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Forced unsteady state methanol synthesis in a reactor network

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Most processes in the chemical industry operate under steady-state conditions, following the prevailing technological outlook that high process efficiency can be mainly achieved operating at constant conditions. However in the last two decades theoretical and experimental investigations show that the performance of chemical processes can be significantly improved when operating under transient conditions. Among the various modes of transient operation, forced periodic operation combines the benefits of permanent unsteady-state regime and fixed averaged production. Periodic operation can be attained mainly in two ways: periodic change in the feed composition and periodic reversal of the flow direction. The second possibility has been widely studied, and has been implemented in several industrial chemical processes [1]. The ring reactor, or reactor network, which consists of a sequence of two or more catalytic fixed bed reactors, is another way to obtain a dynamic sustained behaviour, thanks to the periodical alternation of the feed position. This configuration has been recently investigated for the combustion of lean VOC mixtures [2] and for the mildly exothermic methanol synthesis reaction [3]. The auto-thermal behaviour, with a nearly uniform catalyst exploitation, due to the constant flow direction, and the possibility to achieve an optimal temperature distribution, which makes possible the creation of favourable thermodynamic conditions for exothermic equilibrium-limited reactions, are the main advantages of the network. It however presents a small range of switching times t_c which allow to reach and maintain a pseudo-steady state of operation.

Methanol synthesis in a two and in a three reactors network has been simulated for different values of the switching time and of the inlet temperature proving that the yield and selectivity of the process are generally higher than the reverse flow reactor and the traditional technology. Numerical simulations reveal that both the network of two and three reactors can operate in two regions of t_c values: the high t_c and the low t_c operating range. Figure 1 shows a comparison between the performances of a network of three and two beds for different inlet temperatures and constant total length. The behaviour of the network in these two configurations is quite similar, showing that better results can be expected at low switching time values.

Out of the useful operating regions, two other reactor states are possible: extinction or complex behaviour. In the latter, even if permanent pseudo-steady-state conditions have been reached, the profiles in a generic cycle are different from the others since the material and energy balances have a solution of period greater than the cycle time. As we move away from the stability operation range the period of the solution increases until a non-ignited state is reached. The knowledge of the dynamic behaviour of the reactor out of the operating regions is very important for the control of such a system, because the condition of maximum yield is very close to the zones of complex periodic behaviour.

In order to attain and maintain stable periodic operations, it was adopted a simple open-loop control strategy, consisting in the periodic alternations of the inlet sections at fixed time interval t_c. Numerical simulations show that this open-loop control allows for a safe start-up, with an appropriate choice of the bed pre-heating temperature, but care must be paid when the inlet conditions are subjected to fluctuations. In fact, the open loop control is able to take the system to its original point of operation just only after a short length disturbance (Figure 2, left hand side). Instead, after a long time perturbation (Figure 2, right hand side), the previous state of operation cannot be restored by the simple action of the open loop control and the system can be driven out from the stability range.

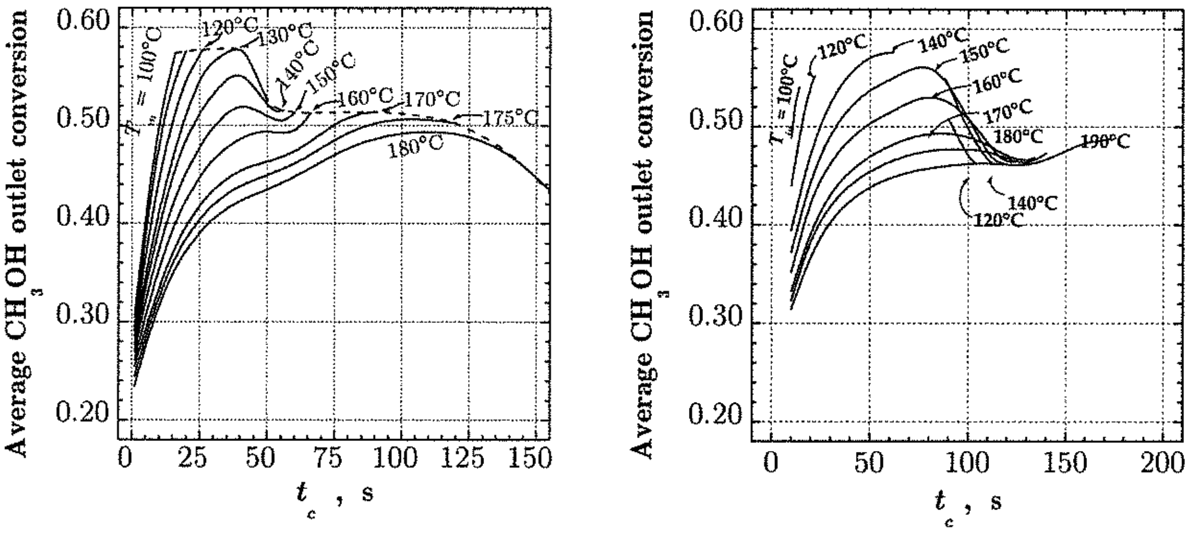

Figure 1 *Average methanol conversion for different values of the switching time and of the inlet temperature. Network of three reactors (left hand side) and network of two reactors (right hand side).*

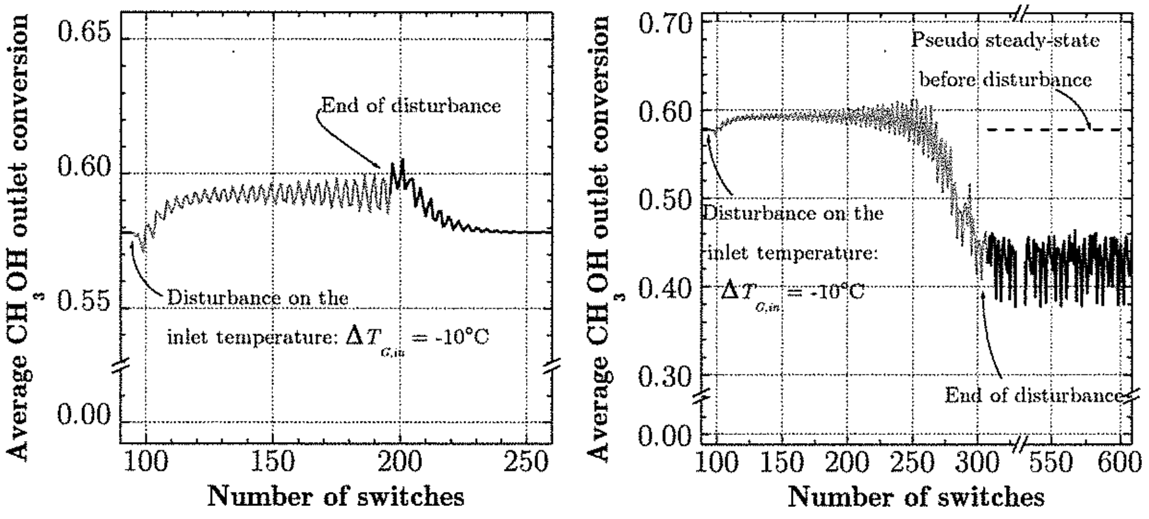

Figure 2 *Left hand side: open loop response to a step disturbance on the inlet temperature and restoration of the original steady-state. Right hand side: open loop response to a step disturbance on the inlet temperature and establishing of a new complex steady state.*

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