

Removal of Volatile Organic Compounds from Polluted Air in a Reverse Flow Reactor: An Experimental Study

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An experimental study of the reverse flow reactor for the purification of contaminated air has been carried out. An experimental reactor with an inner diameter of 0.145 m has been constructed. It almost completely reached the goal of an adiabatically operating system. The influence of several operating parameters such as gas velocity, cycle period, chemical character, and concentration of the pollutants and reactor pressure are discussed. The reactor could be operated autothermally provided that the inlet concentrations were sufficiently high. If a mixture of contaminants is fed to the reactor, it might be necessary to increase the total hydrocarbon concentration to assure an autothermal process. Increasing the reactor pressure will hardly change the axial temperature profiles, if the mass flux is kept constant. Increasing the mass flow rate will lead to a higher plateau temperature. Not only the reactor behavior at fixed operating conditions, but also the response of the reactor toward variations in inlet conditions is reported.

Introduction

Polluted air can be decontaminated from volatile organic compounds by making use of catalytic oxidation in a packed bed reactor. Matros (1989) has shown that unsteady-state reactor operation can be profitable for chemical processes. After the pioneering work of Boreskov, Matros, and co-workers several other research groups started to investigate the reverse flow reactor (RFR): an adiabatic packed bed reactor in which the direction of the feed flow is reversed periodically, so the reactor is forced to operate under transient conditions. Despite the many papers dealing with the behavior of this type of reactor for several processes—see, e.g., van de Beld and Westerterp (1994)—the number of experimental studies has been limited to only a few. The reverse flow reactor, if operated on a large scale, will behave close to adiabatically, and therefore it is important to have adiabatic conditions in a laboratory scale reactor. The dynamics of the system should be well-defined and not influenced by its surroundings. This excludes the usage of insulation to obtain adiabatic conditions, because for a reasonable resistance against heat losses the amount of insulation needed is very large. The heat capacity of the insulation can easily be larger than that of the packed bed and thus a significant additional heat buffer is created. Also applying compensation heating has to be avoided, because in that case the dynamics of the system are certainly influenced and no more well-defined.

In Table 1 an overview is given of published experimental studies on RFR's. In most studies insulation has been applied to suppress radial heat losses, sometimes extended with electric heat loss compensation. Several processes have been studied by the Russian researchers: they reduced the influence of heat losses by choosing a large reactor diameter. They have also demonstrated the applicability of the RFR on an industrial scale. Blanks *et al.* (1990) recognized the problem of achieving adiabatic conditions in a laboratory scale

reactor, and after some preliminary experiments they concluded that the reactor diameter should be increased. Therefore they have built a much larger plant, but still applying large amounts of insulation. In our opinion the best way of achieving adiabatic conditions and minimizing the influence of the surroundings is making use of an evacuated jacket; see also Bos *et al.* (1993). Such a system was also applied by Neophytides and Froment (1992): their reactor dimensions were very small. At high operating temperatures radiation shields should be included in the jacket.

We have studied the catalytic combustion of organic components in a RFR, and in this paper the experimental results are discussed. Applying a RFR for air cleaning makes an autothermal process possible with low inlet concentrations of the contaminants and enables the reactor to handle fluctuations in inlet conditions like gas flow and inlet concentration, as they will appear in industrial practice.

Experimental Setup

In our laboratory an experimental installation containing a reverse flow reactor has been built. In Figure 1 a schematic drawing of the setup is given. The reactor is made of Inconel 601, it has an inner diameter of 145 mm and a length of 1 m, and the wall thickness is 1.6 mm. The reactor is equipped with an evacuated jacket; the total diameter is about 250 mm. The outer tube is made of stainless steel. The pressure inside the jacket is monitored continuously with a vacuum gauge, an Edwards Pirani-11. In the reactor the temperatures can become rather high, and therefore three radiation shields have been placed in the annulus. The first 0.175 m of the reactor is filled with inert α -Al₂O₃ pellets, which have negligible catalytic activity for the oxidation reactions; see van de Beld and Westerterp (1994b). On this inert layer 0.65 m of the catalyst, Pd/ γ -Al₂O₃, is placed; above that the rest of the reactor is again filled with inert material. The particles are cylindrical and have a height and diameter of 4.5 mm. Inside the reactor a frame of Inconel 601 is placed in which 0.5

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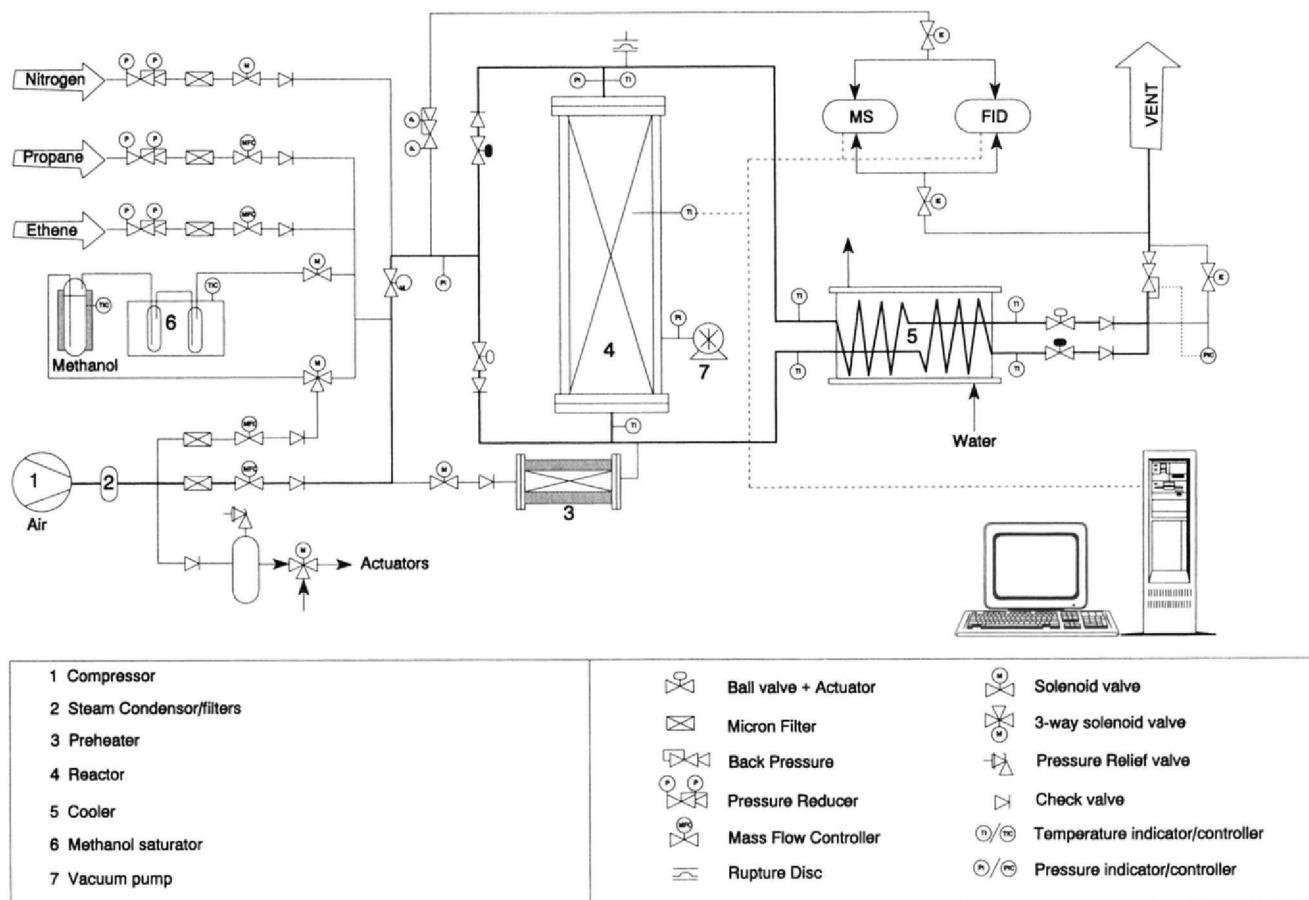


Figure 1. Flowsheet of the experimental setup.

Table 1. Overview of Experimental Studies for the Reverse Flow Reactor

research group	process	reactor dimensions	radial heat loss prevention	references
Institute of Catalysis, Novosibirsk, Russia	SO ₂ ⁻ oxidation	$D = 175 \text{ mm}, L = 0.44\text{--}1.8 \text{ m}$	200 mm insulation	Boreskov <i>et al.</i> (1982); Bunimovich <i>et al.</i> (1990)
	air purification	$D = 175 \text{ mm}, L = 3.0 \text{ m}$	200 mm insulation	Matros (1989); Matros <i>et al.</i> (1993)
	methanol production selective reduction of NO _x	$D = 68 \text{ mm}, L = 0.77 \text{ m}$ $D = 0.175 \text{ m}$	insulation	Matros (1989) Bobrova <i>et al.</i> (1988)
Universität Stuttgart, Germany	air purification	$D = 50 \text{ mm}, L = 0.84 \text{ m}$	285 mm insulation + heat loss compensation	Eigenberger and Nieken (1988); Nieken (1993)
Amoco, Naperville, IL	synthesis gas production	1. $D = 63.5 \text{ mm}$ 2. $D = 570 \text{ mm}, L = 4 \text{ m}$ 30 g of catalyst	insulation + heat loss compensation evacuated jacket	Blanks <i>et al.</i> (1990)
Rijksuniversiteit Gent, Belgium	methanol production		630 mm insulation evacuated jacket	Neophytides and Froment (1992)
Universität Karlsruhe, Germany	air purification	$D \sim 136\text{--}90 \text{ mm}, L = 0.594 \text{ m}$	insulation + heat loss compensation	Züfle (1993)
Twente University, The Netherlands	air purification	$D = 145 \text{ mm}, L = 1.0 \text{ m}$	evacuated jacket	this work

mm thermocouples have been fixed. At 20 different axial positions the gas phase temperature is measured. The total number of thermocouples is limited, so the radial temperature gradient cannot be evaluated. At 12 axial positions the solid phase temperature is measured. To this end a thermocouple has been inserted in a hole, drilled in a catalyst pellet.

The main air flow is delivered by a compressor of Hydrovane with a maximum flow of 840 L (NTP)/min and a maximum pressure of 8 bar. Before feeding into the reactor the air is dried: for all experiments the water content remains equal; the dew point is around 8 °C. To remove all other unwanted components, the air also flows through several filters made by Domnick Hunter. The gas flow is measured and controlled in a thermal mass flow controller supplied by Hitec Bronk-

horst. Ethene, propane, and methanol have been used as the organic contaminants. Ethene and propane are gaseous and can be simply added and controlled via mass flow controllers of Brooks. Methanol as a liquid has to be vaporized. To ensure saturation of the methanol, the following system is used: part of the air stream flows through liquid methanol kept at about 35 °C. Next the methanol loaded air is cooled to 25 °C. The methanol concentration can be varied by changing the air flow rate.

The total hydrocarbon concentration is detected online with a FID analyzer of JUM Engineering. By switching a solenoid valve the inlet or outlet tube is connected. A QMG 112A quadrupole mass spectrometer of Balzer has been installed to follow the conversion of each individual component in the mixtures. The reactor

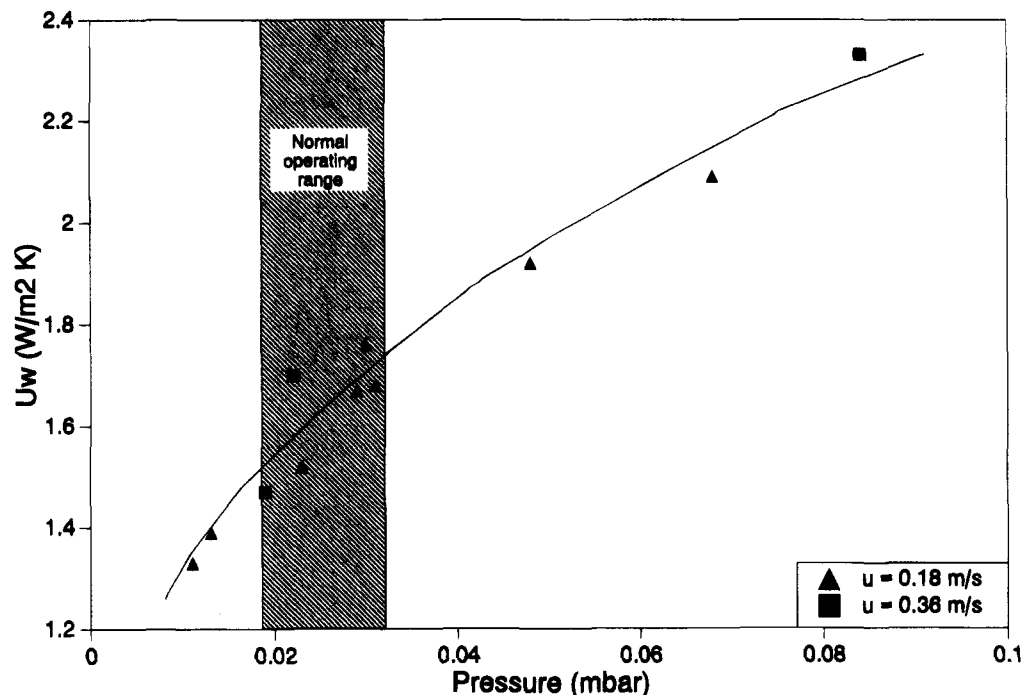


Figure 2. Overall radial heat transfer coefficient as a function of pressure in vacuum annulus. $u_g = 0.18\text{--}0.36$ m/s; $T_0 = 50\text{--}500$ °C.

Table 2. Range of Operating Parameters for the Experimental Installation

pressure	1–5 bar
temperature	<650 °C
gas velocity	0.1–0.6 m/s
flow	6–36 m ³ /h (NTP)
HC concentration	<0.5 vol %
contaminants	ethene, propane, and methanol

pressure is controlled by means of an electronic back-pressure controller. The pressure is measured on the top of the reactor, and depending on the flow direction it indicates the inlet or the outlet pressure of the reactor. For safety reasons a rupture disk has been installed. The direction of the feed flow is switched using four ball valves equipped with pneumatic actuators. The air pressure on all actuators is regulated by one solenoid valve, so all valves will turn at the same moment.

Before an experiment is started, air is heated in an electric heater to 600 °C at maximum and the hot air is fed to the reactor. To protect the downstream equipment—especially during preheating—the outlet stream of the reactor is cooled with water.

Table 2 lists the range of operating parameters. The installation is completely automated, and it can be operated continuously. All process parameters can be changed on the computer, and the computer will take care of all actions needed. The critical parameters like maximum temperature, pressure, and concentration of the combustibles are checked against alarm values, and if necessary, the installation shuts down automatically and the system is purged with nitrogen. The computer itself is checked by a so-called electronic “watchdog”, and if it does not work properly due to soft- and/or hardware errors, the experiments are terminated and the installation is shut down safely. All measured data are collected by a Hewlett Packard Data Acquisition and Control Unit.

Results and Discussion

The influence of several operating parameters on the behavior of the reverse flow reactor will be discussed

on the basis of the axial temperature profiles and overall parameters like maximum temperature and conversion. We will distinguish between the pseudo steady state (PSS) and the dynamic response of the reactor toward variations in operating conditions. In the PSS the temperature profiles are constant over a cycle. As mentioned before, it is important to have adiabatic conditions or at least know the impact of heat losses on the results. Therefore the adiabaticity of the reactor will be discussed first.

Adiabaticity of the Reactor. On a laboratory scale it is very difficult to obtain adiabatic conditions and significant heat losses will always be present. The overall radial heat transport coefficient has been determined in steady-state experiments. Air is heated to a certain temperature and fed to the reactor. For these experiments the direction of the feed flow is *not* reversed and is also *without* chemical reaction. Then for an adiabatic reactor operated under steady-state conditions the inlet and outlet temperatures will be equal to each other. Deviations are now caused by radial heat losses, leading to the following heat balance:

$$\frac{dT}{dz} = -\frac{4U_w}{(\rho C_p)_g u_g D} (T(z) - T_\infty) \quad (1)$$

Integration yields:

$$\ln\left(\frac{T(z) - T_\infty}{T(0) - T_\infty}\right) = -\frac{4U_w}{(\rho C_p)_g u_g D} z = -\frac{1}{HTU} z \quad (2)$$

The steady-state temperature profile has been measured for different inlet temperatures, $T(0)$, and different gas velocities. The overall radial heat transport coefficient, U_w , is sensitive to the pressure in the vacuum annulus. In Figure 2 U_w is plotted as a function of this pressure. Under normal operating conditions the pressure in the annulus is between 0.02 and 0.03 mbar and the overall radial heat transfer coefficient is about 1.5–1.6 W/(m² K). Further the resistance against radial heat transport lays outside the reactor, because increasing the gas velocity does not result in an increase of U_w : probably,

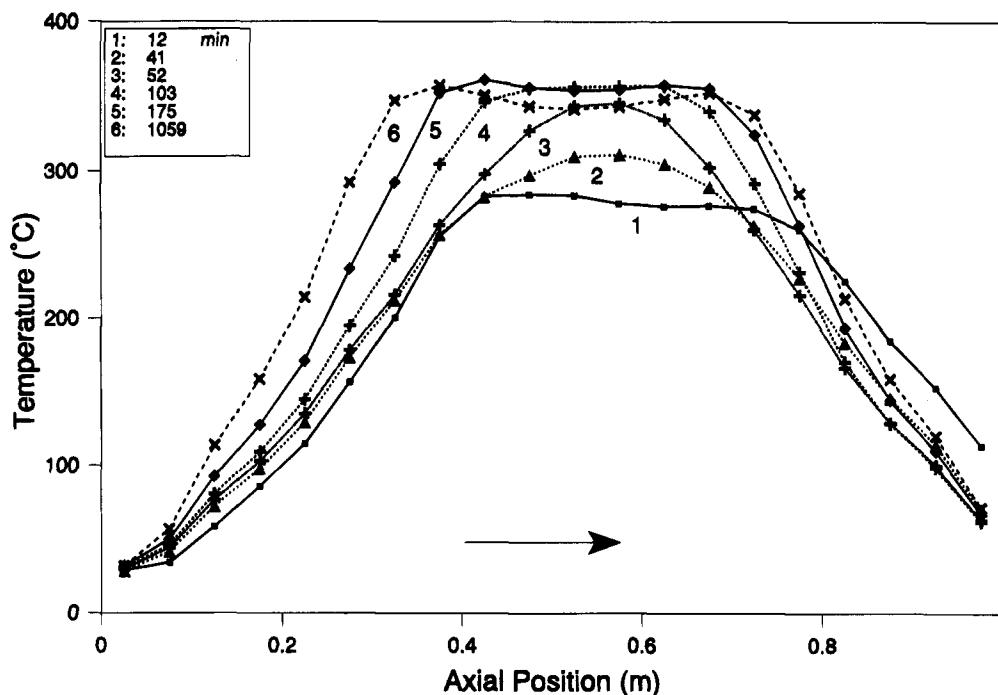


Figure 3. Development of axial temperature profiles. $u_{g0} = 0.55$ m/s; $P = 1.48$ bar; $T_0 = 30$ °C; $t_c = 145$ s; $C_{\text{ethene}} = 0.068$ vol %; $\Delta T_{\text{ad}} = 30.3$ °C.

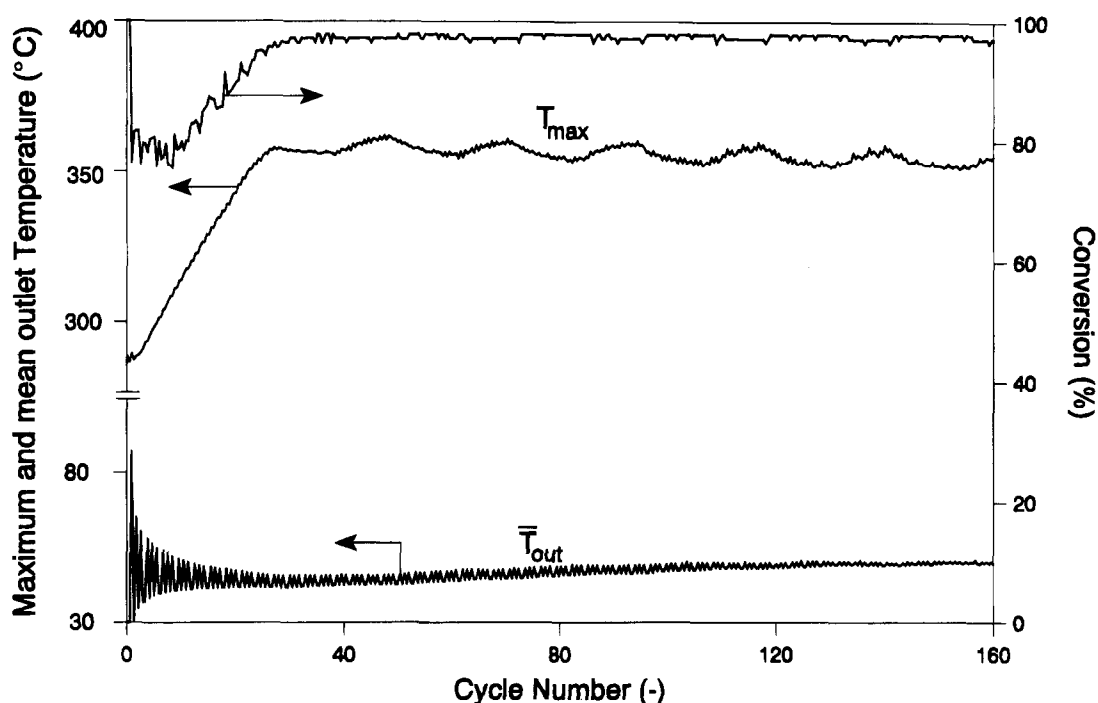


Figure 4. Conversion, maximum temperature, and mean outlet temperature as a function of cycle number. For conditions see Figure 4.

radial concentration and/or temperature gradients can be neglected.

Development of Temperature Profiles. In Figure 3 the development of the axial temperature profiles is shown. For simplicity only the profiles in one flow direction are given. For profile 1 the temperature is low; thus the chemical reaction is slow and reaction only takes place in the center of the reactor. The conversion is about 75%. In the first instance only the temperature in the center of the reactor will increase, because the reaction heat is released there; see profile 2. The profiles will further develop in the direction of the inlet and outlet of the reactor. After a large period of time—in our installation normally 10–20 h—the profiles become

constant over a cycle. For the same experiment the maximum temperature, the mean outlet temperature, and the conversion are plotted as a function of the cycle number in Figure 4. One cycle consists of half a cycle upflow and half a cycle downflow. The measured conversion for this experiment is around 98%. At start-up the maximum temperature increases very rapidly and the maximum is found in the center of the reactor. After that the position of the maximum shifts to the inlet and outlet of the reactor, but the absolute value of the maximum temperature hardly changes. The shifting of the position can also explain the undulations of the T_{max} observed in Figure 4, because with increase of time the location of the highest temperature moves

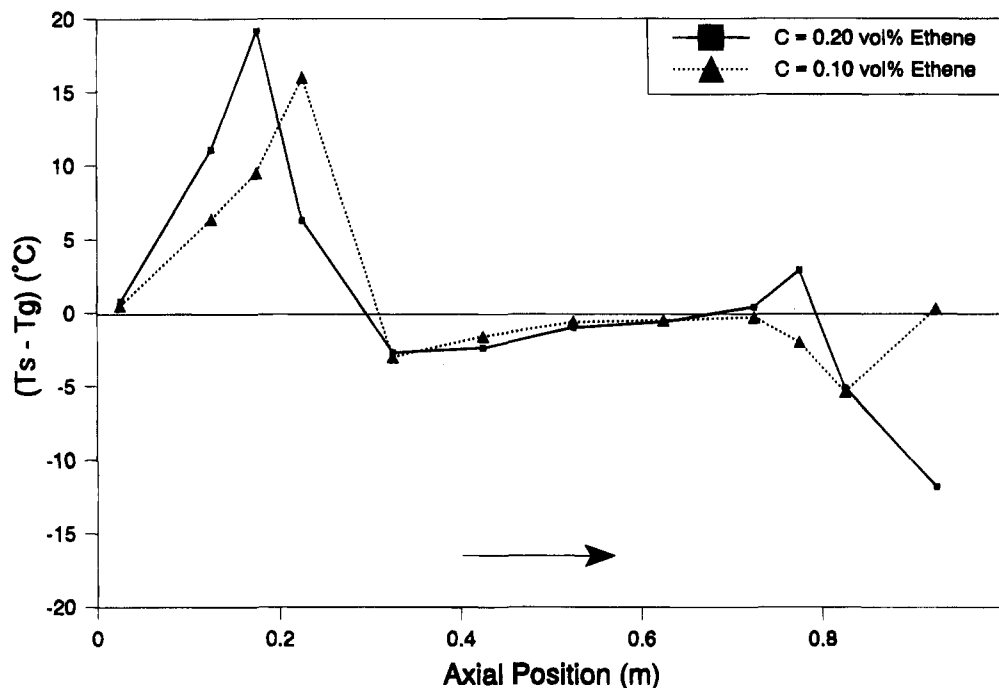


Figure 5. Difference between catalyst and gas phase temperatures for different inlet concentrations of ethene. $u_{g0} = 0.40$ m/s; $P = 1.44$ bar; $T_0 = 30$ °C; $t_c = 400$ s.

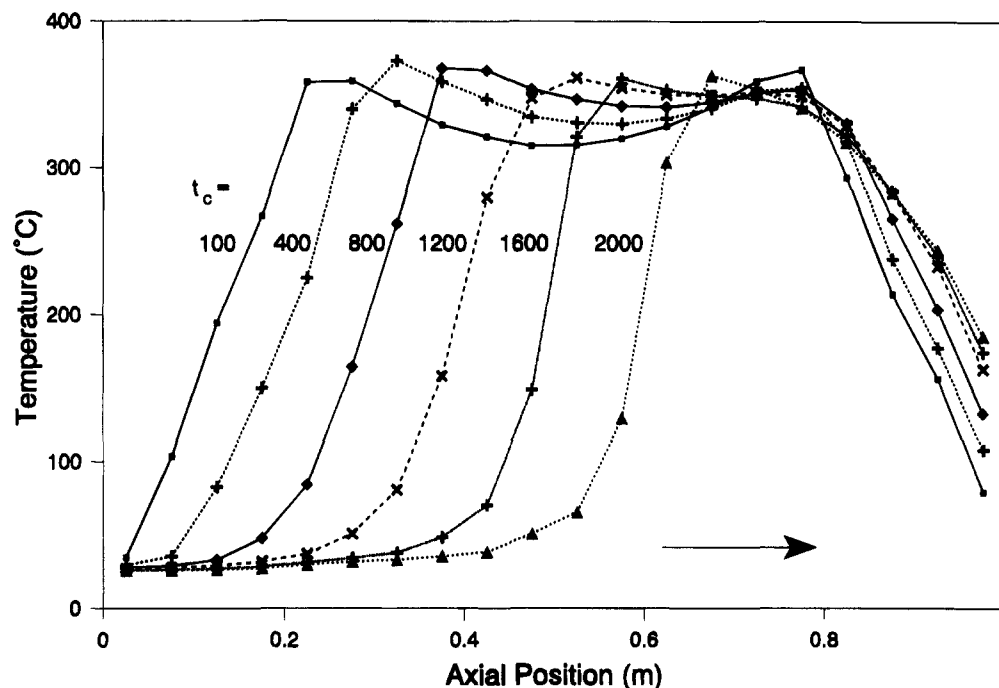


Figure 6. Influence of cycle period on axial temperature profiles. $u_g = 0.40$ m/s; $P = 1.44$ bar; $T_0 = 30$ °C; $C_{\text{ethene}} = 0.097$ vol %; $\Delta T_{\text{ad}} = 43.2$ °C.

to another thermocouple. In the pseudo steady state there is no accumulation of heat in the reactor; the reaction heat is removed from the reactor by convection via an increase in the outlet temperature. For the experimental reactor a correction should be made for the heat losses: the following heat balance should hold:

$$\bar{T}_{\text{out}} = \int_0^{(1/2)t_c} T_{\text{out}}(t) dt = T_0 + \Delta T_{\text{ad}} - \Delta T_{\text{lost}} \quad (3)$$

From Figure 4 we can conclude that for the determination whether the PSS is achieved or not the mean outlet temperature is a better quantity to follow than T_{max} or the conversion, because as long as the temperature

profiles are changing the mean outlet temperature will not be constant. For the experiment given in the Figures 3 and 4 ΔT_{lost} is about 10 °C, which corresponds with 30% of the reaction heat.

In the figures given before only gas phase temperature profiles have been shown; also the solid phase temperature profiles have been measured. In Figure 5 the difference between gas and solid phase temperature is depicted as a function of the axial position and for different inlet concentrations. Although this difference is afflicted with a large experimental error, the shape of the profile is as expected from a theoretical point of view. In the inlet section the solid phase will heat the

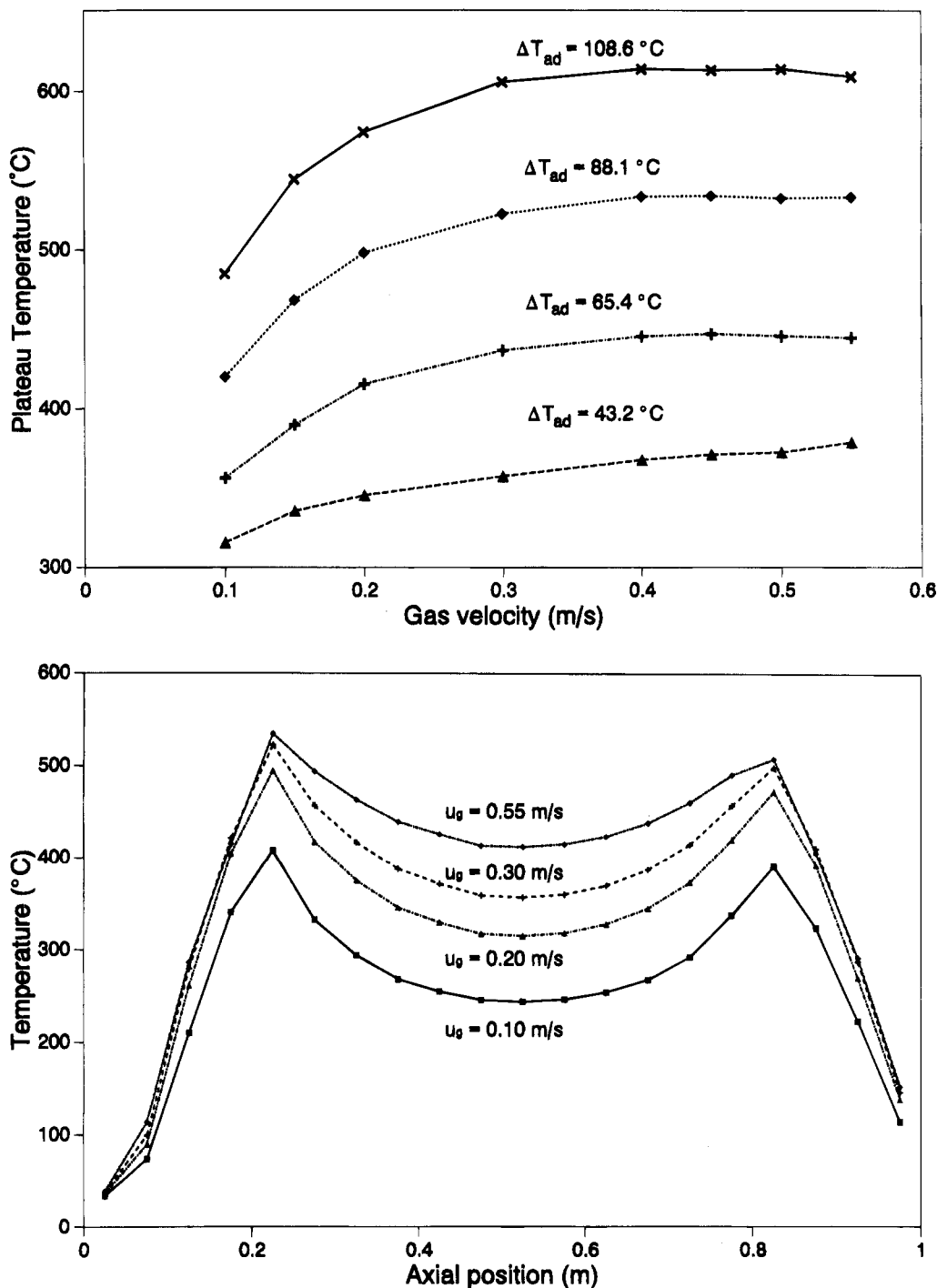


Figure 7. (A) Plateau temperature as a function of superficial gas velocity for different inlet concentrations of ethene. $T_0 = 30^\circ\text{C}$; $P = 1.04\text{--}1.48\text{ bar}$; $u_{gtc} = 80\text{ m}$. (B) Axial temperature profiles for different superficial gas velocities. $T_0 = 30^\circ\text{C}$; $P = 1.04\text{--}1.48\text{ bar}$; $u_{gtc} = 80\text{ m}$; $\Delta T_{ad} = 88.1^\circ\text{C}$.

cold feed gas and in the outlet section the hot gas will heat the solid phase. As the temperature profiles are very steep in these sections, small errors in axial position will cause large errors in the solid-gas temperature difference. The magnitude of this temperature difference depends on the reaction rate and adiabatic temperature rise. The reaction zone shifts toward the reactor inlet with increasing inlet concentration, and thus the maximum temperature difference is also found nearer the reactor inlet. If the direction of the feed flow is reversed, the mirror image with respect to the axial position is found.

Influence of the Cycle Period. It is obvious that the cycle period is an important parameter for operating

a reverse flow reactor. For different cycle periods the axial temperature profiles are given in Figure 6 just before the flow is reversed. The maximum temperature is almost insensitive to a change in cycle period; this is in good agreement with results reported in the literature. If the frequency of flow reversal is too low, too much heat will be removed from the reactor and at the end of the cycle the conversion starts decreasing so an autothermal process is no longer possible. If we look at a particular axial position in the reactor, the temperature changes over a wide range within one cycle and perhaps these temperature fluctuations will damage the catalyst. On the other hand, for a very small cycle period the temperature profiles hardly move. Now

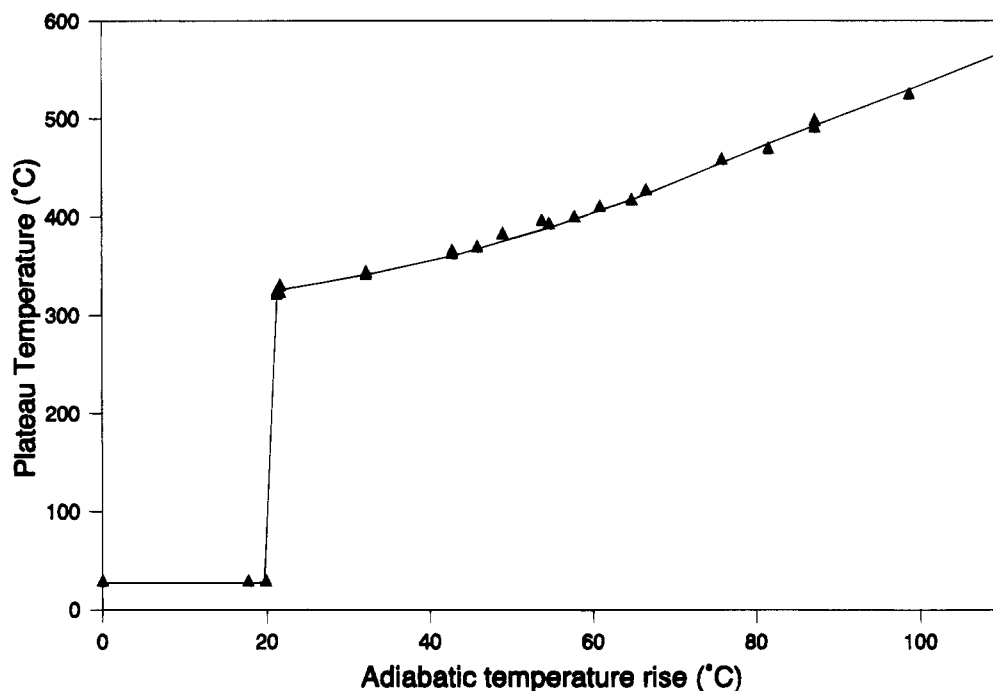


Figure 8. Plateau temperature as a function of adiabatic temperature rise for ethene. $u_g = 0.40$ m/s; $P = 1.44$ bar; $T_0 = 30$ °C; $t_c = 400$ s.

the overall efficiency of the process is lowered, because large amounts of feed gas will bypass the reactor at the moment of flow reversal. So finally we can conclude that the optimum cycle period is such that it allows the heat front to move over a certain distance, but not too far that the whole heat reserve is removed during one cycle. The position of the heat front at the end of a half cycle can be easily calculated with the following equation:

$$z = \frac{t_c}{2} v_h = \frac{t_c}{2} \frac{\epsilon(\rho C_p)_g}{(1 - \epsilon)(\rho C_p)_s} v_g \quad (4)$$

in which the factor 2 is introduced, because the cycle period is defined as two flow reversals. Comparison of results calculated with eq 4 and measured results gave good agreement; see also Westerterp *et al.* (1988).

Influence of the Gas Velocity. In Figure 7A the plateau temperature—the maximum temperature in the PSS—is plotted as a function of the superficial gas velocity and for different adiabatic temperature rises. If the gas velocity changes, the relative change in the heat front velocity is the same. For the experiments in Figure 7A not only the gas velocity but also the cycle period has been changed, such that the displacement of the heat front is the same during a half cycle; thus $u_g t_c = \text{constant}$ (see eq 4). This is validated by the corresponding axial temperature profiles given in Figure 7B. The minimum in the temperature profiles can be explained as follows: the reaction is completed in a small part of the catalyst bed. There is no reaction in the center of the reactor and thus no heat production. Hence the temperature will decrease due to a small heat loss in the radial direction. For low adiabatic temperature rises a monotonously increasing plateau temperature is observed with increasing gas velocity. For high gas velocities the plateau temperature remains nearly constant. From a theoretical point of view a maximum in the T_{\max} in the PSS or a decreasing T_{\max} at increasing values of u_g has to be expected; see van de Beld and Westerterp (1994). Experimentally this maximum is not found, because radial heat losses become more

important at low gas velocities, causing a decrease in the plateau temperature. Under truly adiabatic conditions it is likely the maximum will be found for a gas velocity of about 0.1 m/s.

Influence of the Adiabatic Temperature Rise.

One advantage of applying a reverse flow reactor is the possibility of purifying polluted air with an autothermal process without any external energy supply. To this end a minimum amount of combustibles should be present in the feed. Instead of a minimum feed concentration, it is probably better to consider a minimum adiabatic temperature rise, $\Delta T_{\text{ad, min}}$. In Figure 8 the plateau temperature is given as a function of the adiabatic temperature rise. In our installation an autothermal process could not be guaranteed below $\Delta T_{\text{ad, min}} \approx 22$ °C. Below that value no reaction occurs and the plateau temperature equals the feed temperature. Above this critical value a high conversion was obtained and the temperature rise in the reactor reached values several times the adiabatic temperature rise. Further increasing of the inlet concentration will lead to an increase in the plateau temperature larger than the increase in ΔT_{ad} . The minimum required ΔT_{ad} can be much lower under really adiabatic, such as industrial conditions; it is only slightly sensitive to the kind of combustible used.

Different Components and Mixtures. In practice different organic components or mixtures of components can be present in contaminated air. For our experiments ethene, propane, methanol, or mixtures of these components have been added to the feed flow. In Figure 9 the PSS axial temperature profiles are given for only ethene, only propane, and for mixtures of these two. In all cases the adiabatic temperature rise has been kept at a constant value of $\Delta T_{\text{ad}} = 43$ °C. Ethene is easier to oxidize than propane and thus the oxidation of ethene is completed at lower temperatures; see van de Beld and Westerterp (1995). A reverse flow reactor exhibits some self-controlling behavior: lowering the reaction rate will result in a higher plateau temperature; see, e.g., Eigenberger and Nieken (1988). In Figure 9 it can be observed that using propane results in a higher plateau tempera-

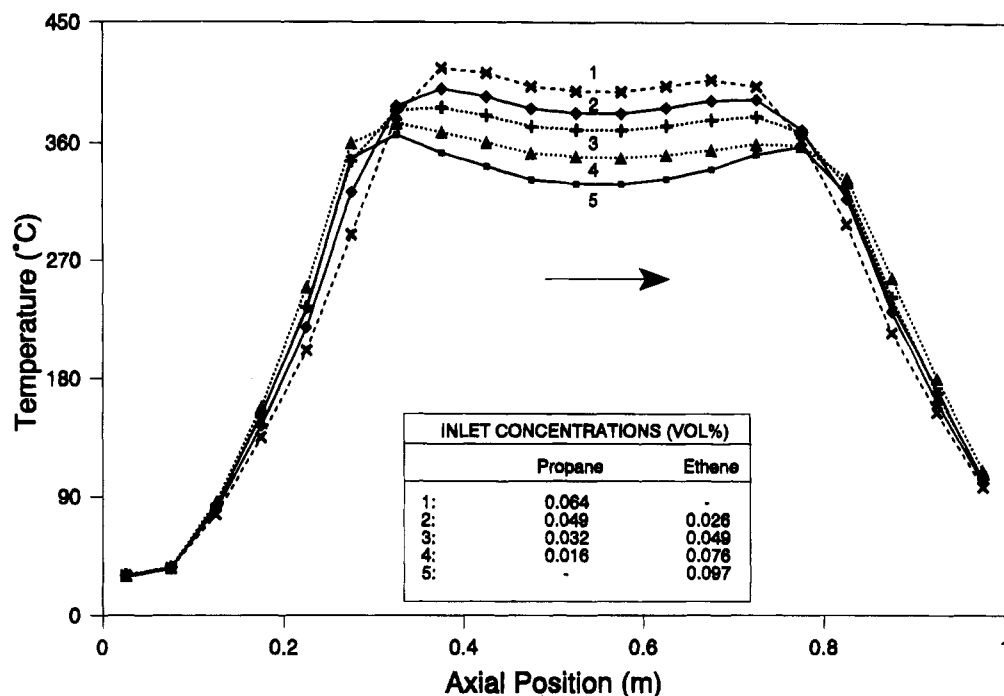


Figure 9. Axial temperature profiles for single components ethene and propane and for mixtures of these two. $u_g = 0.40$ m/s; $P = 1.43$ bar; $T_0 = 30$ °C; $\Delta T_{ad,total} = 43.2$ °C.

ture, whereas the width of the plateau becomes smaller. The temperature profiles for mixtures of propane and ethene are found between those of the single components. For high inlet concentrations the differences between the profiles will disappear, because at the corresponding high temperatures a complete conversion of all components is obtained in the same region of the reactor.

For single components the minimum adiabatic temperature rise, $\Delta T_{ad,min}$, is almost equal. For a mixture of ethene and propane a higher $\Delta T_{ad,min}$ is necessary, because the temperature level is too low to oxidize propane completely. For example, for an experiment with $\Delta T_{ad,ethene} = 15$ °C and $\Delta T_{ad,propane} = 15$ °C ethene was converted completely, but the propane conversion was just about 80%. From this it can be concluded that—if extra combustibles have to be added to the feed to attain higher bed temperatures and thus high conversions of all components—it depends on the temperature level required which component is the most effective additive. Most likely the component itself or a component which reacts in the same temperature range should be added. In principle we aim for a component that will burn in the middle of the bed and not already on the flanks of the hot zone.

Experiments with *methanol* gave totally different results. Although methanol is very easy to oxidize, high inlet concentrations were needed to prevent blowout of the reactor, but even so the conversion was limited to about 70%. This can be explained as follows: methanol adsorbs very strongly and in large quantities on the cold material in the inlet section of the reactor. After flow reversal this part of the reactor will be heated, so that the methanol desorbs and leaves the reactor unconverted. To prevent slip of such components an inert packed bed material should be chosen with low adsorption capability and capacity. This is not relevant for the catalyst material because the temperature of the catalyst in the bed is normally so high that adsorption will hardly take place.

Influence of Pressure. The air purification process will not be carried out at elevated pressure. Nevertheless, it is interesting to study the RFR at higher pressures, because it gives us the opportunity to change the ratio of the heat front and the gas velocities. The influence of the reactor pressure on the axial temperature profiles is shown in Figure 10 for two different adiabatic temperature rises. The heat front velocity is not influenced by the pressure. The cycle period has been kept constant, so again the movement of the heat front is the same during a half cycle for all experiments. It can be seen that the profiles hardly change if the pressure is increased by a factor of 3. The combustion reactions can often be described by first order reaction kinetics with respect to the hydrocarbon. Therefore increasing the pressure will lead to an increase in the reaction rate. In Figure 10 the plateau temperature decreases with increasing pressure and the temperature plateau does not become broader. This seems contrary to the explanation of increasing reaction rates. At the moment of flow reversal part of the reactor holdup is bypassed and leaves the system unconverted. Because the gas holdup increases with increasing pressure and the cycle period remains unchanged, this effect becomes more significant at higher pressures: due to this reactant loss the total heat production in the reactor and the overall conversion are reduced. For the experiment given in Figure 10 the bypass flow at 4 bar is estimated to be about 2% of the total flow. From the bypass point of view a high ratio between gas and heat front velocities is favorable, because it allows for high cycle periods and minimizes the bypass flow. A different, possible explanation for the decrease in the temperature at higher pressures is found in the experimental equipment. The flow delivered by the mass flow controllers can be slightly different at different operating pressures, and according to the FID measurements the concentrations were a little lower at higher pressures.

Response to Variations in Inlet Concentration. For industrial air purification processes obviously inlet

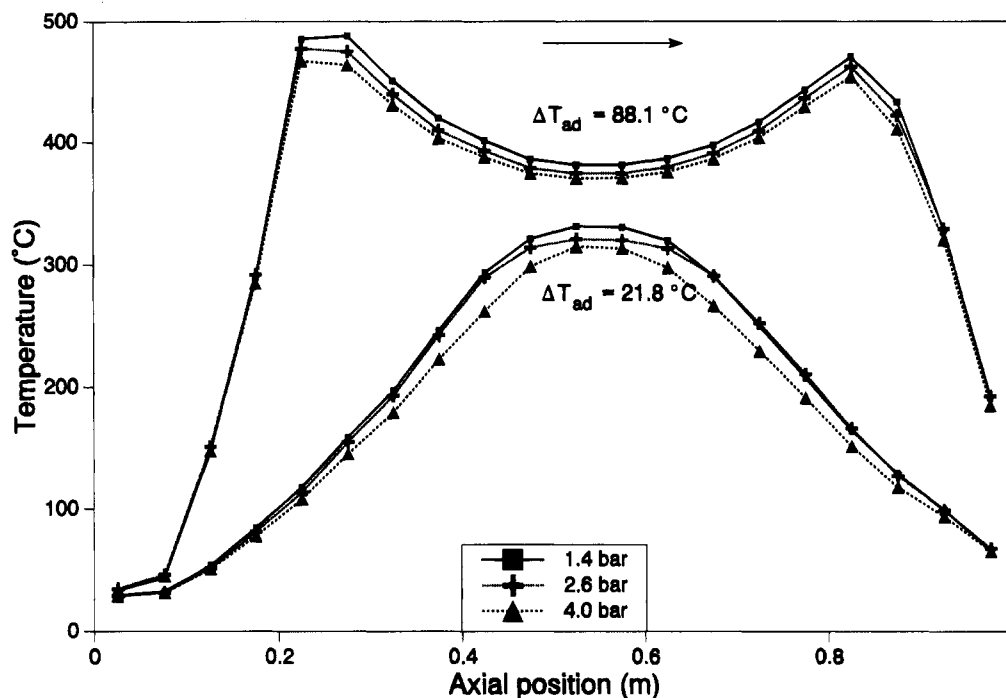


Figure 10. Influence of reactor pressure on axial temperature profiles at a constant mass flux and for different inlet concentration of ethene. $u_g = 0.40$ m/s; $T_0 = 30$ °C; $t_c = 400$ s.

conditions like concentration and chemical character of contaminants and flow rate are subject to fluctuations. In Figure 11 an experiment is shown in which a temporary decrease in inlet concentration has occurred. Initially ethene and propane are present in the feed flow and the concentrations are such that each component gives an adiabatic temperature rise of 35 °C. This assured high conversions of both components. In Figure 11A the maximum temperature and the total conversion are plotted as a function of time. At $t = 800$ min the inlet concentrations of both ethene and propane are decreased by a factor of 7. At these conditions an autothermal process with high conversion of the contaminants is no longer possible. Immediately after the change in the inlet concentrations the maximum temperature decreases; however, it takes some time before the conversion decreases. It depends on the specific conditions before and after the change for how long high conversions can be maintained. Using the mass spectrometer, we followed the individual conversions of ethene and propane, but due to the low concentrations the scatter in these results was rather large; the propane conversion started to decrease sooner. This is evident because the ethene reaction proceeds at lower temperatures. At $t = 970$ min the concentrations are reset to their original values, and full conversion is obtained very rapidly again. This behavior has been explained in more detail by van de Beld and Westerterp (1994). In Figure 11B the corresponding axial temperature profiles are given at different times, t_1 – t_6 . At $t = t_1$ the temperatures in the reactor are too low to achieve full conversion and reaction only takes place in the center part of the reactor. Initially, after the concentrations increase, the same conversion is obtained, but because the absolute concentration is much higher much more heat is released by the reaction and the bed temperatures will increase. Next the temperature profiles will further move toward the inlet and outlet of the reactor and much time is needed before the pseudo steady state is reached again.

Also the flow rate may change. To this end some experiments have been carried out. The influence was rather small, and the observed differences in temperature profiles can be explained by the increased radial heat losses at lower gas velocities. Switching from one component to another gives a different picture: if the new component is easier to oxidize and the concentration is high enough the process is continued without any troubles. If the new component is more difficult to convert, the situation is completely different: it depends on the temperature in the reactor whether the reaction starts at all. The inlet concentration determines whether enough heat is released to achieve a stable ignited pseudo steady state, or otherwise it may be necessary to supply additional heat to the system.

In a reverse flow reactor the catalyst is exposed to high temperatures and rapid temperature changes. This may cause a severe deactivation of the catalyst, in which case the plateau temperature will increase. A standard experiment has been carried out regularly to check whether the catalyst deactivates or not. After more than 6000 h of experiments we could not observe a noticeable change in the plateau temperature or in the temperature profiles. Small differences were found randomly over this period.

Conclusions

The purification of polluted air in a reverse flow reactor has been studied experimentally. The influence of several operating parameters on the reactor behavior has been discussed. Although radial heat losses cannot completely be excluded on a small scale, by making use of an evacuated jacket and several radiation shields the dynamics of the system are well-defined and the heat losses are minimized. The overall radial heat transfer coefficient was determined to be about 1.5 W/(m² K).

The reactor was operated successfully and high conversions of the contaminants were obtained without additional energy supply, provided that the inlet concentrations were not too low. In case of different components in the feed a high total hydrocarbon con-

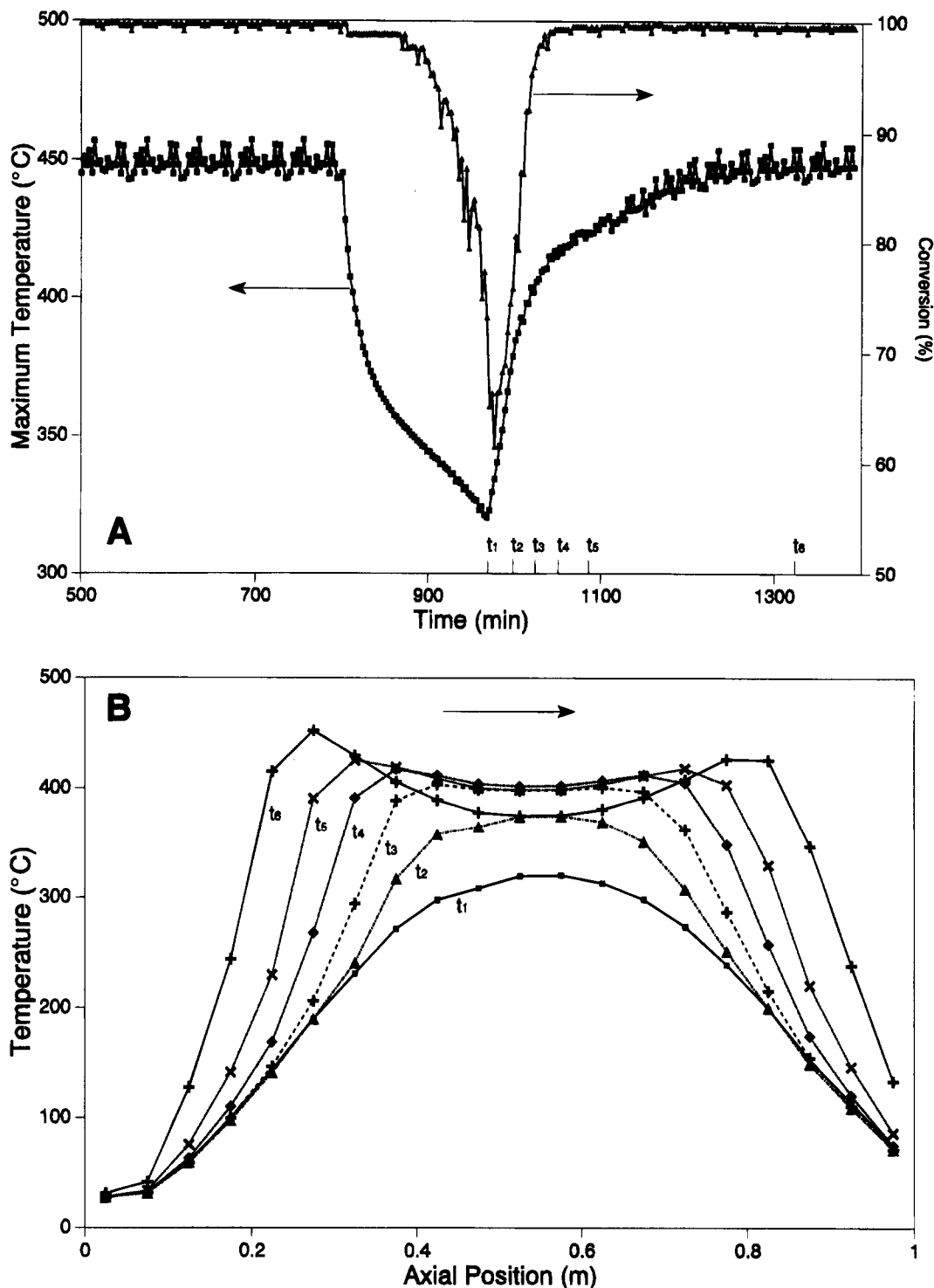


Figure 11. (A) Maximum temperature and conversion as a function of time. (B) Axial temperature profiles at different times. $u_g = 0.40$ m/s; $P = 1.45$ bar; $T_0 = 30$ °C; $t_c = 400$ s. Initial concentrations: $C_{\text{ethene}} = 0.079$ vol %, $C_{\text{propane}} = 0.051$ vol %; $\Delta T_{\text{ad}} = 70$ °C. At $t = 800$ min the inlet concentrations are decreased for 170 min: $C_{\text{ethene}} = 0.011$ vol %, $C_{\text{propane}} = 0.0074$ vol %; $\Delta T_{\text{ad}} = 10$ °C. $t_1 - t_6 = 970, 996, 1023, 1050, 1083, \text{ and } 1323$ min, respectively.

centration does not necessarily guarantee that all components individually achieve high conversions. Operating a reverse flow reactor under higher pressures and at equal mass flow rate does not significantly change the axial temperature profiles. Increasing the mass flow rate or gas velocity will result in a higher plateau temperature.

Besides the reactor behavior at fixed inlet conditions, the response of the reactor toward fluctuations in inlet conditions has been studied. A temporary change in inlet conditions—such that an ignited pseudo steady

state is no longer possible—could be handled quite well. The response of the reactor toward these variations is very slow.

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Nomenclature

C_p = specific heat capacity, J/kg K
 D = reactor diameter, m
 $HTU = (\rho C_p)_g u_g D / 4 U_w$, height of a transfer unit, m
 L = reactor length, m
 T = temperature, °C
 t_c = cycle period, s
 u_g = superficial gas velocity (at NTP), m/s
 v_g = interstitial gas velocity (u_g/ϵ), m/s
 U_w = overall radial heat transfer coefficient, W/(m² K)
 v_h = heat front velocity, m/s
 z = axial position, m

Greek Symbols

ϵ = bed porosity
 ρ = density, kg/m³

Sub- and Superscripts

g = gas
 s = solid
 ∞ = outside the reactor/surroundings

Literature Cited

- Beld, L. van de; Westerterp, K. R. Air purification by catalytic oxidation in a reactor with periodic flow reversal, *Chem. Eng. Technol.* **1994**, *17*, 217.
 Beld, L. van de; Westerterp, K. R. The catalytic oxidation of organic contaminants in a packed bed reactor. Accepted, *Chem. Eng. Sci.*
 Blanks, R. F.; Wittrig, T. S.; Peterson, D. A. Bidirectional adiabatic synthesis gas generator. *Chem. Eng. Sci.* **1990**, *45*, 2407.
 Bobrova, L. N.; Slavinskaya, E. M.; Noskov, A. S.; Matros, Yu. Sh. Unsteady-state performance of NO_x catalytic reduction by NH₃. *React. Kinet. Catal. Lett.* **1988**, *37*, 267.

- Boreskov, G. K.; Bunimovich, G. A.; Matros, Yu. Sh.; Ivanov, A. A. Catalytic processes under non-steady-state conditions. I. Switching the direction for the feed of the reaction mixture to the catalyst bed. Experimental results. *Kinet. Catal.* **1982**, *23*, 335.
 Bos, A. N. R.; Beld, L. van de; Overkamp, J. B.; Westerterp, K. R. Behavior of an adiabatic packed bed reactor. Part 1: Experimental study. *Chem. Eng. Comm.* **1993**, *121*, 27.
 Bunimovich, G. A.; Strots, V. O.; Goldman, O. V. Theory and industrial application of SO₂ oxidation reverse-process for sulphuric acid production. *Unsteady state processes in catalysis: proceedings of the international conference*, June 5-8, 1990, Novosibirsk, USSR; V.S.P. BV: Utrecht, The Netherlands, 1990; p 7.
 Eigenberger, G.; Nieken, U. Catalytic combustion with periodic flow reversal. *Chem. Eng. Sci.* **1988**, *43*, 2109.
 Matros, Yu. Sh. Catalytic processes under unsteady-state conditions. *Studies in Surface Science and Catalysis*; Elsevier: Amsterdam, 1989; Vol. 43.
 Matros, Yu. Sh.; Noskov, A. S.; Chumachenko, V. A. Progress in reverse-process application to catalytic incineration problems. *Chem. Eng. Prog.* **1993**, *32*, 89.
 Neophytides, S. G.; Froment, G. F. A bench scale study of reversed flow methanol synthesis. *Ind. Eng. Chem. Res.* **1992**, *31*, 1583.
 Nieken, U. Abluftreinigung in katalytischen Festbettreaktoren bei periodischer Strömungsumkehr. *VDI-fortschrittber., Reihe 3*, **1993**, No. 328.
 Westerterp, K. R.; Fontein, H. J.; Beckum, F. P. H. Decoking of a fixed bed of catalyst. *Chem. Eng. Technol.* **1988**, *11*, 367.
 Züfle, H. Betriebsverhalten eines Festbettreaktors mit periodischer Strömungsumkehr. Dissertation, Universität Karlsruhe, Germany, 1993.

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