

# Optimization of structured catalyst carriers for VOC combustion

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## Abstract

Typical catalytic afterburners do not offer sufficient mass transport properties for volatile organic compounds (VOC) combustion. Therefore, displaying highly increased transport properties, short-channel microstructures of various geometries and wire gauze structures have been designed and evaluated as an alternative to monoliths. The structures have been deposited with Pd promoted cobalt oxide catalyst using Langmuir–Blodgett technique. The catalyst has proved to meet the specific requirements of VOC combustions and also not to change the microstructure geometry. According to the models of mass transfer proposed in this study, short-channel and gauze structures have up to 15 times higher mass transfer coefficients, and up to 90 times higher volumetric coefficients than standard monoliths. The results of experiments performed using the stacked wire gauze structure were in accordance with the results derived from modelling giving high convergence for higher Reynolds number values ( $> 15$ ). According to the evaluation criteria proposed by us, the majority of short-channel microstructures have shown very high mass transfer efficiency both in diffusional regime (40 times higher than monoliths) and in levelled diffusional and kinetic regime (over 20 times higher).

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## 1. Introduction

A serious environmental problem of automotive CO/NO<sub>x</sub> emission has been successfully solved for past two decades. Since then, catalytic afterburners based on ceramic monoliths of long strait channels with three-way noble metal catalyst have become a worldwide standard [1]. Their outstanding performance is due to high specific area (more than 4000 m<sup>2</sup>/m<sup>3</sup>), low pressure drop, especially comparing to packed beds, and finally, excellent mechanical and thermal resistances. The catalyst composition and deposition techniques on ceramic supports practically leave nothing to be desired.

In this study, we have focused on another vital environmental problem—emission of volatile organic compounds (VOC). Their monitoring and removal entails large financial loss in a great number of countries all over the world. For example, within urban areas of the USA, benzene

exceeded its allowable concentration in ambient air by 16 times, while in the most occupied areas—by 1200 times, according to the standards given by US Environmental Protection Agency [2]. There, a total yearly cost of VOC monitoring and controlling reaches 4 billion USD [3]. In Poland, the overall emission of VOC (excluding methane) exceeded 900 thousands tonnes in 2000 [4], whereas in other countries, the main sources of VOC are road transport, solvent use, oil industry and agriculture.

A few techniques are available to reduce or prevent VOC emission, all of them having certain range of applicability (types of compounds, their concentration). These are:

- low-temperature condensation—limited mainly to solvents, energetically costly [5];
- biochemical methods—selective and concentration sensitive [7];
- adsorption based techniques—for highly diluted VOC [6];
- non-catalytic combustion—high concentration of VOC in end-pipe streams; and finally
- catalytic combustion—suitable especially for highly diluted VOC.

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## Nomenclature

$a$	specific surface area ( $\text{m}^2/\text{m}^3$ )
$b$	structure characteristic diameter (m)
cpsi	number of channels per square inch
$C_A$	concentration of reagent A ( $\text{kmol}/\text{m}^3$ )
$d$	wire diameter of gauze (m)
$d_h$	channel hydraulic diameter (m)
$D_A$	kinetic diffusivity ( $\text{m}^2/\text{s}$ )
$E$	activation energy (kJ/mol)
$k_C$	mass transfer coefficient (m/s)
$k_r$	kinetic rate constant (m/s)
$k_\infty$	pre-exponential factor of Arrhenius equation (m/s)
$L$	reactor length (m)
$R$	gas constant ( $8.314 \text{ J}/(\text{mol K})$ )
$Re$	Reynolds number for short-channel structures ( $Re = w_0 \rho d_h / \varepsilon \eta$ )
$Re_d$	Reynolds number for wire gauzes ( $Re = w_0 \rho d / \eta$ )
$Sh$	Sherwood number, $Sh = k_C d_h / D_A$ (short-channel structures), $Sh = k_C d / D_A$ (gauzes)
$Sc$	Schmidt number
$t$	temperature ( $^\circ\text{C}$ )
$T$	temperature (K)
$w_0$	fluid superficial velocity (m/s)
$x$	axial co-ordinate (m)

## Greek letters

$\chi$	mass efficiency of structure
$\varepsilon$	void fraction
$\eta$	dynamic viscosity (Pa s)
$\rho$	gas density ( $\text{kg}/\text{m}^3$ )

## Subscripts

1, 2	reactor inlet or outlet, respectively
D	diffusional limitation
m	monolith
S	catalyst surface

Catalytic combustion seems useful for a wide range of various compounds, which are understood as VOC. But indeed, to be efficient in a particular application, the process requires thorough elaboration of both a converter construction and catalyst structure.

Since catalytic oxidation of VOC significantly differs from that of  $\text{CO}/\text{NO}_x$ , the question arises what the specific requirements for VOC combustion are. Firstly, VOC unlike  $\text{CO}/\text{NO}_x$  are highly diluted (up to ppm concentration) in huge streams of end-pipe gases. Under such conditions, catalytic combustion runs in diffusional regime, where reaction rate is limited by the mass transfer between catalyst and flowing gas mixture. This, in turn, results in an undesirable increase in the reactor length (raising investment

costs) and flow resistance (raising pumping costs). From a chemical point of view, VOC pose a collection of organic compounds, mostly of high molecular mass. For this reason, their oxidation under oxygen lean conditions can be accompanied either by the formation of toxic side products, such as dioxins and furans, or the deposition of coke. During VOC combustion, coke except sintering can be a major reason for catalyst deactivation.

To meet the challenge, we decided to develop a novel microstructured reactor able to overcome the drawbacks of monoliths such as low mass transfer inside capillary channels and sensitivity to clogging by coke agglomerates. The study consists of the following parts: (i) designing the reactor microstructure; (ii) development of cobalt oxide catalyst; (iii) modelling the transfer coefficients and experimental verification; (iv) reactor modelling and developing new criteria to evaluate performance of the structures.

## 2. Microstructure concept

Classic monolithic reactors, especially ceramic ones, assure reasonable flow resistance in long strait parallel channels. However, fluid flow in a small and long channel is laminar and well-developed in almost whole channel. Typical monolith channels are 0.8–4 mm in diameter [1] and their length exceeds the entrance length by several times. Molecular diffusion is therefore a predominant mass transport mechanism, which gives rise to low mass transfer coefficients, and finally, brings an undesirable increase in the reactor length. To overcome the above transport limitations, we have proposed two alternative structures: a short-channel structure and a wire gauze structure. The first type of the structures has already been partly considered in other publications by us [8,10]. The geometric parameters of the structures considered in this study are summarized in Table 1.

To place our results in a broader context, in next paragraphs, we consequently compare both types of structures used in this study to monoliths of appropriate cpsi and length: the gauze structures to 200 cpsi and the short-channel structures to 50 cpsi. The parameters of a monolith were chosen in such a way so that they could geometrically correspond to a counterpart structure.

While mass transport is rather poor for the laminar flow in long fluid ducts, it is well known that within the entrance region of the duct, transfer coefficients are significantly higher [9]. Thus, it seems reasonable to enhance mass transport by replacing a single long channel by several short ones. This is an idea of the short-channel structure with enhanced transport presented in this study.

Short-channel structures of various cross-sectional shapes are presented in Fig. 1A–C. Reactor is filled with several stacked structure units separated by gaps to allow good mixing of reagents (Fig. 1E). The dimension (hydrodynamic diameters) of the channels is of the order of ceramic monoliths, from about 0.5–5 mm. What all

Table 1  
Geometric parameters of structures, monoliths and gauzes

Structure	$b$ (mm)	$L$ (mm)	$d_w$ (mm)	$N$ (wires per linear in.)	$a$ (m <sup>-1</sup> )	$\varepsilon$	Remarks
Sine	3	3	–	–	1100	0.835	This work
Triangle	3	3	–	–	1000	0.850	This work
Square	3	3	–	–	667	0.900	This work
Gauze 40 mesh	–	–	0.076	40	4960	0.905	[11]
Gauze 80 mesh	–	–	0.076	80	10100	0.808	[11]
50 cpsi monolith	1.48	250	–	–	917	0.680	[1]
200 cpsi monolith	0.76	250	–	–	1898	0.730	[1]
Gauze 62 mesh	–	–	0.16	61.7	8186	0.673	This work

presented structures have in common is the short length of the ducts, comparable or smaller to the length of simultaneous development of velocity and concentration profiles (so-called entrance length). More details concerning advantages of the short-channel structures can be found in another paper by us [10].

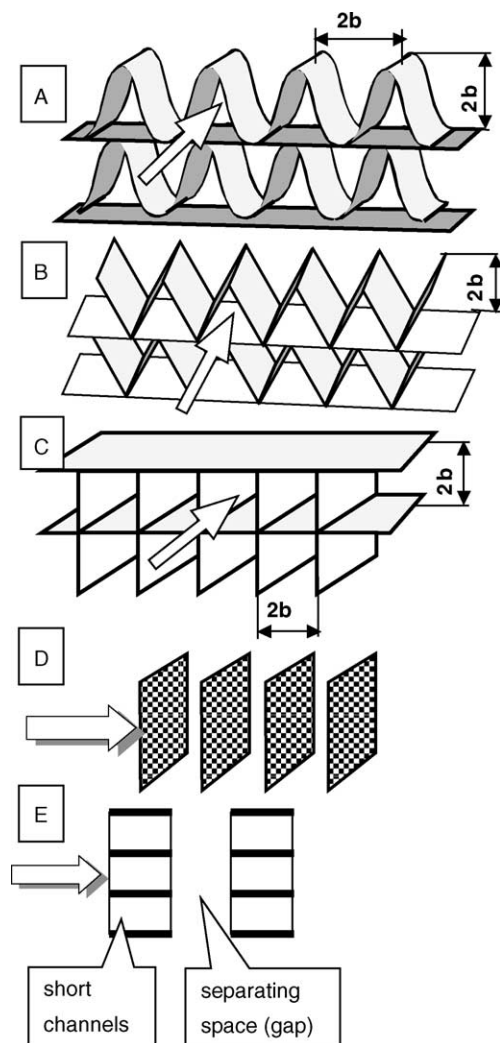


Fig. 1. Designed microstructure units of different cross-sectional shapes: (A) sine, (B) equilateral triangle, (C) square, (D) stacked wire gauzes (gauze structure) and (E) arrangement of subsequent units with a separation gap. White arrows indicate gas flow.

The wire gauze structure, presented in Fig. 1D, is built of single gauzes stacked perpendicularly to the flow direction (forming a gauze package). The design is quite similar to that used in reactors for ammonia oxidation. Combustion of hydrocarbons on gauze-type catalysts has already been studied by Satterfield and Cortez [11] and by Ahlström-Silversand and Odenbrand [12]. A commercial structure of the same kind is Microlith<sup>®</sup> described in ref. [13].

The designed structures have proved to be less sensitive to clogging by coke agglomerates. In a classic monolith (Fig. 2A), coke being formed in a channel can block it and thus eliminate it from the process. Additionally, in the long-channel structure, coke agglomerates are difficult to burn even at elevated temperature, as oxygen molecules migrating through long channels have to cover a long distance to reach them. Unlike in monoliths, in short-channel structures (Fig. 2B) gas can get to the agglomerate through non-blocked channels from both directions (upward and backward), and through the gaps between them. Suppose the deposit was not burnt and stayed in a short channel, only a small part of the reactor becomes dead because the fluid is still able to bypass it.

### 3. Cobalt oxide catalyst

Deposition of catalyst material on metallic surface is another crucial question to be solved, and is probably a reason why, until now, metallic structured carriers have been barely adapted to industrial practice. The problem lays in obtaining catalytic layers of strictly controllable and

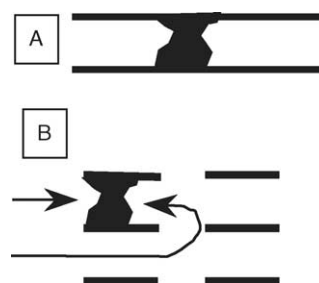


Fig. 2. Coke agglomerates formed in: (A) long channel of classic monolith (difficult oxygen access) and (B) short-channel structure or gauze eyelet (easy oxygen access from both sides indicated by arrows).

uniform thickness, firmly attached to the carrier surface. Such layers should not perturb carefully elaborated geometry of a microstructure. Commonly used methods (deposition from suspension, sol–gel method) are hardly applicable in this case as they give relatively thick layers of difficult to control uniformity and texture (especially at places where wires or sheets are connected). Therefore, more advanced methods should be tested such as plasma spraying, electrochemical deposition or Langmuir–Blodgett technique.

Detailed information on the preparation method of the catalyst and its characterisation can be found in our accompanying paper [14] and only a brief description is presented below.

Talking of surface morphology of the chromium–aluminium steel (CrAl, 20 wt% Cr and 5 wt% Al) used to prepare the structures, upon calcination at high temperature, a uniform and highly dispersed layer of  $\text{Al}_2\text{O}_3$  (2  $\mu\text{m}$  thick, partly containing  $\alpha\text{-Al}_2\text{O}_3$ ) was formed. Such a layer can be regarded as a support or an oxide matrix for catalytic material (washcoat and catalyst). The wash-coat ( $\gamma\text{-Al}_2\text{O}_3$ ) and catalyst ( $\text{Co}_3\text{O}_4$  spinel) were introduced to the surface using Langmuir–Blodgett depositing method [15,16] enabling to control dispersion and amount of the material. After catalyst activation by oxidation (800 °C) some deposited samples were additionally promoted with Pd or Pt using a chemisorption technique. The surface of the samples at all stages of their preparation was examined by SEM, EDX, XPS and AFM techniques. The activated samples were tested in oxidation of *n*-hexane.

The only reaction products of *n*-hexane oxidation using cobalt catalysts both unpromoted and promoted with noble metals were  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . Assuming oxygen excess and a first-order reaction kinetics, the parameters of Arrhenius equation were derived for all the prepared samples [14]. For modelling of the microstructures presented in next paragraphs, two catalyst types were chosen:  $\text{Co}_3\text{O}_4$  catalyst (0.7 wt% related to  $\text{Al}_2\text{O}_3$ , denoted here as Co catalyst) and  $\text{Co}_3\text{O}_4$  catalyst of the same content promoted with Pd (Co/Pd catalyst). It has to be pointed out that the reactive experiments were significantly affected by the diffusional transport of reactants to the catalyst surface. Thus, the derived parameters of Arrhenius equation should be regarded as apparent, and as more qualitative than quantitative:

$$k_r = k_\infty \exp\left(\frac{-E}{RT}\right) \quad (1)$$

The values of activation energy (*E*) were, however, quite reproducible and thus can be treated as a reliable factor describing catalyst activity. The pre-exponential factor ( $k_\infty$ ) showed a significant experimental scatter resulting from a random arrangement of catalyst pieces (CrAl sheets with the catalyst) in the reactor, and thus, an unknown impact of diffusion on the reaction rate. In order to eliminate casualty

from the kinetic results, the values of  $k_\infty$  were averaged for a certain type of catalyst. Thus, a mean  $k_\infty$  value, representing apparent mean catalyst activity, has been taken for further calculations and evaluation of structures efficiency. The Arrhenius equation parameters obtained in this way are the following:

- pre-exponential constant:  $k_\infty = 21.4 \text{ m/s}$  (average for all catalysts);
- activation energy:  $E = 57 \text{ kJ/mol}$  for Co catalyst,  $E = 48 \text{ kJ/mol}$  for Co/Pd catalyst.

Such an endeavour used in our calculations should be treated formally as a tool to compare the structures considered in this study. In the nearest future, to achieve intrinsic kinetics, the experiments are planned in a gradientless reactor to provide absolute kinetic parameters.

#### 4. Mass transfer

By and large, the effective mass transport of reactants to the catalyst surface is the main and common feature of heterogeneous catalytic reactors, and hence also, of reactors for VOC combustion. A reliable estimation of mass transfer coefficients is therefore necessary for further reactor modelling.

At the present stage of our study, the estimation of mass transfer in short-channel structures relies mainly on theoretical calculations, although some experimental work has also been done until now to support them. The literature is an excellent source of the information on heat transfer for laminar flow for various duct shapes and boundary conditions. Theoretical results are confirmed by experimental studies, whose number is, however, on the low side because of experimental difficulties with the so-called entrance region. The differential equations governing mass and heat transfer are analogous, which gives the opportunity to adapt the equations formulated by Shah and London [9] to study mass transport phenomena. The details concerning their estimation and discussion can be found in ref. [10]. In brief, selected results on mass transfer for short-channel structures are presented in Fig. 3 as Reynolds number dependence of volumetric mass transfer coefficient ( $k_{Ca}$ ). The transfer coefficients of the microstructures are related to those of a standard ceramic monolith (50 cpsi, and 250 mm long). According to our estimations, volumetric mass transfer coefficients are up to around ten times higher for the structures designed than for the monolith.

Conversely, as regards wire gauze structures, the literature concerning mass and heat transfer is rather scarce and the results fairly out-dated. An interesting model approach to this kind of microstructure is presented in the study on catalytic combustion of hydrocarbons on Pt gauzes in mass transfer controlling regime by Satterfield and Cortez [11]. In the light of this approach, wire gauze resembles an

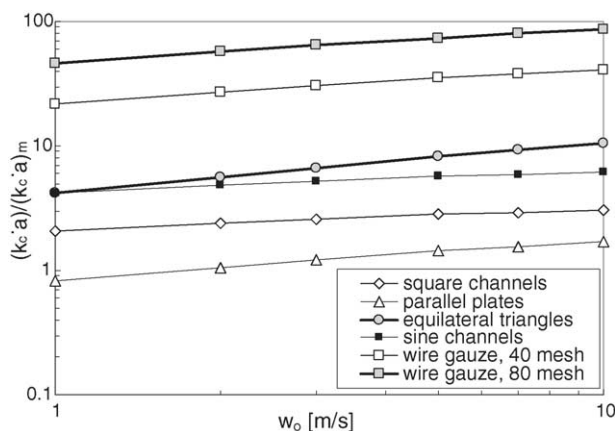


Fig. 3. Superficial gas velocity dependence of volumetric mass transfer coefficients,  $(k_c a)/(k_c a)_m$ , calculated for various short-channel structures (related to 50 cpsi monolith) and gauzes (related to 200 cpsi monolith). Structure description in Table 1.

infinite cylinder, and thus, Reynolds number can be defined using wire diameter. Satterfield and Cortez [11] proved that Colburn analogy applies to derive mass transfer coefficients from the heat transfer and vice versa. They combined their own results with other literature data by: Gay and Moghan [17] (gas-phase mass transfer), Coppage and London [18] (gas-phase heat transfer), Vogtlander and Bakker [19] (liquid-phase mass transfer) and Dixon and Longfield [20] (gas-phase mass transfer). Basing on them, Satterfield and Cortez proposed general correlation equation:

$$Sh = 0.865 Re_d^{0.352} Sc^{1/3} \quad (2)$$

The above equation is a starting point in further modelling in the present study.

As can be inferred from our calculations (Fig. 3), the transfer coefficients for gauzes are the highest among the studied structures; their mass transfer coefficients are up to 15 times higher and the volumetric coefficients up to 90 times than the corresponding values found for the 200 cpsi monolith. In an attempt to verify the theoretical derivations performed for short-channel microstructures and to compensate for the lack of the literature data concerning wire gauzes, the transfer coefficients for the selected structured were also determined experimentally. Taking advantage of the above mentioned analogy between heat and mass transport it was possible to measure exclusively heat transfer to further transform the results into mass representation.

The experiments were carried out in a test reactor of a rectangular cross-section, 45 mm × 30 mm, which was filled with several structures: the short-channel structures and stacked gauzes. Air flow through the reactor was measured using flow-meters. The inlet and outlet air temperature was controlled by thermocouples. Electric current (up to 70 A, carefully stabilised) heated all the

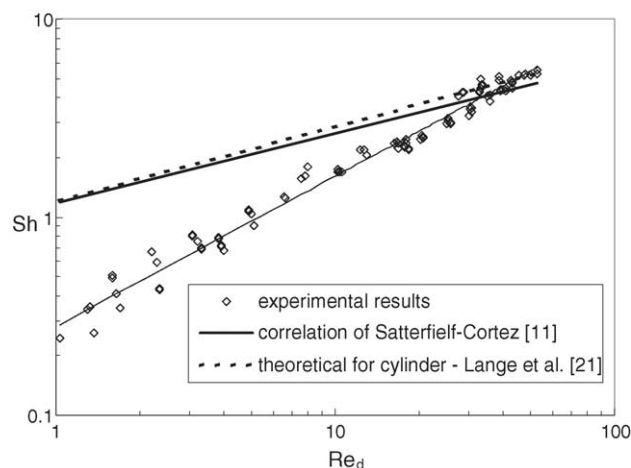


Fig. 4. Mass transfer from experimental results collected for 62 mesh wire gauze (Table 1): Sherwood vs. Reynolds number. Curves represent Satterfield and Cortez correlation [11] and theoretical results for infinite cylinder by Lange et al. [21].

structure units, attaining reasonable heating power, and an appropriate temperature gradient between the structure and flowing gas. Additionally, temperature of each element of the structure was measured with small thermocouples attached to them, and thoroughly electrically insulated although still assuring very good heat conduction. All the devices were connected to computer data acquisition system. During the experiments, typical temperature range of the gas was 30–100 °C and of structure elements 60–150 °C. The temperature difference between gas and structure varied within the range 15–30 °C. The gas Reynolds numbers based on the wire diameter varied within the range from 1.0 to 52.7.

Complete results collected for one package of the wire gauzes (62 mesh, wire diameter of 0.16 mm, 6 gauzes in package) are presented in Fig. 4 together with the fitted correlation curve of the following formulae:

$$Sh = 0.207 Re_d^{0.758} Sc^{1/3} \quad (3)$$

The correlation is based on 121 experimental points and mean deviation amounts to 8.1%. The correlation of Satterfield–Cortez [11] as well as the theoretical results of Lange et al. [21] for an infinite cylinder are also presented in Fig. 4. For higher Reynolds number ( $Re > 15$ ), the results differ from those of Satterfield–Cortez by less than 25%, while for  $Re < 15$  the discrepancy is more significant (even over 50%). For the low Reynolds range, the heat losses attained up to 25% of electric power supplied to the gauzes, which does not seem to be a reason of the observed deviations. Possible deviation sources can be attributed to significant differences in the gauze parameters. For example, the gauzes used in ref. [11] show considerably higher void fraction and twice lower wire diameter than the gauze structure used by us (Table 1). Moreover, the transfer values measured in ref. [11] could also be too high because of a



significant contribution of homogeneous combustion of hydrocarbons to the overall combustion, that occurs at high temperatures in mass transfer controlling regime, especially at low gas velocities (long residence times).

Judging by the increased mass transfer properties, the structures proposed by us seem very competitive to classic monolithic reactors. However, the price of enhanced transport properties is a higher pressure drop due to the momentum, energy and mass transfer analogy. We tackled the problem in our former study [10].

## 5. Reactor modelling

A reliable evaluation of various structures proposed in this study requires formulation of appropriate criteria able to estimate their efficiency from different points of view, or more precisely, considering various regimes of the process. Assuming a steady-state catalytic combustion of highly diluted VOC in a tubular reactor, both reaction heat and heat transfer effects can be neglected. Thus, determining steps can be reduced to the diffusional transport of reactants to catalyst surface and the reaction kinetics. The differential balance of a component A is given by the equation:

$$-w_0 \frac{dC_A}{dx} = k_C a (C_A - C_{AS}) \quad (4)$$

where  $k_C$  is an average mass transfer coefficient (in fact, mass transfer intensity differs along the reactor). Component A, in our case, is simply VOC or a compound or a mixture of compounds chosen as a model VOC in catalytic tests. The mass transfer is balanced by the catalytic reaction expressed as first order kinetic equation:

$$k_C (C_A - C_{AS}) = k_r C_{AS} \quad (5)$$

The integration of the above equations gives the expression which defines mass efficiency,  $\chi$ :

$$\chi = \frac{k_C a L}{(1 + k_C/k_r)w_0} = \ln \frac{C_{A1}}{C_{A2}} \quad (6)$$

Additionally assuming  $k_r \gg k_C$  (diffusional limitation), the equation can be simplified as follows:

$$\chi_D = \frac{k_C a L}{w_0} = \ln \frac{C_{A1}}{C_{A2}} \quad (7)$$

Let us note that the right side of the equations (6) or (7) depends only on the external requirements of the reactor, where  $C_{A1}$  is an initial concentration of the main reactant (typical end-pipe concentration of VOC) and  $C_{A2}$  is a concentration at the reactor outlet (adjusted due to environmental rules). Following this train of thought, a proper reactor design should assure a sufficient value of mass efficiency,  $\chi$ , in order to maintain the assumed level of a pollutant at the outlet.

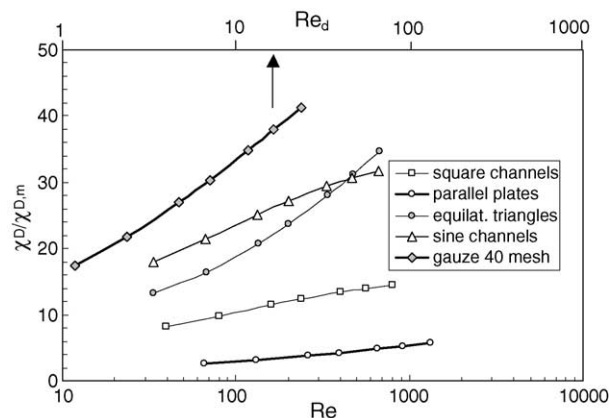


Fig. 5. Reynolds number dependence of mass efficiency: the case of diffusional limitation (equation (7)). The structures description in Table 1. The gauze—upper axis ( $Re_d$ ). Structures related to 50 cpsi monolith, gauzes to 200 cpsi one.

The dependence of the mass efficiency on Reynolds number (the case of diffusional limitation, equation (7)), is presented in Fig. 5. The common trend is that, the gauze structures are the most efficient from all studies structures (20–40 times more efficient in comparison with the 200 cpsi monolith). Moreover, among the short-channel structures the sine and triangular structures in turn show the highest efficiency (15–30 times higher than 50 cpsi monolith).

The efficiencies found for the general case, when both diffusional and kinetic resistances are comparable, are calculated from the equation (6). Three types of reactor internals are compared in Fig. 6: wire gauze 40 mesh, sine short-channel structure and the 200 cpsi monolith. To calculate the efficiency of each structure, the kinetic parameters obtained for the two catalysts described above (Co and Co/Pd) are used. Since the Co/Pd catalyst is more

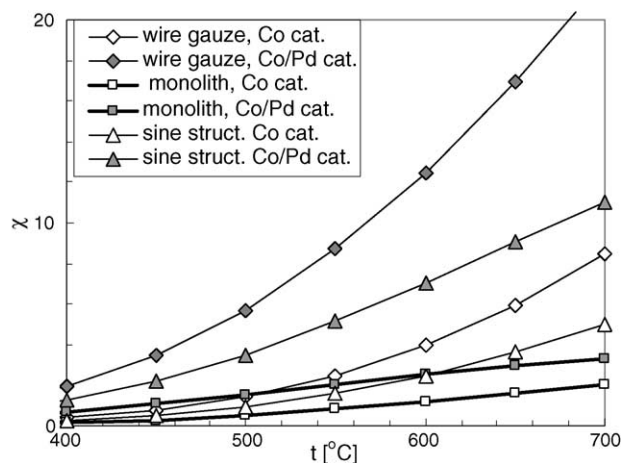


Fig. 6. Temperature dependence of mass efficiency: the general case (equation (6)). Gas velocity  $w_0 = 1$  m/s. Wire gauze 40 mesh, sine structure and 200 cpsi monolith as in Table 1.

active than the Co catalyst, it is clear that it enhances the efficiency of the structures, especially those of better mass transfer properties. At higher temperatures, diffusion starts to play a more important role. The efficiency of the gauze structure with Co/Pd catalyst is almost seven times higher than for the monolith. Above 600 °C, monolith reaches a certain saturation level of efficiency slightly affected by catalyst type while the gauze efficiency continuously increases with temperature.

## 6. Conclusions

The microstructured reactors considered in this study emerge as a powerful tool for the improvement of VOC combustion processes.

- The mass transfer properties for the developed structures are highly increased comparing to classic monoliths. Mass transfer coefficients are up to 15 times higher for gauzes (9 times for short-channel structures), while volumetric mass transfer coefficients are up to 90 times higher for gauzes (10 times for structures). Specific surfaces of short-channel structures are comparable to monoliths, while those of wire gauzes are twice as high.
- The experimental values of transfer coefficients obtained for stacked gauzes are in keeping with the literature data [11] only for higher Reynolds numbers ( $Re > 15$ ). For lower Reynolds number, the results showed a considerable negative deviation.
- The activity of the studied cobalt catalysts is satisfactory taking into account an extremely low amount of the catalysts. The selectivity is excellent as the only products were CO<sub>2</sub> and H<sub>2</sub>O.
- The developed efficiency concept allows comparing different catalytic structures in an easy but comprehensive way. In the case of strong diffusional limitation, the efficiency is around 40 times (gauzes) or 30 times (short-channel structures) higher as compared to standard monoliths. In the general case, when both diffusional and kinetic limitations are leveled, the efficiency is up to 7 times (gauzes) or 3.5 times (structures) higher and strongly dependent on the reaction kinetics.

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