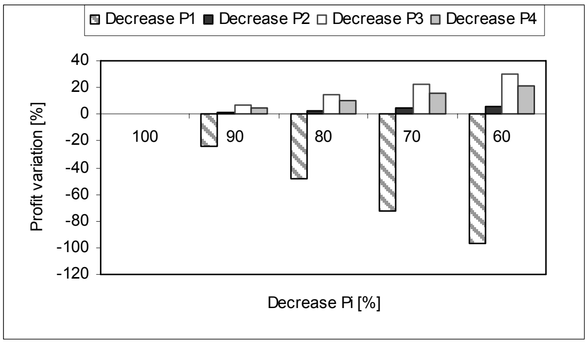

Fig. 5. Impact of economical coefficients on F(x) due to decrease in each one

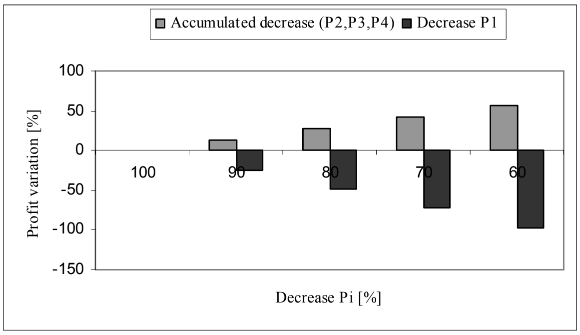

Fig. 6. Impact of economical coefficients on F(x). Accumulated decrease in P2, P3 and P4

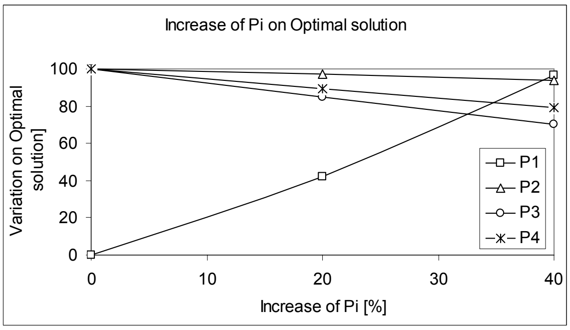

Fig. 7. Impact of economical coefficients on the optimum due to increase in each one

Similar results can be observed in case of Figs. 7 and 8, where it was analyzed the influence of the economic coefficients on the optimum. In that sense, the energetic integration of the plant is very important. In this respect, the energy cost in the plant can be reduced by means of an adequate integration between the activation plant and the pyrolysis plant. Also, the residual heat of the heating gas must be recovered as a heating mean for different plant sections. This topic will be addressed in a next contribution.

Finally, it was evaluated the influence of the variation in the operation variables V_i (for i = 1,…, 4) on the objective function by means of changing the value of each variable between 10 to 40% and keeping the others constant. The results obtained are presented in Fig. 9 and Fig. 10 for the increasing and the decreasing tests respectively. Clearly, the changes in the product flow rate has the main influence on the objective function because of it has the highest price, but it can not be considered in this study, since it is a dependent variable. Hence, the operation variables which have the main influence can be ranked as follows: water vapor, heating gas and raw material flow rates, so much in the positive variation as in the negative. The increase of such variables produce a reduction in the profit, however an increase could produce a lost in the product quality.

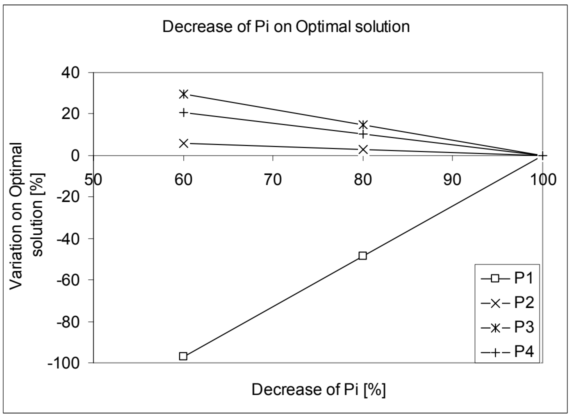

Fig. 8. Impact of economical coefficients on the optimum due to decrease in each one

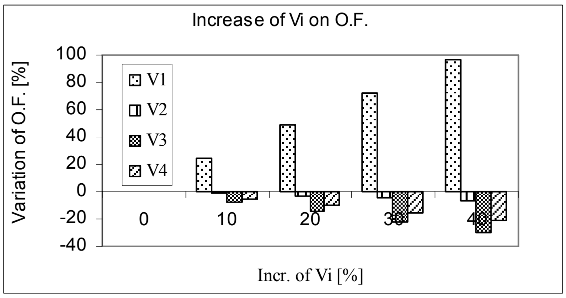

Fig. 9. Influence ranking of the operation variables on the objective function due to increase in each one

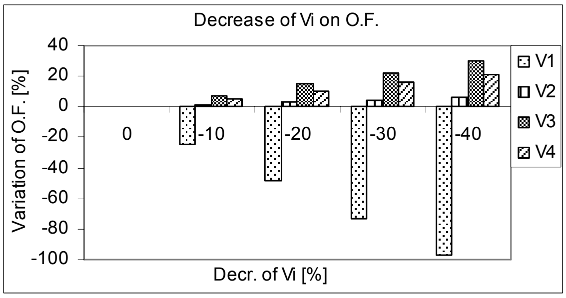

Fig. 10. Ranking of the operation variables influence on the objective function due to decrease in each one

III. CONCLUSIONS

An optimization study of the steady operation of a pilot rotary kiln, used to manufacture activated carbon from eucalyptus wood has been presented. A sensitivity analysis of the optimal solution under the influence of model parameters and manipulated variables was carried out too. Also, the ranking of economical coefficients and manipulated variables in the objective function has been presented. The problem has been approached by means of a coupled solution of an optimization code with a DAE solver in Matlab framework.

Due to the problem complexity, which is highly nonlinear with constraints formed by ODE system, a quasi-optimal solution has been obtained and, a set of optimal operation conditions have been achieved for a maximum operation profit. The optimal operation conditions have been satisfactorily verified by simulation, however to achieve more reliable optimal solutions, additional works about the estimation of model parameters must be done. Thus, model parameters such as, gas viscosity, reaction enthalpy and gas thermal conductivity must be predicted more accurately. In respect of the variables, it can be stated that the product flow rate must be under control, as well as, the minimum vapor/solid and gas/solid ratios at the kiln inlet must be maintained to preserve the optimal operation of the kiln.

The information thus obtained together with the already achieved by means of steady state and dynamic simulation will be a great contribution to enhance the knowledge and insight about the physical activation process. Lately, all this information will be very useful for the scaling up of the pilot plant.

In a next contribution, taking into account that the process is intensive heat consuming and, because of the cost of water vapor and heating gas in the plant are important, a study about the energy integration of the rotary kiln will be addressed. Such a study will consider the energy integration of the activation process so much with that of pyrolysis like the recovery of the residual heat of the heating gas.

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