ANALYSIS OF REACTING MIXTURES IN A HEAT INTEGRATED SIMULATED MOVING BED REACTOR

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Summary

A SMBR with adiabatic segments is used for an autothermal operation. Feed mixtures are common in industry which motivates a thorough investigation of the steady state properties and the dynamics of the reactor. Based on experimental work the operation windows are acquired and discussed.

Keywords

Novel reactor technologies, Dynamics and control of chemical reacting systems, Environmental Reaction Engineering

Introduction

In recent years energy savings have become a crucial reactor design criterion. Thereby heat integrated concepts can offer substantial advantages in contrast to traditional processes and have been investigated theoretically and also experimentally[1].

For gas phase reactions conducted in catalytic fixed bed reactors it was shown that regenerative heat exchange is very efficient, especially for the purification of industrial waste gases by catalytic total oxidation. This is because the application of heat exchangers (recuperative heat exchange) offers only a limited performance due to their unfavorable ratio between heat exchange area to equipment size. The unsteady operated reverse flow reactor developed by Matros and co-workers exploits the regenerative heat exchange in a fixed bed and is industrially applied[2]. Cold feed gases are entering a catalytic fixed bed and are converted within a narrow reaction front, which propagates in the direction of the gas flow. Before leaving the fixed bed the flow direction is reversed and a heat front moves in the opposite direction. This process shows also several unwanted features. Reversing the flow direction leads to a release of unconverted material immediately after the switching event. This "slip" can be reduced e.g. with the help of guard beds. An also undesired property is the non-uniform catalyst exploitation due to the narrow region the heat front repeatedly moves along.

Another attractive option to achieve an autothermal operation is the simulated moving bed reactor as suggested e.g. by Haynes a Caram[3,4]. The reactor is equipped with a certain number of adiabatic fixed bed segments connected in series. Again the cold feed gas is entering the reactor and a heat front propagates through the segments in the direction of the gas flow (fig. 1). After a specific time the feed and product ports are switched in the flow direction by one reactor segment. Hereby the cold feed segment is shifted to the end of the cascade and is reheated again.

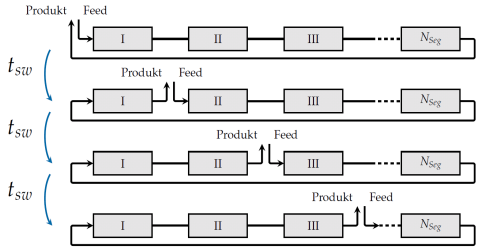

Fig.: 1 Principle of a simulated moving bed reactor

This operation has the advantage of an unidirectional propagation of the heat front and a uniform exploitation of all fixed beds. Although direct dynamic calulations provide a valuable description of the process dynamics, one is also interested in methods describing the qualitative behaviour in terms of process parameters. In our recent study we have developed a reduced model to approximate the periodic process and to predict the transition between ignited and extinguished states of the reactor[5]. It was shown that the following reduced switching time is usefull to investigate parameter dependencies for simple reactions.

$$\gamma = \frac{t_{sw} N_{Seg}}{(1-\varepsilon) L_R} \frac{u_g}{Le}$$

Where t_{sw} is the switching time, N_{seg}, L_R and ε are scaling parameters, u_g is the inlet gas velocity and Le the Lewis number. Our present work is devoted to experimentally and theoretically analyze the reactor behavior in case of

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more complex reaction mixtures. Due to different reaction enthalpies and activation energies the operation window can be affected significantly. Different reactor control schemes and the self-adaptivity of the reactor are studied.

Procedure and Results

The total oxidation of ethene and propene on $CuCrO_x/Al_2O_3$ was investigated. A large set of kinetic data was analyzed and kinetic parameters have been identified. The dynamics of the propagating reaction zone was investigated for the pure components and for the mixture on large fixed beds. Model based analysis of these experiments helped to identify appropriate transport parameters and to perform further theoretical studies.
The intrinsic property of the simulated moving bed reactor is a solution isola with respect to the switching time, meaning, that if the switching time is adjusted to the front velocity, accumulation of energy is possible. This kind of focusing leads to a high temperature stable solution with full conversion, separated from the stable extinguished by an unstable one.

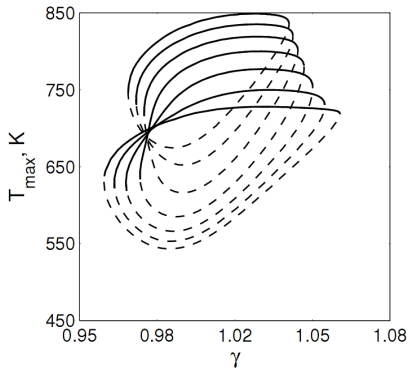

Fig.: 2 A family of isola solutions of a mixture of ethene/propene corresponding to a total adiabatic temperature rise of 30 °C at a constant low inlet temperature of 300 °C (extinguished solutions not shown).

The shape and the dimension of the characteristic operation region depend on physicochemical (E_a, ΔH_R, k_0) and operating parameters (γ, T_{in}, u_g, c_{in}…). For the particular example, each individual component shows different ignition temperatures and different maximum temperatures. Lower activation energies and higher reaction enthalpies lead to a larger area of possible switching times to ensure an ignited state. The maximum temperature also decreases when the components with lower ignition temperature dominate. With increased fraction of the component with low activation energy an almost smooth transition of the operating region is observed.
Results for the mixtures are shown for the parameter plane (γ, T_{in}) in figure 3. This figure illustrates the position of the limit points, separating stable from unstable states, e.g. two limit points exist for the isola (fig. 2) at a certain inlet temperature. For higher inlet temperatures the isola should become larger and finally at very high inlet temperatures an auto ignition is possible (fig. 3). This analysis elucidates the influence of the activation energies.

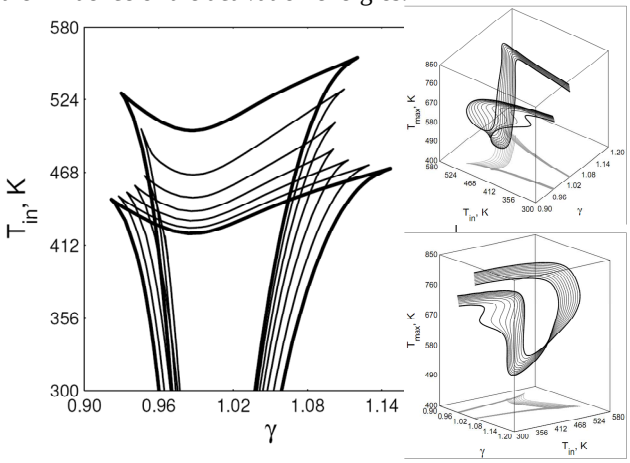

Fig.: 3 A family of limit point curves enclosing the region of three steady states in the parameter plane of the flow rate ratio γ and the inlet temperature T_{in}. The solutions of mixtures (ethen /propene) are shown. The graphs right illustrate the maximum temperatures.

It will illustrated that an isola center possesses the most singular behavior and our method allows for a fast evaluation of the reactor performance under various conditions. Also control schemes which are capable to stabilize the reactor are compared.

Summary and Outlook

This contribution investigates the behavior of an adiabatic simulated moving bed reactor to totally oxidize mixtures of hydrocarbons. The analysis is based on experimental work of short chained hydrocarbons. It exploits singularity analysis to calculate dependencies. Dynamic simulations demonstrate the stability of control schemes.

References

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