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CENTRO INTERUNIVERSITARIO DI RICERCA
SUI SISTEMI DISORDINATI E SUI FRATTALI
NELL'INGEGNERIA CHIMICA

Giornate di Studio

Dinamica e Controllo Nonlineare nell'Ingegneria di Processo

DINIP 2000

Roma 28-30 giugno 2000
Università di Roma La Sapienza
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Dinamica e Controllo Nonlineare nell'Ingegneria di Processo, DINIP 2000
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DYNAMICS AND CONTROL OF FORCED-UNSTEADY-STATE CATALYTIC COMBUSTORS

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INTRODUCTION

Catalytic oxidation is a suitable technology for the treatment of waste gases containing low VOC concentrations. Catalytic combustion requires a certain temperature of the catalyst bed, which can be provided by the gas stream itself or by external heating. Large amounts of energy can be required to maintain the reaction temperature, and the heat exchange section can take unreasonable dimensions, especially if the mixture has a very low calorific value. In this case, the use of apparatuses for continuous yet forced non-stationary operation, with a regenerative heat transfer section, appears very promising. Two different designs will be considered in detail in this work: a network of three reactors in series and the reverse-flow reactor developed by Matros.

The first design has been proposed by the authors and its feasibility has been shown in a previous paper (Brinkmann *et al.*, 1999). Each reactor consists of a large inert section for heat exchange and a relatively small catalytically active part near the outlet. A set of valves enables to vary the feed position, thus changing the sequence of reactors. Contrary to the reverse-flow reactor, the flow direction is maintained in this way.

The reverse flow device consists of a catalytic reactor and a set of valves at the inlet and outlet so that the gas flow direction can be reversed; both ends of the reactor act as heat exchangers and may be replaced by inert material.

They represent an interesting example of a reactor (or a sequence of reactors) that cannot operate in the steady state, but for which there exists a successful periodic control policy that switches feed respectively between the two reactor sides or among the reactors. The choice of the best control policy is very important in the treatment of very lean mixtures, in order to avoid reactor extinction or off-range waste emissions, in case of variations of the inlet conditions; for this type of device it is also necssary to develop simultaneously reactor and control design.

A open-loop periodic control with fixed switching periods can give good performances if the inlet conditions are relatively constant. In particular it has been shown (Barresi *et al.*, 1997) that a reverse-flow reactor with the same amount of catalytic and inert material does not perform better than a reactor network operated at fixed cycle time. This is due to wash-out at the beginning of each period, which becomes more important at shorter cycle time. Reducing the amount of inert in order to diminish this effect decreases maximum temperature and may impair the desired autothermal operation at constant catalyst mass. At the same time, the cycle time becomes much shorter than in the reactor network. But this simple logic is unsuitable for the network of reactors; as it is very sensitive to model parameters, is confined to a narrow range and is not stable at variable inlet conditions. The reverse flow reactor is more robust, and the fixed periodic switch can be used in many applications, but not when very low and variable concentrations of VOC have to be treated.

In this work a more robust periodic strategy implemented through feedback control is discussed; the aim is to make this devices reliable for the control of gaseous emissions from iron making processes.

THE MODEL

The model adopted for the simulations is presented below. The PDE system is reduced to a set of ODEs, by discretizing the spatial derivatives on a grid of evenly spaced points. Finally, the ODE system is solved through the Fortran routine "LSODE" included in package "ODEPACK".

The pollutant that has been adopted for the simulations is methane and the catalyst is composed by perovskites.

$$\frac{\partial \tau_S}{\partial s} = \frac{\lambda_S \varepsilon}{c_{p,S} L u_0 \rho_S} \frac{\partial^2 \tau_S}{\partial z^2} + \frac{k_G a L(-\Delta H)\rho_G \varepsilon y_{G,0}}{c_{p,S}(1-\varepsilon) u_0 \rho_S M T_0}(Y_G - Y_S) - \frac{haL\varepsilon}{c_{p,S}(1-\varepsilon)u_0\rho_S}(\tau_S - \tau_G) \tag{1}$$

$$\frac{\partial \tau_G}{\partial s} = -\frac{\rho_{G,0}}{\rho_G}\frac{\partial \tau_G}{\partial z} + \frac{k_{eff}\varepsilon}{c_{p,G} L u_0 \rho_G}\frac{\partial^2 \tau_G}{\partial z^2} + \frac{haL}{c_{p,G} u_0 \rho_G}(\tau_S - \tau_G) - \frac{4Lh_{w,ov}}{D_R c_{p,G} u_0 \rho_G}(\tau_G - \tau_E) \tag{2}$$

$$\frac{\partial Y_G}{\partial s} = -\frac{\rho_{G,0}}{\rho_G}\frac{\partial Y_G}{\partial z} + \frac{D_{eff}\varepsilon}{L u_0}\frac{\partial^2 Y_G}{\partial z^2} - \frac{k_G a L}{u_0}(Y_G - Y_S) \tag{3}$$

$$k_G a (Y_G - Y_S) = r(Y_S, \tau_S) \tag{4}$$

$$r = k_\infty \exp\left({E_a}/{RT_0\tau_S} \right) y_{G,0} Y_S \tag{5}$$

NETWORK OF REACTORS

In this type of device the proper switching period is closely related to the velocity of the heat front: if it is too long, the first section is cooled too much, and the gases do not reach the catalysts at the required temperature, while if it is too short there is no time to accumulate enough heat in the second reactor before switching. The increased average heat loss at unfavorable periods cannot be compensated by reactive heat development, and the system gradually "lights off". The heat front velocity, on the other hand, depends only on the ratio of solid and gas heat capacity and, which is the most important, on the mass flow of gas, if heat transfer is sufficiently fast.

This explains the sensitivity and instability evidenced by the simulations. It is possible to adopt a fixed-time switching policy only if the period can be fixed in advance with an error smaller than a few percent. But if gas velocity is subject to fluctuations, this strategy obviously is inadequate for any kind of reactor configuration. For the reasons stated above, feedback control has been investigated: previous work (Barresi *et al.*, 1999) evidenced that a single-point control strategy

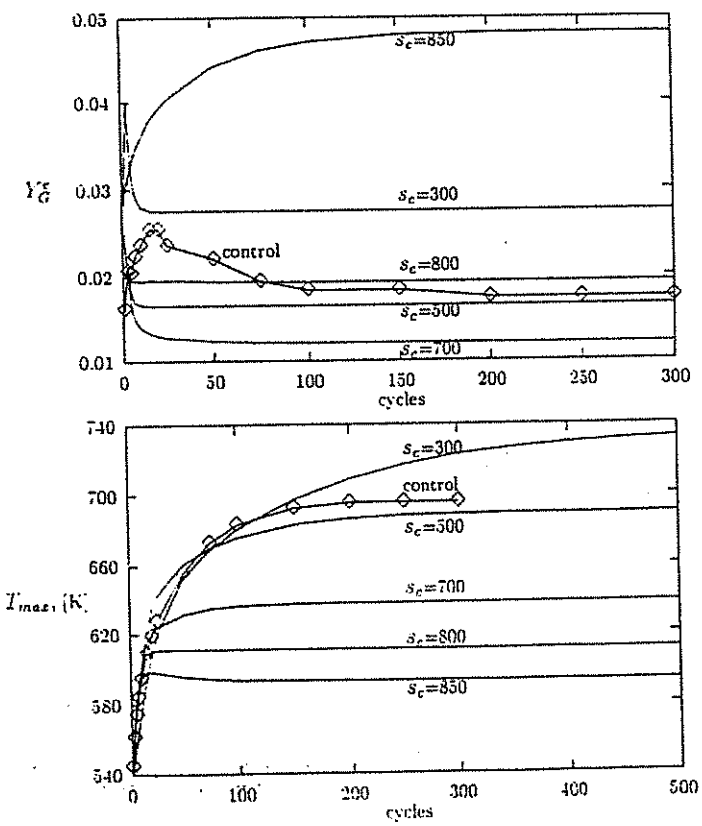

Fig. 1 – Comparison of fixed period and one point control in reverse flow reactor. Ethene, 300 ppm; set point T= 530 K.

(based on the measurement of the temperature at the beginning of the first catalytic bed) is not efficient.

To improve the robustness of the system, an additional temperature measurement located at the beginning of the second catalyst bed has been implemented, and the switch condition has been given as follows: the feed position is changed only if the local temperature in the first reactor drops below set point 1 and in the second reactor is above set point 2.

This strategy, in the following referred to as two-point control, enables stable autothermal operation even at low combustible load, with a reasonable choice of set points.

As the controller in the first section only determines the period length during the first few cycles, its set point has no effect on the final conversion, which is determined by the second one.

REVERSE FLOW REACTOR

In order to improve the performance of the reverse-flow reactor, a one-point control strategy has been considered also for this device, with a temperature measurement located at either end of the active portion. Flow direction is changed when temperature at the controller located close to the inlet drops below a certain setpoint.

The simulations evidenced that at higher setpoint also the maximum temperature is increased, but at least for large portions of inert material conversion cannot be arbitrarily improved by this strategy. This is due to the shorter period at higher setpoint, resulting in a stronger wash-out effect. In addition Figure 1 shows that while maximum temperature is constant soon, it takes much longer time to reach the periodically stable state with respect to conversion as compared to a fixed period.

The conclusion is that a different strategy has to be adopted, as the previous one does not perform better that the simplest one, and does not avoid the risk of extinction in case of low VOC concentration.

As experimental data evidenced that the gaseous emissions from the kind of plants considered may be highly variable in flow rate and pollutant concentration, and the variations can be periodic, the response of the system to this type of disturbances has been investigated (Cittadini *et al.*, 2000).

Two kinds of variations have been taken into consideration: in the inlet concentration and in the flow rate. In both cases, the variations cause relevant fluctuations in the maximum temperature of the bed and in conversion. These fluctuations, especially for long feeding cycle periods, can endanger the correct operation of the reactor. In addition, for particular ratios between the feeding cycle period and the flow-reversal cycle period, the interaction between the two cycles may cause serious problems of instability, represented by large low-frequency fluctuations of the maximum temperature in the case of variations in the inlet concentration, and by a dangerous flow asymmetry in the case of variations in the flow rate. As these may compromise the thermal stability of the reverse-flow reactor, leading the reaction to irreversible extinction, even if in normal conditions it works well, the burner needs a

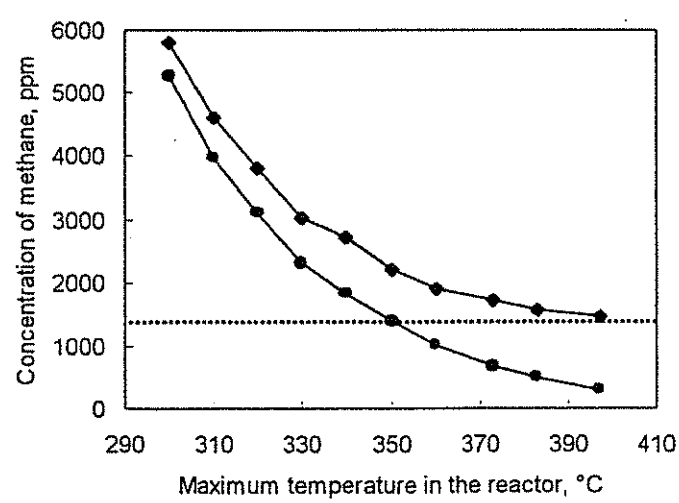

Fig 2. Minimum inlet concentration of methane (♦) for the restoring of the reaction, depending on the maximum temperature in the reactor, and corresponding outlet concentration (•).

control system, that can operate when the reaction is close to extinction. The simplest and better kind of intervention is the addition of auxiliary fuel. The aim of the research is to find a simple and cheap control system that allows low fuel consumption and safe operation.

The control can be based on three parameters: input concentration, output concentration and temperature inside the reactor. According to the results of our simulations, the best control parameter seems to be the temperature inside the reactor. In every single axial point of the bed the temperature changes periodically for the shifting of the hot zone due to the flow reversal, but the shape and the values of the moving profile change very slowly, even in presence of a wide variation in the inlet conditions. The whole profile, for its peculiar shape, can be summarized by two important parameters: the length of the hot zone and the maximum temperature. These two values give accurate information about the development of the reaction: in order to avoid extinction, the latter must be higher than the ignition temperature and the former must be wide enough (depending on T_{max}) to allow the almost complete destruction of the pollutant. The maximum temperature, in particular, gives an accurate indication about the trend of the inlet concentration (c_{in}) in the last minutes of operation: if c_{in} is higher than the minimum concentration that allows autothermal operation, T_{max} raises up to a safe and high value; if c_{in} is lower, T_{max} decreases down to extinction. The length of the hot zone, on the contrary, is determined mainly by the flow-reversal cycle period; if this period is fixed, such a length can not change appreciably and it has only a slight influence on the lower limit for T_{max}, above which extinction is irreversible.

For the measurement of the temperatures inside the reactor, the presence of a few thermocouples in different axial points of the bed has been simulated. Three thermocouples placed in the proper positions (determined by the flow-reversal cycle period) can give a good real-time approximation of T_{max}, while several thermocouples would be needed for a real-time approximation of the length of the hot zone. The recording and the processing of the measurements of only one central thermocouple during the whole flow-reversal cycle can give a good approximation of the temperature profile.

The control system that has been tested is based on the evaluation and the control of only the maximum temperature. The system operates with two set-points for T_{max}: a lower value, below which the introduction of a fixed concentration of auxiliary fuel is activated, and an upper value, above which the feeding of auxiliary fuel is stopped. The concentration of the auxiliary fuel is the minimum one that allows the recovering of the reaction at the lower limit of T_{max}; this last one is chosen high enough to keep low the outlet concentration of pollutant even when T_{max} is close to that limit. The upper limit is chosen high enough to keep low the frequency of switch of the auxiliary-fuel feeding. The simulations have shown that the normal operation can be restored even when the progressive cooling down has led the maximum temperature to very low values (< 300°C), but the lower is the maximum temperature, the higher is the concentration of auxiliary fuel that must be introduced, causing, among other things, heavy discharges of unburned pollutant (Fig. 2). It is evident that better results can be obtained introducing the auxiliary fuel before a deep cooling of the reactor takes place (at 400°C, for instance).

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