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NONSTATIONARY CATALYTIC DESTRUCTION OF LEAN WASTE GASES IN A NETWORK OF BURNERS AND A REVERSE-FLOW REACTOR UNDER NONADIABATIC CONDITIONS

A.A. Barresi, M. Vanni, M. Brinkmann and G. Baldi

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NONSTATIONARY CATALYTIC DESTRUCTION OF LEAN WASTE GASES IN A NETWORK OF BURNERS AND A REVERSE-FLOW REACTOR UNDER NONADIABATIC CONDITIONS

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Abstract - The performances of the two configurations have been considered with respect to influence of radial heat losses and control policy. The network of three reactors reduces the emissions due to wash out of untreated gases, and may be competitive with the reverse flow reactor.

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: a network of three reactors in series and the reverse-flow reactor developed by Matros (1985).

The first design has been proposed by the authors and its feasibility has been shown in a previous paper (Brinkmann *et al.*, 1996). 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 (see Figure 1). 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. Both modelling and applicative aspects of the periodically reversed flow reactor have been widely investigated, even if little data are available on nonadiabatic systems (Boreskov and Matros, 1986; Matros and Bunimovich, 1996).

The model

A heterogeneous one-dimensional mathematical model has been developed. It is assumed that no accumulation of mass occurs at the solid surface, where the combustible reacts according to a first order law; pressure loss inside the reactor is neglected, and heat capacities of gas and solid are assumed to be constant. The details for the adiabatic case have been given in a previous work (Brinkmann *et al.*, 1996); non-adiabacity in the reactor model is accounted for including a term for heat loss to the environment in the gas-phase energy balance.

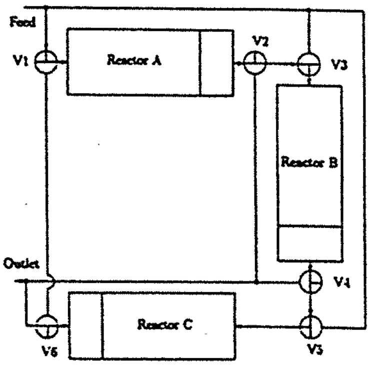

Fig. 1 - Scheme of the network of reactors.

The balance equations for the catalytically active sections thus write:

$$\rho\frac{\partial T_G}{\partial t}=-v\rho\frac{\partial T_G}{\partial x}+\frac{k_{eff}}{c_p}\frac{\partial^2 T_G}{\partial x^2}+\frac{h_t a}{c_p\varepsilon}(T_S-T_G)-\frac{4h_W}{c_p\varepsilon D_R}(T_G-T_W) \tag{1}$$

$$\frac{\partial T_S}{\partial t}=\frac{k_G a(-\Delta H)}{\rho_S c_{p,S}(1-\varepsilon)}(c_G-c_S)-\frac{h_t a}{\rho_S c_{p,S}(1-\varepsilon)}(T_S-T_G) \tag{2}$$

$$\frac{\partial(\rho y_G)}{\partial t}=-\frac{\partial(v\rho y_G)}{\partial x}+D_{eff}\frac{\partial^2(\rho y_G)}{\partial x^2}-\frac{k_G a}{\varepsilon}\rho(y_G-y_S) \tag{3}$$

The interstitial velocity v has been used and an effective gas conductivity has been considered, which includes the effects of the bed conductivity; subscripts G, S and W refer to gas, solid and wall respectively. In the inert section no mass transfer and reaction occur, and consequently there is no heat generation. For the gas phase Dankwerts boundary conditions are assumed.

In the following the results are given in terms of the dimensionless variables: axial position, $z=x/L$; radial position, $y=r/D_R$; exit concentration, $Y^e_G=y_{G,out}/y_{G,feed}$; gas temperature, $\tau_G = T_G/T_0$; Stanton number at the wall, based on overall transfer coefficient, $\mathrm{St}_W=4Lh_W/D_R v\varepsilon\rho_0 c_{p,G}$. A 0.2 m diameter, 3 m long packed bed reactor, with particles of 6 mm (void fraction 0.4) has been modelled; the active fraction is 20%. The gases are fed at 293 K, with a surface velocity of 0.4 m/s.

It must be evidenced that the expanded monodimensional model is not suitable to obtain information on the extent of radial temperature and concentration profiles. Furthermore, it is not possible to quantify the smoothing effect of wall conductivity on axial temperature profiles. Therefore some simulations have been carried out (for the reversed flow reactor) using a two-dimensional model that accounts for radial heat dispersion within the bed, bed-to-wall and external heat transfer coefficients.

The apparatus is operated in forced non-stationary conditions; anyway, a periodic state can be reached after typically 100 to 200 switches. This periodic state is characterised by identical temperature profiles at the beginning of each cycle, and by constant time-average conversion; therefore it is also referred to as pseudo-steady-state.

The easiest possible control strategy is to define a fixed cycle period. This is usually done for the reverse flow reactor; with the network of burners it is possible to use this strategy, but only a very narrow range of cycle periods determined by flow velocity is allowed. It has been observed that for an adiabatic reactor, periods of 2025 - 2075 residence times give good conversion, for the case considered, while shorter or longer switching times do not result in a stable state of operation. The increased loss of heat at unfavourable periods cannot be compensated by reactive heat development, and the system gradually "lights off".

This means that the period has to be known in advance with an error smaller than two percent. But if the gas velocity is subjected to fluctuations, this strategy obviously is inadequate for any kind of reactor configuration. It is not assured that an effective pseudo steady-state can be reached, since in all simulations the maximum temperature does not converge even after 200 switches. Finally, there is no way to control the maximum temperature in the reactor, so that overheating and consecutive catalyst destruction may occur.

Therefore a two-point temperature control has been adopted for the network of reactors: the first temperature control is at the beginning of the first catalytic bed, while the second one is at the beginning of the second catalyst bed: the feed position is changed only if the local temperature drops below set point 1 and is above set point 2. $T_{sw,1}$ =530 K and $T_{sw,2}$ =535 K have been chosen. The choice of the set-points has not a significant effect on the cycle time.

Results

The influence of non-adiabatic operation has been investigated by means of the previously described monodimensional model. The standard value of St_W has been assumed to be 0.0343 according to a heat flux density of 50 J/m²sK at a temperature of 500K.

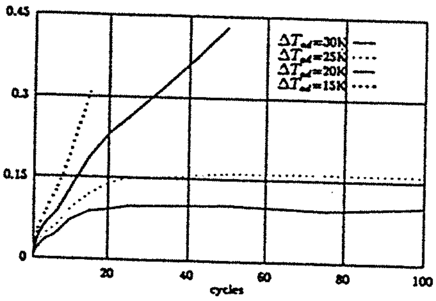

Fig. 2 - Influence of the adiabatic temperature raise under non adiabatic conditions in the three-bed reactor.

As Fig. 2 shows, the minimum adiabatic temperature rise for autothermal operation in the network of three reactors is 20-25 kelvin instead of 10K under adiabatic operation, with the same parameters and control strategy. The additional radial heat loss leads to a decreased maximum temperature and thus reduced conversion and heat recovery as demonstrated in Fig. 3 for the case ΔT_{ad}=30K. An increase of St_W leads to lightoff, which means that the effect of St_W is very pronounced as well.

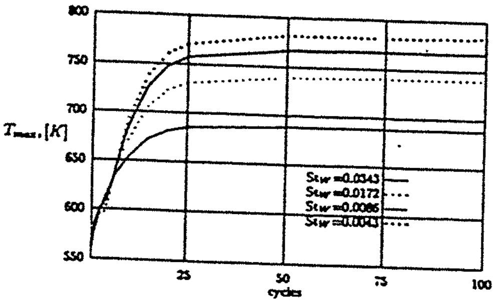

Fig. 3 - Influence of the wall Stanton number on maximum temperature at ΔT_{ad} = 30 K in the three-bed reactor

The analysis of the two-dimensional model reveals the complex behaviour of the actual system. As an example the reverse-flow reactor operated at ΔT_{ad}=10K and with a dimensionless period of $s_c = tv_0/L$=500 is chosen, and the results are demonstrated in Fig. 4. The axial temperature profiles correspond qualitatively to the adiabatic system. Radial profiles behind the catalytically active section are monotonously decreasing toward the reactor wall due to radial heat loss. However, wall conductivity and capacity smooth out the axial profiles near the wall. As a consequence the radial profiles on the ascending part of the temperature peak shows a minimum at about y=0.8. This minimum also affects local concentration to some extent (see Fig. 5). Anyway these effects should not seriously affect the overall performance, but a comparison with the monodimensional model remains interesting.

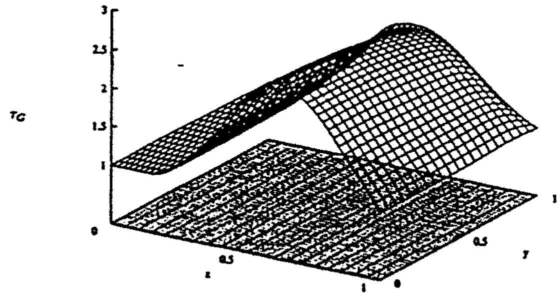

Fig. 4 - Dimensionless gas temperature in non-adiabatic reverse flow reactor.

A comparison of the performances of the network of three burners and the reverse flow reactor is difficult since the reverse-flow reactor is usually designed with at most 40% inert material, while the network reactors may contain about 80% inert. Furthermore, the reverse-flow system is usually operated at a fixed cycle time without temperature control to cope for load and flow fluctuations. On the other hand, we have shown that only a narrow range of fixed switching periods exists for the reactor network, and that two-point control enables a safe start-up at the expense of final conversion (at least if preheat temperature is not too high).

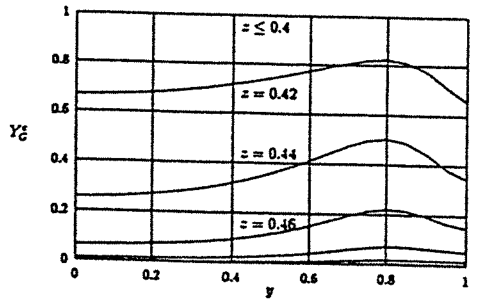

Fig. 5 - Radial concentration profiles in non-adiabatic reverse flow reactor.

The temperature profile in the reverse flow reactor is such that the active portion is always at high temperature and thus active, whereas in the reactor network a part of catalyst is always cold and thus inactive. Therefore, a reverse flow reactor with the same amount of catalyst and inert should be superior to the reactor network, but wash out of unburned gases must also be taken into account. The simulations show that maximum conversion in a reverse flow reactor corresponding to the complete network is about 99% at $\tau_c = 900$, with $T_{max} = 680K$. The reactor network with a fixed dimensionless period of 2050 performs even better: conversion is 99.5%, with $T_{max} > 740K$. The reason for the maximum in conversion vs. cycle time is due to the decreasing effect of washout at the beginning of the cycle with increasing period time. With further increasing cycle duration, the maximum temperature decreases until conversion drops and lightoff occurs. Note that a larger reactor shows higher maximum temperature at the same dimensionless period.

Conclusions

Catalytic reactors in forced non-stationary operation enable autothermal VOC-oxidation even at extremely low adiabatic temperature rise. The effect of heat loss at the wall has been investigated; non-adiabacity can be considered using an overall heat transfer coefficient to the exterior. In practical applications this coefficient approximately depends on the reactor insulation. Radial heat loss increases the minimum load required for autothermal operation. Reactor geometry may be optimised for reduced external surface area, but flow conditions and possible radial dependencies should be considered in this case.

Numerical simulation reveals that the network of three catalytic reactors in series with large inert sections is a suitable design for VOC oxidation at low concentration. The system can be controlled by a simple set of two switches acting according to temperature set points. This strategy serves for start-up and is stable with respect to fluctuating inlet concentrations and also of gas flowrate, but conversion depends on set point and thus on preheat temperature.

In contrast to this, fixed switching periods show excellent conversion, but are confined to a narrow range and are not stable at variable inlet conditions.

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. However, more information on both designs is necessary in order to decide which one is favourable in a particular application.

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