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Design of an Catalytic Oxidation Unit for Elimination of Volatile Organic Compound and Carbon Monoxide

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ABSTRACT: Treatments of waste gas as well as solid and liquid wastes are important activities for environmental protection. Thermal oxidation is the most used method for volatile organic compound (VOC) abatement from process off-gases. However, the consumption of natural gas is often considerable, and the running costs of such equipment are very high. This is the main reason why companies are searching for technologies that decrease the consumption of natural gas but still meeting current emission limits. In many cases, thermal oxidation is appropriate to replace catalytic oxidation technology. A mathematical model of an ideal plug-flow packed-bed adiabatic reactor for catalytic oxidation of VOC and carbon monoxide was created to support the design of a catalytic oxidation unit (pilot-plant reactor). Industrial catalyst "EnviCat" based on platinum and palladium applied on the Al_2O_3 support was used. In the first proposed mathematical model, the maximum working temperature of the catalyst was exceeded. Thus, the catalytic bed had to be divided into two parts. Cooling air was injected between the two catalytic beds. The catalytic oxidation unit (pilot-plant reactor) was designed according to the results of the mathematical model. The experimental data are in good agreement with the results of the mathematical model, although a very simplified model was used.

1. INTRODUCTION

Recently, the pressure of lowering of emission limits is increasing, especially for volatile organic compounds (VOCs).[1] The greenhouse effect and negative influence on the environment and human organisms are the main reasons. Particular VOCs have dangerous properties, such as irritant, toxic, and dangerous for the environment, and harmful.[2] Emissions of VOCs are produced by chemical industry companies, which treat organic material such as organic solvents, paints, fuels, cleaning compounds, etc. In many cases, industry emissions of VOCs are removed by thermal oxidation at an incinerator unit. The running costs of an incinerator using thermal oxidation are often considerable, and this is the main reason why companies are searching for more modern and cheaper technologies. The technology of catalytic oxidation is one of most developing methods for removal VOCs.[3] Fixed- or packed-bed reactors are the equipment most often used. The catalysts used for catalytic oxidation are, among others, noble metals such as platinum (Pt), palladium (Pd), and rhodium (Rh) or transition-metal oxides such as chromium (Cr) or nickel (Ni).[4−6] Commonly used catalyst supports are Al_2O_3, TiO_2, SiO_2, or zeolites.

1.1. First Approach to Catalytic Reactor Design. The packed-bed reactor was chosen for the design of a pilot-plant reactor. It has been suggested that the reactor will be connected to a branch of the process off-gas pipeline prior to the entrance of the incinerator unit. The process off-gas composition, which was simulated in the *HYSYS* program environment, is stated in Table 1. For the mathematical model, it is assumed that the composition of the process off-gas was stable in time and was calculated with the maximal possible concentrations of VOC and carbon monoxide (CO) of 10000 and 7000 mg/Nm^3, respectively.

The process parameters of the mathematical model were adjusted according to observed real conditions. The operation

Table 1. Simulated Composition of the Process Off-Gas in *HYSYS*

compound	formula	y_i
acrylic acid	$C_3H_4O_2$	0.00072
acetic acid	$C_2H_4O_2$	0.00167
propylene	C_3H_6	0.00136
propane	C_3H_8	0.00143
formaldehyde	CH_2O	0.00356
acrolein	C_3H_4O	0.00041
carbon monoxide	CO	0.00569
oxygen	O_2	0.03410
water	H_2O	0.31116
carbon dioxide	CO_2	0.01392
nitrogen	N_2	0.62598

conditions and parameters of the catalytic and inert beds of the reactor are summarized in Table 2. The design of the catalytic reactor is also based on the operational parameters of the used catalyst EnviCatVOC-5565 listed in Table 2. This catalyst is a mixture of noble metals Pt and Pd on the Al_2O_3 support. This combination is mostly used in the industry for gas cleaning containing CO and VOCs.[4]

For the pilot technology design, these key parameters were used. The volume of the catalyst was calculated by space velocity and gas volumetric flow:

$$V_{cat} = \frac{V_g}{VHSV} = 0.01 \ m^3 \tag{1}$$

Table 2. Operation Conditions and Parameters of the Catalyst and Inert Beds of the Catalytic Reactor

Operation Conditions	
inlet gas temperature, T_0 (°C)	310
inlet gas pressure, p_0 (Pa)	106352
inlet gas volumetric flow, V_g (N m³ h⁻¹)	100
Catalytic Bed Properties	
starting temperature, T_{min} (°C)	290
maximal operational temperature of catalyst, T_{max} (°C)	650
volumetric hourly space velocity for design of catalytic bed, VHSV (h⁻¹)	10000
linear velocity for the design of the catalytic bed, $v_{g,linear}$ (N m s⁻¹)	0.56
catalyst density, ρ_{cat} (kg m⁻³)	600
catalytic bed porosity, ε_{cat}	0.44
hydraulic diameter, $d_{h,cat}$ (m)	0.005
shape	spheres
shape factor	1
Inert Bed Properties	
inert density, ρ_{inert} (kg m⁻³)	700
inert bed porosity, ε_{inert}	0.7
hydraulic diameter, $d_{h,inert}$ (m)	0.0041
shape	Raschig rings
shape factor	0.3

The mass of the catalyst was calculated by the mass density and volume of the catalyst:

$$m_{cat} = V_{cat}\rho_{cat} = 6 \text{ kg} \qquad (2)$$

The surface area of the catalytic bed was calculated by the gas volumetric flow and linear velocity of the gas:

$$A_{cat} = \frac{V_g}{v_{g,linear}} = 0.0496 \text{ m}^2 \qquad (3)$$

The catalytic bed inside the catalytic reactor was square-shaped. The length of the edge of the square surface was 0.23 m. The height of the catalytic bed was calculated as 0.2 m. For the calculation, the starting temperature of the oxidation reaction was 290 °C (for the design, 310 °C was chosen) and the maximal operational temperature of the catalyst was 650 °C (for the design, 600 °C was chosen).

Before construction of the pilot-plant reactor, it was necessary to verify the behavior of the reactor, mainly the VOCs and CO removal efficiency, and to find out whether, during the reaction, the expected operating parameters of the catalyst are met. A mathematical model of an ideal plug-flow packed-bed adiabatic reactor for catalytic oxidation of VOCs and CO was used to support the design of a catalytic oxidation unit (pilot-plant reactor).

2. MATHEMATICAL MODEL OF A PILOT-PLANT REACTOR

2.1. Kinetics. It was necessary to obtain kinetic data of oxidation reactions for optimal setup of the mathematical calculation of the reactor. Kinetic data obtained under similar conditions were found in the literature and are stated in Table 3 and below in particular examples of oxidation.

$$C_3H_8 + 5O_2 \rightarrow 4H_2O + 3CO_2$$
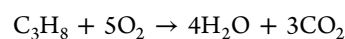
2.1.1. Propane. Garreto et al.[13] examined oxidation of propane on a Pt catalyst on different catalyst supports. He

**Table 3. Activation Energy and Frequency Factors for Individual VOCs and CO**

compounds	formula	E_a (J mol⁻¹)	A (mol s⁻¹ kg$_{cat}$⁻¹ Pa⁻¹)
propane	C_3H_8	74300[a]	0.2[a]
propylene	C_3H_6	65700[a]	0.45[a]
formaldehyde	CH_2O	57050[b]	0.45[a]
acrylic acid	$C_3H_4O_2$	65700[a]	0.45[a]
acetic acid	$C_2H_4O_2$	65700[a]	0.45[a]
acrolein	C_3H_4O	65700[a]	0.45[a]
carbon monoxide	CO	48950[c]	0.388[c]

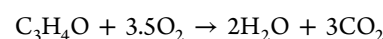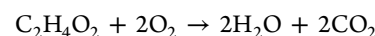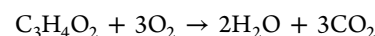[a]Data from refs 7 and 8. [b]Data from refs 9 and 10. [c]Data from refs 11 and 12.

discovered that the activation energy of oxidation is lower with an Al_2O_3 support than with a zeolite support. Despite of reduction of the activation energy on an Al_2O_3 support, the propane reached 80% conversion at temperatures above 450 °C in the catalytic bed.[14] The oxidation of propane is very slow; it starts at high temperatures above 400 °C, and 100% conversion of propane can be achieved at temperatures above 550 °C.[15]

$$C_3H_6 + 4.5O_2 \rightarrow 3H_2O + 3CO_2$$
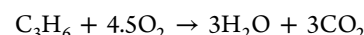
2.1.2. Propylene. Propylene can be a product of the partial oxidation reaction of propane;[16] thus, oxidation conversion of propylene is higher at the same temperatures than that of propane.

$$CH_2O + O_2 \rightarrow H_2O + CO_2$$
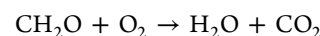
2.1.3. Formaldehyde. Formaldehyde (opposite to propane) starts oxidizing on a Pt catalyst at room temperature, and 100% conversion is reached at temperatures above 100 °C.[9,10] The same frequency factor was used as in the case of propylene.

$$C_3H_4O_2 + 3O_2 \rightarrow 2H_2O + 3CO_2$$

$$C_2H_4O_2 + 2O_2 \rightarrow 2H_2O + 2CO_2$$

$$C_3H_4O + 3.5O_2 \rightarrow 2H_2O + 3CO_2$$

2.1.4. Organic Acid and Acrolein. Kinetic data of organic acids and acrolein oxidation have not been published in the literature, but they were counted with the fact that organic acids and acrolein are products of the oxidation reaction of propylene.[16] Hence, we supposed that the activation energies of organic acids and acrolein can use the same values as those of propylene (Table 3). There is no reason to assume that these organic acids and acrolein will react as quickly as CO or formaldehyde or that they will react as slowly as propane.

$$CO + 0.5O_2 \rightarrow CO_2$$
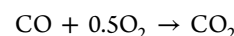
2.1.5. CO. CO is another compound contained in the process off-gas. CO is oxidized at low temperature of around 200 °C on a Pt catalyst with an Al_2O_3 support. In addition, the activation energy is low, and catalytic oxidation is very quick.[11] The lower concentration of CO is better for VOC oxidation, because CO is an inhibitor of VOC oxidation reactions.[17]

2.2. Simplification of the Reaction Rate. The process of oxidation of organic compounds is an irreversible reaction, thus approaching equilibrium can be considered as zero. As a kinetic model for calculation of the reaction rates, the following power law was used:

$$r = kp_{VOC}{}^a p_{O_2}{}^b \qquad (4)$$

where a and b are reaction orders of the oxidation reaction. The presumption of a large excess of oxygen for complete oxidation of VOCs was taken. In such cases, the partial pressure of oxygen is always greater than the partial pressure of the organic substances. The partial pressure of oxygen during the reaction is thus nearly constant. VOC oxidation under these conditions behaves as a first-order reaction. The zero order for a decrease of the oxygen can be used because of the high partial pressure of oxygen.[18,19] The zero order for oxygen was experimentally verified by Garrett et al.[13] for oxidation of propane on a Pt catalyst on an Al_2O_3 support. Because of these assumptions, catalytic oxidation of CO and VOCs can be described as the monomolecular irreversible reaction of one component. This assumption is correct for the oxidation process of the residual organic compounds, which are present in small amounts in the waste gas and if there is an excess of oxygen (O_2) for oxidation reactions. The reaction rate from eq 5 is simplified to the form

$$r_i = k_i y_i p \tag{5}$$

where k_i is the reaction rate constant

$$k_i = A_i \exp\left(\frac{E_{a,i}}{RT}\right) \tag{6}$$

and p is the current pressure in the reactor (initial pressure minus the pressure drop in a given place of the reactor).

2.3. Reactor Model. An adiabatic reactor model with plug flow was used for the mathematical calculation of the above-described processes. The behavior in the reactor was determined by calculation of the mass and energy balances, pressure drop, and axial dispersion. The energy losses in the model were neglected.

2.3.1. Mass Balance. The mass balance is formulated as a change of the molar flow (dF) of component i for passing through a catalytic bed (dW):

$$\frac{dF_i}{dW} = r_i \tag{7}$$

The initial conditions of the molar flows of reaction products were $F_{CO_2}(W{=}0) = 0$ mol s^{-1} and $F_{H_2O}(W{=}0) = 0$ mol s^{-1}. The molar flows of CO_2 and H_2O on the inlet were counted as inerts.

2.3.2. Energy Balance. A change of the gas temperature (dT) for passing through the catalytic bed (dW) was derived from the enthalpy balance:

$$\frac{dT}{dW} = \frac{\sum\left[-\Delta H_{r,i}(T)\,(-r_i)\right]}{\sum F_i c_{p,i}} \tag{8}$$

The initial temperature condition was $T(W{=}0) = 583.15$ K.

2.3.3. Pressure Drop. The pressure drop (dp) increased by passing through the catalytic bed (dW). The pressure drop was calculated by the Leva equation:[20]

$$\frac{dp}{dW} = \frac{2f_m v_g^2 \rho_g L_r (1-\varepsilon)^{3-N}}{d_p \Phi^{3-N} \varepsilon^3} \tag{9}$$

f_m and N were determined by a graphical dependence on the Reynolds number.[20] The initial condition of the pressure drop was $dp(W{=}0) = 0$ Pa.

2.3.4. Axial Dispersion: Deviation from Ideal Plug Flow. The presence of the axial dispersion was determined with a

dispersion coefficient D_a, which was obtained from a graphical dependence of the dimensionless numbers Re_m and Sc:[21]

$$Re_m = \frac{d_p \rho_g v_g}{\eta_g} \tag{10}$$

$$Sc = \frac{\eta_g}{\rho_g D_g} \tag{11}$$

The density of the gas was calculated by the ideal gas equation

$$\rho_g = \frac{p \sum M_i y_i}{RT} \tag{12}$$

The dynamic viscosity of the gas was calculated by Chapman−Enskog theory with Lennard-Jones parameters:[22]

$$\eta_g = 2.6693 \times 10^{-5} \frac{(T \sum M_i y_i)^{0.5}}{\sigma^2 \Omega} \tag{13}$$

The diffusion coefficient of the gas was calculated by the Chapman−Enskog equation:[23]

$$D_g = \frac{1.86 \times 10^{-7} T^{1.5} \left(\frac{1}{\sum M_i y_i}\right)^{0.5}}{p \sigma^2 \Omega} \tag{14}$$

The inverse value of the Peclet number indicated the size of the axial dispersion in the reactor. The Peclet number was calculated by the dispersion coefficient:[24]

$$\frac{1}{Pe} = \frac{D_a}{v_g L_r} \tag{15}$$

The ordinary differential equations, reaction rates, and axial dispersion were solved by numerical integration for catalyst weight $W = 6$ kg, with a step of 0.1 kg, by *MATLAB*.

3. DESIGN STUDY

Computation of the mathematical model of the catalytic reactor was performed by parameters that were written herein before. The results of computation, i.e., conversion of compounds and temperature profile all over the length of the catalytic bed, can be seen in Figure 1. The value of the maximal operational temperature of the catalyst was exceeded ($t > 650$ °C) in the first mathematical model. This was the reason for reaction mixture cooling. Cooling of the catalyst can be provided in many ways. The simple and cheap method is cooling by air. The advantages of this method are energy savings and the fact that air brings another amount of oxygen for the oxidation reactions. Thus, the catalytic bed was divided into two parts in a ratio of 1:3. Cooling air with temperature 20 °C and pressure 1 bar was fed between the two catalytic beds, as can be seen in Figure 2.

3.1. Mixing with Fresh Cooling Air. The gas composition and temperature were calculated by the enthalpy and mass balances of the reaction mixture with fresh (cooling) air. The result of iteration was the following: the fresh (cooling) air must be added in a ratio of 1:20 to inlet gas volumetric flow to decrease the outlet temperature to a value of 600 °C and simultaneously reach minimal 98% conversion of VOCs and CO. A temperature of 600 °C was chosen as a safety threshold
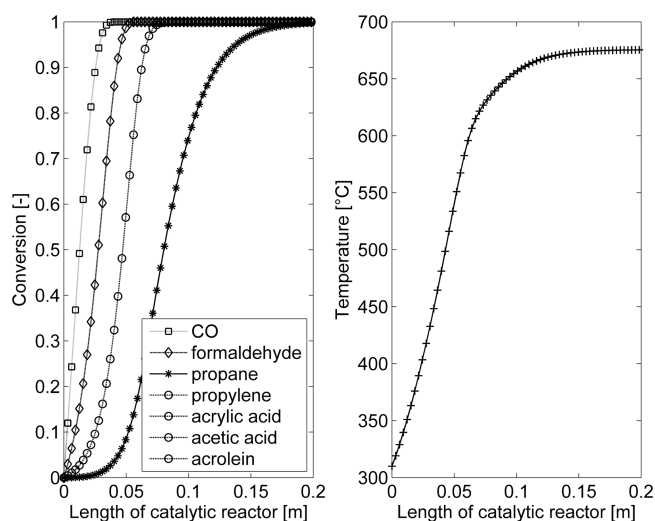
Figure 1. Conversion of VOCs and CO and a change of the temperature in the reactor with one catalytic bed (model conditions).
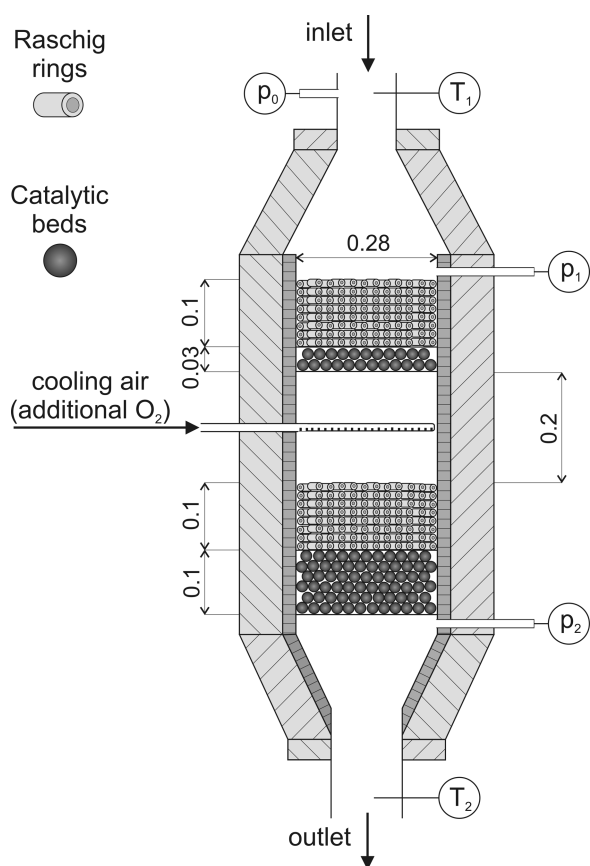
Figure 2. Scheme of the catalytic oxidation unit (pilot-plant reactor). Dimensions are in meters.

because of oscillations of the VOC concentration in the inlet, and this prohibits an increase in the temperature above 650 °C.

4. EXPERIMENTAL PARTS

4.1. Catalytic Oxidation Unit (Pilot-Plant Reactor). The pilot-plant reactor of catalytic oxidation was built in terms of the knowledge stated above. The scheme of the catalytic oxidation unit (pilot-plant reactor) is shown in Figure 2. The pilot-plant reactor was connected to a pipeline with the process

off-gases on the way from the technology to thermal combustion chamber. An expected value of the inlet gas volumetric flow to the reactor was approximately 100 N m^3 h^{-1}. The inlet temperature of the process off-gas (inlet temperature into the reactor) was around 310 °C. The starting temperature of catalytic oxidation on "EnviCat" is 290 °C, so the process off-gas has propriety temperature. The diameter of the inlet pipeline used was DN80 with regard to the inlet gas volumetric flow and inlet temperature. The maximal operational temperature is 650 °C, but for safety reasons, 600 °C was chosen. The diameter of the outlet pipeline used was DN100 with regard to the outlet gas volumetric flow and outlet temperature. The diameter of the cooling air pipeline was DN15.

Sibral heat isolation with 0.03 m thickness was put inside the reactor. Thermal isolation, mineral wool, with 0.1 m thickness was put on the shell. The internal size for the catalytic bed was 0.28 × 0.28 m. The height of the catalytic bed (weight of catalyst, W = 6 kg) was recalculated to 0.13 m. This height was divided into two parts with a ratio of 1:3 (the height of the first bed was 0.03 m; the height of the second bed was 0.1 m). The inert layers of Raschig rings with the height of bed 0.1 m were loaded prior to the catalytic beds. The size of the ceramic Raschig rings was 15 × 15 × 2 mm. The particles of the catalyst have spherical shape with diameters in the range 4−6 mm. The volumetric flow of cooling air was used in the ratio of 1:20 compared to inlet gas volumetric flow; it was predicted by the mathematic model. Feedback control on the outlet temperature (T_2) was installed for safety reasons.

4.2. Measurement and Data Treatment. The inlet (T_1) and outlet (T_2) temperatures of the pilot-plant reactor were measured in an online regime by Pt thermocouples. The pressure drop was measured in an online regime by pressure sensors (p_1 and p_2). The inlet pressure of the gas (p_0) was measured in an online regime by a pressure sensor. The gas volumetric flow was measured by an orifice plate with a pressure sensor.

The concentrations of VOCs in the process off-gas on the inlet and outlet of the reactor were measured by a flame ionization detection analyzer with a range of 0−10000 ppm CH$_4$. The sample of off-gas was taken through a heated Teflon pipeline.

The concentrations of CO, CO$_2$, and O$_2$ in the process off-gas on the inlet and outlet of the reactor were measured by an IR analyzer. The sample of off-gas was taken through a heated Teflon pipeline to a condensation unit with a filter to separate water and solids.

4.3. Experimental Check of the Function. The gas volumetric flow in the pilot-plant reactor was 103.5 ± 14.0 N m^3 h^{-1}. The temperature on the inlet (T_1) was 310 ± 0.4 °C, and the pressure on the inlet (p_0) was 101860 ± 51.4 Pa. The concentration of the VOC in the process off-gas was 8642 ± 270 mg$_{TOC}$ N^{-1} m^{-3}. The volumetric flow of cooling air was set to 6.2 N m^3 h^{-1} (with regard to a ratio of 1:20). The oscillating values of off-gas volumetric flow and concentration of the VOC were a technological problem in the plant. All variables were recorded with 30 Hz frequency. The final value was determined as the median from the recorded data, and the relative deviation from the median was calculated. The experimental measurement lasted for 60 min in steady state. The VOC concentration of the process off-gas during experimental measurement was similar to that in Table 1, only with little correction to 8642 mg$_{TOC}$ N^{-1} m^{-3} VOC.

5. RESULTS AND DISCUSSION

The catalytic oxidation unit (pilot-plant reactor) was built according to the results from the mathematical model of the reactor. The calculated conversion of VOC and CO and temperature trend through the reactor with two catalytic beds under real conditions can be seen in Figure 3. The catalytic
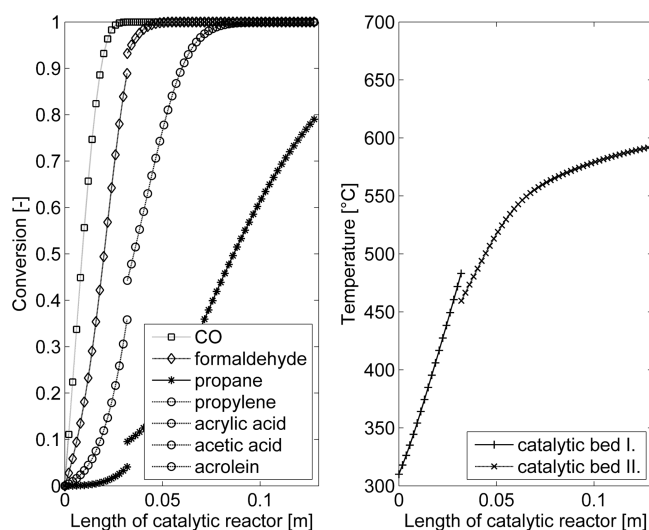
Figure 3. Conversion of VOC and CO and change of temperature in the reactor with two catalytic beds (real conditions).

oxidation of the process off-gas with the content of VOC and CO was performed in this pilot-plant reactor. A comparison of the results from the mathematical model with the experimental results can be seen in Table 4.

Table 4. Comparison of the Results from the Mathematical Model with the Experimental Results from the Pilot-Plant Reactor

variable	model	experiment	absolute difference (%)
outlet temperature (T_1, °C)	589.3	557.4 ± 3.3	5.7
pressure drop ($p_2 - p_1$, Pa)	389	362 ± 31	7.5
conversion of VOC (%)	96.4	98.0 ± 0.2	1.6
conversion of CO (%)	100.0	99.1 ± 0.1	0.9

The experimental results are similar to the results of the mathematical model. The temperature on the outlet of the reactor was lower than that in the mathematical model. The theoretical temperature was overestimated, but this error has a positive impact because it is beneficial for the safety of the catalyst. It is supposed that the difference between the theoretical and experimental pressure drop is given by use of the Leva equation (9), where the porosity played the main role for the calculation. The small deviation from the theoretical porosity has a big influence on the calculated pressure drop. The final conversion of VOC and CO in the experiment is nearly the same as that in the mathematical model. Obviously, the real process of VOC and CO conversion in the catalytic bed can be different because of the kinetic data for organic acids and acrolein were simplified and because of the fact that the laboratory kinetic data measured on the used catalyst were not collected and used. Although a simplified mathematical model was used for computation, the error of the results was at most

10% (this was an acceptable result for the design). The dispersion coefficient and Peclet number were calculated in the mathematical model and used to find out whether the axial dispersion is presented. The axial dispersion in the catalytic reactor was in range of 0.07−0.2. These values showed the medium-to-high amount of the axial dispersion presented in the reactor. A considerable deviation of behavior from an ideal plug-flow model in the reactor is expected. This deviation has an influence on process parameters such as conversion of VOC and CO, reaction temperature, and pressure drop.

6. CONCLUSIONS

On the basis of the concepts of VOC oxidation chemical reactions obtained from the available kinetic data, a design calculation of the catalytic reactor was performed. On the basis of this design calculation, a real catalytic oxidation pilot unit in the company Momentive Specialty Chemicals Inc. was implemented. The catalytic oxidation unit (pilot-plant reactor) was designed for the catalytic oxidation of CO and VOC from process off-gas as a possible technology replacement for the current incinerator unit. Efficiencies of the catalytic pilot unit of 98.0% for VOC and 99.1% for CO were reached. The amounts of VOC and CO on the output from the catalytic unit were in agreement with the emission limits according to valid legislation.

Experimentally identified relevant data on the maximum temperature in the catalyst bed and conversion in the catalytic bed are in good agreement with the calculated values in the mathematical model with a maximal relative deviation of 10%. Although the model includes many simplifications (simple kinetics without exact data of all compounds or neglected energy losses), it turns out that this mathematical model is a good tool for future design calculations of pilot-plant reactors of this type.

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Notes
The authors declare no competing financial interest.

■ NOMENCLATURE

A frequency factor (mol s^{-1} kg$_{cat}$$^{-1}$ Pa^{-1})
c_p heat capacity (J mol^{-1} K^{-1})
D_a dispersion coefficient (m^2 s^{-1})
D_g gas diffusion coefficient (m^2 s^{-1})
d_p hydraulic diameter of the particle (m)
F molar flow (mol s^{-1})
f_m frictional coefficient
k reaction rate constant (mol s^{-1} kg$_{cat}$$^{-1}$ Pa^{-1})
L_r length of the catalytic bed (m)
p current pressure (initial pressure with a pressure drop) (Pa)

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p_0 initial pressure (Pa)
Pe Peclet number
r reaction rate (mol s^{-1} kg$_{cat}$$^{-1}$)
Re_m Reynolds number
Sc Schmidt number
T temperature (K)
v_g gas velocity (m s^{-1})
W weight of the catalyst (kg)
y molar fraction
ΔH_r reaction enthalpy (J mol^{-1})
ε porosity
η_g gas viscosity (Pa s)
ρ_g gas density (kg m^{-3})
Φ shape factor
DN diameter nominal
TOC total organic carbon
VOC volatile organic compound

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