STRUCTURED FIXED-BED ADSORBER BASED ON ZEOLITE/SINTERED METAL FIBRE FOR LOW CONCENTRATION VOC REMOVAL

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A novel adsorber with a structured fixed-bed made of sintered metal fibres (SMF) plates has been developed for purification of low-content VOCs gas-streams. The surface of the metal fibres was coated by a thin, homogeneous MFI-type (ZSM-5, Silicalite-1) zeolite film. The zeolite/SMF composite has been shown as an efficient adsorbent, in which VOCs in low content streams are first concentrated and desorbed in a second step with higher concentrations suitable for oxidation. The advantages of this structured zeolite/SMF composite are a low resistance to internal mass transfer, a relatively low pressure drop and compactness compared to conventional powders or extrudates. Propane was used as a model VOC compound. The isotherms and dynamics of adsorption (breakthrough points) were measured to characterize the properties of the adsorbent. A mathematical model was developed to simulate the experimental data.

Keywords: *structured reactor; zeolite coating; sintered metal fibres; adsorption; VOC; breakthrough.*

INTRODUCTION

The VOC emissions are subjected to legal limits (Ghoshal and Manjare, 2002). The choice of technique for VOC control depends on the type and concentration of VOC. Catalytic oxidation is the method of choice if the concentration of VOC allows performing the process adiabatically. For low VOC concentrations a two-step adsorber-incinerator process can be used (Schoubye, 1980; Kullavanijaya *et al.*, 2000). In this process the VOCs are first concentrated by adsorption at low temperature until the breakthrough occurs. The adsorbent is regenerated by desorption by passing a heated inert gas through the bed. The desorbed concentrated VOC is then passed through an incinerator and converted to harmless compounds by catalytic oxidation. The heat of combustion can be used for desorption.

Propane was chosen as a model compound for this study. The emission limit for propane is 26 ppmv in Germany according to the German clean air act: 'TA-Luft' (Bundesministerium für Umwelt, 2002). Furthermore, the minimum concentration needed to keep the adiabatic flame temperature at 200°C was estimated to be 2600 ppmv. Hence, there is a real need for environmentally friendly processes such as the proposed two-step adsorber-incinerator process to clean up VOC containing effluent gases.

Adsorption of VOCs is normally carried out on activated carbon or zeolites. Activated carbon is cheaper, but zeolites have the advantages of being non-flammable, thermally stable and hydrophobic (Khan and Ghoshal, 2000). Therefore, zeolites are preferred for the treatment of waste gases containing large amounts of water and for the processes with repeated adsorber regeneration by heating.

Adsorbents in randomly packed beds are used in form of pellets. Structured reactors are known to offer open macro-structure leading to low pressure drop during gas-passage, narrow residence-time distribution and enhanced heat- and mass-transfer (Cybulski and Moulijn, 1998; Yuranov *et al.*, 2003).

Structured sintered metal fibre (SMF) plates have already been used for structured catalytic beds due to their homogeneous and open macro structure (70–90% porosity) and low pressure drop (Yuranov *et al.*, 2003, 2005). The metallic fibre matrix of the SMFs has a high thermal conductivity, which ensures a radial heat transfer coefficient in the bed two times higher than the one attainable in randomly packed beds. This avoids 'hot-spots' and run-away problems (Cahela and Tatarchuk, 2001). Furthermore, the SMF offer mechanical and thermal stability and acid resistance for stainless steel fibres (Sterte *et al.*, 2001).

Zeolite interfaces have recently been used in small and micro-scale applications due to their adsorption capacity and catalytic properties (Coronas and Santamaria, 2004) during the removal of VOCs (Shiraishi *et al.*, 2003; Aguado *et al.*, 2004). The composite material used in this

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study is based on SMF filters as supports. The SMF is coated by a thin film of micro-crystallite MFI-type zeolites (silicalite-1, ZSM-5) grown by the 'seed film method' (Hedlund et al., 1997; Yuranov et al., 2005). The homogeneous film of zeolite has low internal mass-transfer resistance compared to conventional pellets due to its short diffusion path. The improved mass-transfer properties lead to a short response time and better control of the desorption.

The aim of this work was to develop a structured adsorber based on the zeolite/SMF composites. Thin MFI-zeolite film on SMF has been characterized by X-ray diffraction, atomic absorption spectrometry, BET specific surface area and scanning electron microscopy. The performance of the material was characterized by the adsorption isotherms of propane, obtained by a dynamic method of adsorption. The breakthrough curves were measured and simulated. Furthermore, the pressure drop as a function of the zeolite film thickness was determined.

METHODS AND MATERIALS

Zeolite Synthesis

The synthesis is based on the 'seed film method' (Hedlund et al., 1997). The SMF was made from stainless steel (AISI 316L), 10–15 μm fibre diameter, and available as panels of 0.28 mm thickness, non-graded, 75–81% porosity, (Southwest Screens & Filters, Belgium). Pieces of SMF (5 cm × 7 cm) were calcined in air at 550°C to clean and create an oxide layer on the metal surface. The calcined support was treated with a 0.4% aqueous solution of the cationic polymer (Poly(dimethylamine-co-epichlorohydrin, 50 wt%, Aldrich) for 20 min, rinsed in 0.1 M NH$_3$ and air dried. Next, the support was treated with a seed solution of anionic, colloidal silicalite-1 crystals for 20 min to adsorb silcalite-1 seeds on the surface, rinsed four times in 0.1 M NH$_3$ with sonification, dried and calcined at 550 C. The seed solution was prepared by a method outlined by (Persson et al., 1994). We used Tetraethoxysilane (TEOS) (>98%, Acros) as silicon source, Tetrapropylamoniumhydroxide (TPAOH) (technical, 20% in H$_2$O, FLUKA) as template in distilled water and ethanol (purum, absolute >99.8% (v/v), FLUKA). The molar composition of the solution was: 100 EtOH: 500 H$_2$O: 25 TEOS: 9 TPAOH. The solution was hydrolysed for 48 h with reflux at 100 C. Afterwards the seeds were rinsed three times after centrifugation and redispersion in distilled water. The final seed concentration was adjusted to approximately 1.8 wt% and pH 10 by adding NH$_3$OH (FLUKA). The seeded SMF panels were placed in a 200 mL PTFE lined autoclave with a synthesis solution and put in an oven at 125 C for 8–24 h. The synthesis solution was prepared from the same chemicals as the seed solution, but in the molar ratio: 2000 H$_2$O: 4 TEOS: 1 TPAOH. Sodium aluminate (Anhydrous, technical, Riedel-de-Haën) was added when synthesizing ZSM-5. This solution was hydrolysed at room temperature by stirring until the solution became clear (~ 2.5 h). After the synthesis, the samples were washed in deionized water for ~20 min and calcined at 550 C. The obtained Na-ZSM-5 was ion-exchanged three times in 0.1 M NH$_4$NO$_3$ at 85 C and calcined at 550 C to obtain H-ZSM-5.

Characterization

Scanning electron microscope (SEM)-photos were recorded using a Philips XL 30 SFEG. The Si/Al ratio of the zeolite film was determined by atomic absorption spectrometry (AAS) on a Shimadzu Atomic Absorption Spectrometer AA-6650 after dissolving the film in an aqueous HF (~1%) solution. A Siemens Kristalloflex 805 X-Ray Diffractometer was used for determining the crystalline phase of the zeolite film and powders. The BET specific surface area (SSA) was determined from the N$_2$ adsorption-desorption at 77 K using a SORPTOMATIC 1990, Carlo Erba Instruments, after pre-treatment at 250°C for 2 h in vacuum.

Adsorption Isotherms

Adsorption isotherms of propane on the SMF filters covered with a MFI-type film were determined from the adsorption capacity obtained by a transient step-up, step-down method. The measurements were made using a Micromeritics AutoChem 2910 connected to a mass spectrometer (Balzers Quadstar 422, Version 6.02). The reactor consisted of a quartz tube, ID = 10 mm, designed for the set-up. The vertical reactor was packed from the bottom (downstream) in a following way: first quartz wool, then 10–20 disks cut from the synthesized SMF panels (diameter 10 mm), quartz wool, ca. 4 cm quartz powder (<450 mm) and the rest (ca. 11 cm) with quartz beads (diameter ~4 mm). The reactor was placed in a heating/cooling bath for temperature control ($\pm$0.05°C). The concentration of propane was varied by diluting the mixture containing 5 or 10% of propane (>99.95%) in Argon (>99.996%) with He (>99.999%). The total volumetric flow for all experiences was in the range of 40–100 mL min^{-1}. All gases were supplied by Carbagas (Switzerland). The desorption was carried out in He at the same flow rates as used for the propane adsorption. Isotherms were constructed by varying temperature and propane concentration. An example of the adsorption cycle measurements is shown in Figure 1. The adsorption capacity measurements consisted of three steps:

(1) *Stabilization*: stabilization of the propane containing gas in the bypass (30–60 min).
(2) *Adsorption*: the propane containing gas with tracer (Ar) is passed through the reactor and adsorption occurs (5–15 min).
(3) *Desorption*: by purging in He for 30–60 min at the same flow rates as used for the adsorption followed by TPD: 5 K min^{-1} in He keeping the flow rate constant.

The amount of propane adsorbed is determined by integrating the area between the curves at the reactor outlet of the tracer (Ar) and the propane in the adsorption part (see Figure 1) and multiplying by the molar flow rate of propane. The tracer does not adsorb on the zeolite and therefore gives the 'empty reactor' response. The amount of desorbed propane is calculated in a similar manner from the step-down response integrating the area between the two curves in the desorption step.
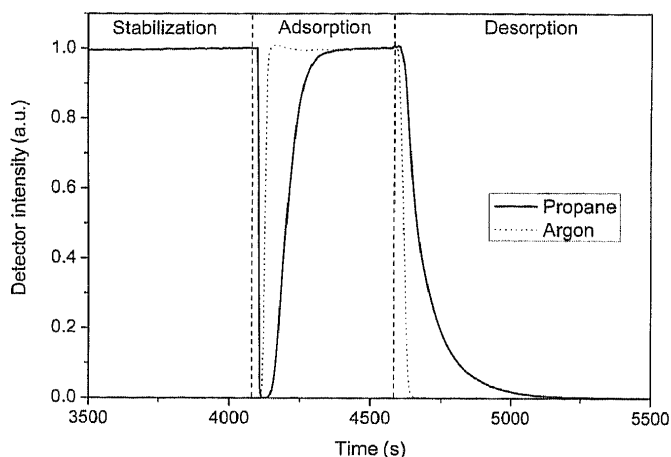
Figure 1. Propane adsorption. Sample: 0.665 g of 5.7% silicalite-1 on SMF at 50.0°C. 2.58% propane in Ar, flow = 19.5 mL (STP) min⁻¹, pressure = 101 kPa.

Breakthrough Curves

Adsorption breakthrough curves of propane through a fixed-bed of zeolite/SMF composites were obtained from dynamic measurements in a tubular pilot reactor (Stainless steel, ID = 19 mm, length 390 mm) at atmospheric pressure reported elsewhere (Yuranov *et al.*, 2003). The SMF filters were cut into disks (diameter = 19 mm) and placed coaxially in the middle of the reactor between two stainless steel tubes (ID = 16.0 mm, OD = 18.9 mm). The inner tubes and filter disks were pressed together by placing rings of PTFE or graphite at the ends sealing the space between the walls of the inner and outer tube to avoid any gas bypass. The space above and below the fixed bed was filled with glass beads. The samples were pretreated at 450°C for 3 h in 67 mL (STP) min⁻¹ Ar flow and cooled down in Ar. 800 ppmv propane in argon was used with a total flow rate of 100 mL (STP) min⁻¹. The measurements were made at atmospheric pressure and 30°C. The concentration was monitored at the outlet using an FID detector in a Perkin Elmer, Auto System XL, Gas Chromatograph.

Adsorption Model

The fixed-bed reactor system was modelled based on a previously proposed compartment model for the adsorption of heavy metals in a fluidized bed (Yang and Renken, 2000). The simplified model in this study uses the tanks-in-series model to describe the hydrodynamics and a lumped mass transfer coefficient for the gas-film diffusion to the solid phase. It is assumed that the adsorption in the solid phase is instantaneous and follows the Langmuir isotherm. Since the experiments in this work were undertaken in the linear part of the Langmuir isotherm a simpler analytical model could have been used (Ruthven, 1984). Nevertheless, we wish to use a general model which is not only restricted to the Henry's law regime. The Langmuir parameters used in the simulations for the adsorption equilibrium, were determined experimentally by the transient response method as described previously.

The material balances in the following are shown for the *j*th tank.

The mass balance for the bulk gas phase for the adsorbate (propane), mixed flow is

$$\frac{dC_j^b}{dt} = N\frac{C_{j-1}^b - C_j^b}{\tau} - \frac{a'k_{lump}}{\varepsilon}(C_j^b - C_j^s) \qquad (1)$$

where k_{lump} is the overall gas-film mass transfer coefficient and N is the number of tanks-in-series. The concentration of adsorbate at the surface of the fibres was calculated from the mass balance at the gas–solid interphase:

$$\varepsilon\frac{dC_j^s}{dt} = a'k_{lump}(C_j^b - C_j^s) - (1 - \varepsilon)\rho\frac{dq_j}{dt} \qquad (2)$$

The accumulation in the solid phase is related to the interphase concentration and derived from the Langmuir isotherm:

$$\frac{dq_j}{dt} = k_f\left(C_j^s(q_m - q_j) - \frac{1}{K}q_j\right) \qquad (3)$$

The initial and boundary conditions are

$$C_j^b = 0, \; C_j^s = 0, \; q_j = 0 \quad \text{at } t = 0 \qquad (4)$$

$$C_{j-1}^b = C_0 \quad \text{for tank } j = 1, t > 0 \qquad (5)$$

The following dimensionless variables are introduced:

$$f = \frac{C}{C_0} \qquad (6)$$

$$\xi = \frac{q}{q_0} \qquad (7)$$

$$\theta = \frac{t}{\tau_m} \qquad (8)$$

$$\tau_m = \frac{mq_0}{QC_0} \qquad (9)$$

τ_m is a residence time based on the dynamic capacity of the adsorbent in equilibrium with the gas concentration C_0. The system in dimensionless variables is as follows:

$$\frac{df_j^b}{d\theta} = \frac{N\tau_m}{\tau}(f_{j-1}^b - f_j^b) - \frac{a'k_{lump}\tau_m}{\varepsilon}(f_j^b - f_j^s) \qquad (10)$$

$$\frac{df_j^s}{d\theta} = \frac{a'k_{lump}\tau_m}{\varepsilon}(f_j^b - f_j^s) - \frac{\tau_m}{\tau}\frac{d\xi_j}{d\theta} \qquad (11)$$

$$\frac{d\xi_j}{d\theta} = \frac{k_f mq_0}{Q}(f_j^s(\xi_m - \xi_j) - (\xi_m - 1)\xi_j) \qquad (12)$$

The parameter τ_m was calculated from the isotherm results of the propane adsorption. The number of tanks, N, and the hydraulic residence time, τ, were found from a fit to the 'empty' reactor response. The only parameter to be fitted in the model for the breakthrough curves was the lumped mass transfer coefficient, k_{lump}. The response at the end of the reactor system is the concentration in the last tank, f_N. Equations (10), (11) and (12) must be solved simultaneously subject to the initial and boundary conditions given by (4) ($f_j^b, f_j^s, q_j = 0$ for $\theta = 0$ ($1 \leq j \leq N$)) and (5) ($f_{j-1}^b = 1$ for $j = 1$ and $\theta > 0$).
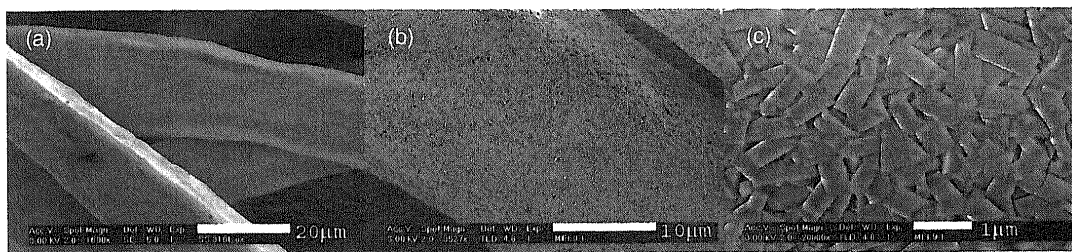
Figure 2. SEM images of the SMF: (a) calcined fibres, (b) and (c) coated by silicalite-1 (loading 9.1 wt%).

The set of 3 times N first order ordinary differential equations (ODEs) was solved using a FORTRAN program. The program uses the IVPAG routine of the IMSL MATH/ library by Visual Numerics, Inc., which uses an algorithm based on the backward differentiation formulas (BDF), also known as Gear's stiff method.

Pressure Drop

The pressure drop was measured in the same tubular reactor as described above for the breakthrough curves. For the pressure drop the reactor space above and below the fixed-bed was left empty. The pressure drop was measured using air and a membrane pump.

RESULTS AND DISCUSSION

Synthesis and Characterization of MFI/SMF

The oxidized SMF surface was modified by a polyelectrolyte to obtain a positive surface charge. The filters were then submerged in a solution of colloidal, silicalite-1 nanocrystals called 'seeds'. Seeds were adsorbed on the support and treated by ultrasonification in 0.1 M ammonia. The film of MFI-type zeolite was grown on the seeded support by hydrothermal synthesis and calcined. The amount of zeolite loading was varied by using different synthesis time or by repeated synthesis. Finally, in the case of ZSM-5 films the obtained Na-ZSM-5 was ion-exchanged and calcined to give H-ZSM-5.

This synthesis method gives a homogeneous film consisting of 1 μm sized prismatic crystallites of MFI zeolite as seen on the SEM images (Figure 2). The difference between oxidized and MFI coated SMF is seen. Homogeneity of the coating was retained for different Si/Al ratios of the synthesis solution. Few cracks in the film were observed after the synthesis. Furthermore, the thermal stability of the film was controlled by SEM after fast heating up to 550 C followed by quick cooling to room temperature. After this treatment a few cracks or holes in the film were observed in the outer regions of the fibre matrix.

XRD patterns were recorded for silicalite-1 and ZSM-5 films and compared with the zeolite powders deposited in the autoclave during the synthesis. The powder spectra were in agreement with the reference spectrum of calcined ZSM-5 (MFI) (Treacy and Higgins, 2001) and the film spectra showed the characteristic peaks ($2\theta = 6-9$ and 22–25) whereas the less intensive peaks disappeared.

A linear correlation was found between the ZSM-5 loading and the BET specific surface area (SSA) of the composite. The SSA calculated for the zeolite was 300–370 m^2 g^{-1} and this material was nanoporous. For MFI-films on SMF from Fecralloy and Inconel alloys (Yuranov *et al.*, 2005) the SSA was found to be 300–320 m^2 g^{-1}. This agrees with the values reported for crystalline ZSM-5 powders: 376–430 m^2 g^{-1} (Narayanan *et al.*, 1996; Seijger *et al.*, 2000; Nijkamp *et al.*, 2001) and that of ZSM-5 crystals supported on metal grids: 302 m^2 g^{-1} (Louis *et al.*, 2001). No effect on the BET surface area was due to the Si/Al-ratio or the film loading.

A significantly higher Si/Al-ratio in the synthesized film compared to the synthesis solution was observed. This indicates the hindering of the isomorphous substitution of Si^{4+} by Al^{3+} in the MFI lattice during the slow controlled growth when applying the seeding method (Ulla *et al.*, 2003).

Adsorption Isotherms

The adsorption of propane was carried out by the dynamic method as described in the experimental part. An example of the isotherms is shown in Figure 3.

The Langmuir model has been fitted to the experimental data using a multivariable, nonlinear least-squares method by minimizing the sum of the squared residuals (SSR) between the model predictions and the experimental data. The variables of the model were: saturation capacity (q_{max}), the pre-exponential factor of the adsorption equilibrium constant (K_0) and the heat of adsorption (ΔH_{ads}^0)
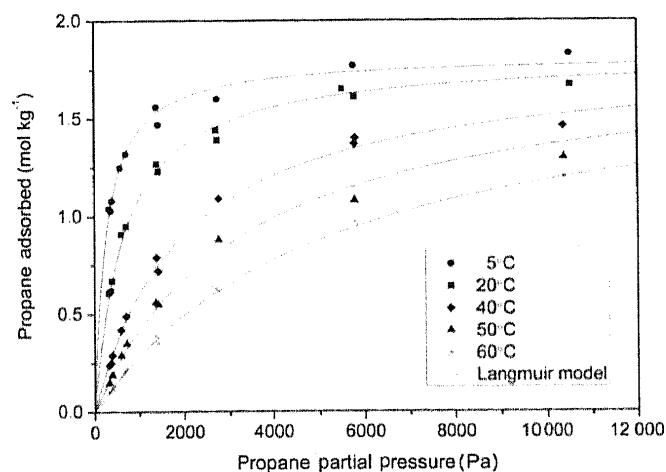

Figure 3. Propane isotherms. Sample: 0.696 g of 10% H-ZSM-5 (Si/Al ~ 200) on SMF.

Table 1. Langmuir model parameters. Si/Al-ratio = ∞ corresponds to silicalite-1; the two other samples are H-ZSM-5.

Si/Al-ratio	∞	~500	~200
q_{max} (mol kg^{-1})	1.85	1.80	1.82
K_0 (Pa^{-1})	3.44×10^{-11}	4.22×10^{-11}	4.11×10^{-11}
ΔH^0_{ads} (kJ mol^{-1})	−43.1	−42.0	−42.5

according to the Langmuir model:

$$q = q_{max} \frac{KP_{propane}}{1 + KP_{propane}}$$

where

$$K = K_0 \exp\left(\frac{-\Delta H^0_{ads}}{RT}\right)$$

The fitted values of the model parameters for three different samples are shown in Table 1.

The results obtained for the three samples of silicalite-1 and H-ZSM-5 with different Si/Al-ratios are very similar. From this data, no trend in the change of adsorption capacity with chemical composition can be concluded. Therefore, the three compounds can be considered to have identical adsorption capacities for propane. The saturation capacity reported is between 1.90 and 2.02 mol kg^{-1}, for pure silicalite-1 in the temperature range 298–308 K (Sun *et al.*, 1998; Zhu *et al.*, 2000a, b), being in the same range as found in the present study.

The heat of adsorption was found to be $\Delta H^0_{ads} = -43$ to −42 kJ mol^{-1}, which is in line with reported data (Hufton and Danner, 1993; Zhu *et al.*, 2000a). The condensation heat for propane in the temperature range of 5–60°C is known to be −12 to −16 kJ mol^{-1} (Majer and Svoboda, 1985). This fact suggests chemical adsorption and not only pore condensation. The isosteric heat of adsorption at zero coverage was found to be identical to the heat of adsorption. Furthermore, the isosteric heat of adsorption as a function of propane loading was constant, indicating

that the synthesized silicalite-1/ZSM-5 layers are energetically homogeneous for the adsorption of propane.

Breakthrough Curves

The breakthrough experiments carried out serve as an example of the dynamics of the novel adsorbent material along with a mathematical model capable of describing the breakthrough curve. The comparison of the simulation results with the experimental data of the breakthrough curves are shown in Figure 4. The hydrodynamic parameters of the model (number of tanks-in-series, N, and the hydraulic residence time, τ) were first fitted to the 'empty' reactor response. The 'empty' reactor was packed in a similar way as the normal reactor except that the SMF filters had no zeolite coating. In other words the 'empty' reactor had no adsorption capacity due to the zeolite coating, but the same hydrodynamics were assumed. The pressure drop over the SMF filters does increase with up to 50% with the increased loading of zeolite. However, the column with the adsorbent was furthermore packed with glass beads, so even though the pressure drop over the active part of the adsorbers was not identical it was negligible in comparison with the rest of the column and the hydrodynamics of the three systems should be nearly identical. The best fit was obtained with $N = 30$ and $\tau = 97$ s. These parameters describe the hydrodynamics and were used for the simulations of the breakthrough. With the adsorbent in the reactor, the breakthrough is seen to be retarded. The sample with the 9.8% loading had approximately two times the mass of silicalite-1 as compared to the sample with 5.1%. In Figure 4(a) the observed breakthrough time approximately doubled. For the longer breakthrough time a broadening of the curve was seen. In Figure 4(b) the breakthrough curves in dimensionless form are seen to nearly overlap. In dimensionless form the F-curves/step-curves for the same hydrodynamics (identical number of tanks-in-series) should be identical, independent on the residence time of the system. Therefore, the increased broadening of the curves in real time is due to the change in adsorption capacity and not a difference in
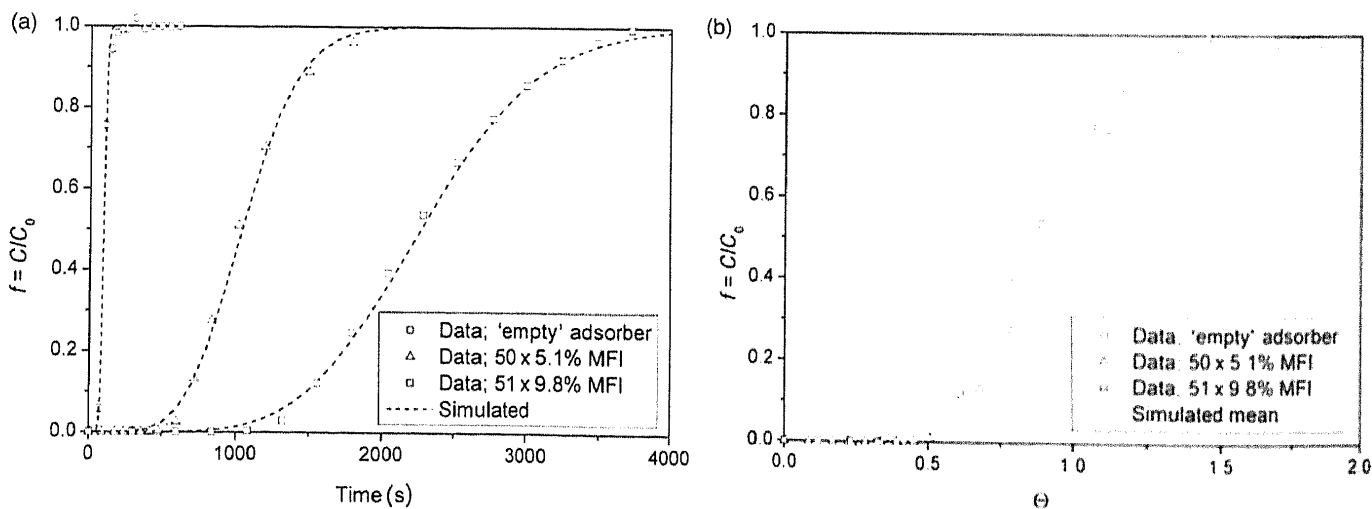

Figure 4. Breakthrough curves obtained in pilot adsorber (a) real time, (b) dimensionless time. Experimental conditions: 800 ppm propane in Ar, 100 mL (STP) min^{-1}, atmospheric pressure and 30°C.
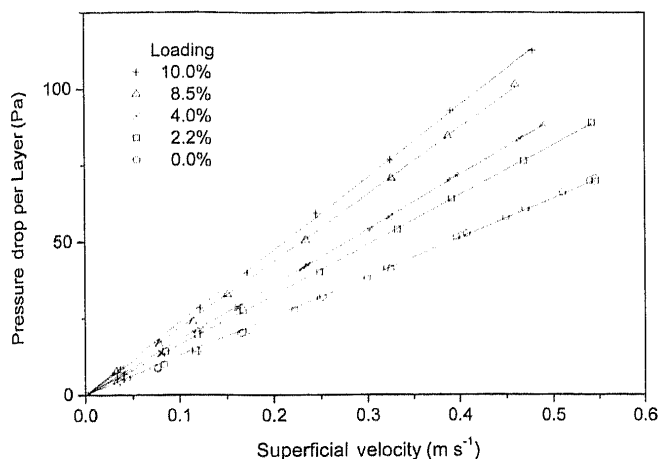
Figure 5. Pressure drop across stacked SMF filters as a function of the gas flow rate.

the internal mass transfer resistance. The best fit of the mass transfer coefficient results in a Damköhler number of $Da\!I = (a'k_{lump}\tau/\varepsilon) \geq 30$ for both loadings. The hydraulic residence time of the active part of the reactor containing the zeolite coated SMF is 1.5 s resulting in a volumetric mass transfer coefficient of $k_{lump}a' \geq 15\,\text{s}^{-1}$.

Since the mass transfer is found to be identical in the two cases, it can be concluded that diffusion resistance in the zeolite film can be neglected.

Pressure Drop

The pressure drop was measured in the laminar flow regime and shows a linear dependence on the flow rate (Figure 5). Darcy's law for the pressure drop in the laminar flow range was fitted to the data by a least-squares method, minimizing the sum of the squared residuals (SSR) between the model and the experimental data:

$$\frac{\Delta P}{\Delta h} = \frac{\mu \cdot Q}{k \cdot A}$$

The variable in the model used was the permeability factor, k. Figure 6 shows the expected falling tendency of
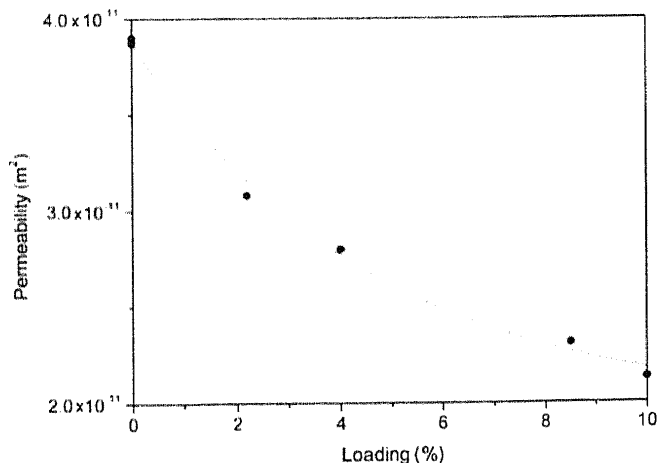

Figure 6. Permeability of SMF filters as a function of the zeolite loading.

the fitted permeability as a function of the zeolite loading on the SMF. Higher loading leads to a lower permeability and a higher pressure drop. Zeolite in the form of powder or particles can also be used in a fixed-bed for VOC adsorption, but the pressure drop will be much higher. We estimated the pressure drop over a fixed-bed of 1 μm spherical particles with the Ergun equation, porosity $\varepsilon = 0.4$. This particle size is in the same order of magnitude as the individual crystals in the zeolite film (Figure 2). When comparing the pressure drop in a randomly packed bed with the same mass of zeolite as on the SMF filters with 10% loading, the pressure drop would be three orders of magnitude higher compared with the structured adsorber. Conventional adsorbents are found in the form of spherical beads of 2–3 mm diameter. It is clear that a zeolite film of 3 μm has very different dynamics than spheres of 2–3 mm diameter. If spheres made entirely out of zeolite were employed the pressure drop would be much smaller, but the dynamics would change the breakthrough curve considerably and the amount of unused zeolite would increase. We have therefore compared the pressure drop in two fixed beds with identical mass of zeolite made up of the SMF based material and another made of 2 mm spheres with a 3 μm, active zeolite shell. In this case the internal mass transfer characteristics would be similar. It is found that the pressure drop of the fixed beds is on the same order of magnitude in the laminar flow regime. The pressure drop is a factor 2 higher in the fixed bed of SMF at a flow rate of 0.5 m s^{-1}. However, the packed bed of spheres is 50 times larger (volumetric basis) than that of SMF.

CONCLUSIONS

A novel, structured adsorber based on SMFs coated by a thin, homogeneous MFI-type (ZSM-5, Silicalite-1) zeolite film has been developed, tested and characterized. The zeolite/SMF composite is an efficient adsorbent for the removal of low concentration VOCs. When VOC breakthrough occurs, the adsorbent can be regenerated by desorbing VOC with the concentration suitable for their catalytic oxidation in adiabatic mode. Heat delivered by oxidation can be used for desorption. The chemical and physical analyses of the synthesized zeolite films correspond well with reported values for MFI zeolites in powder and supported form. The pressure drop across the filters was investigated and shows an increase with increasing loading (as expected). The thin film of zeolite leads to a short diffusion path and lower resistance to internal mass transfer. Breakthrough curves of propane through a fixed-bed of adsorbent were measured and compared with simulated results. The overall volumetric mass transfer coefficient was found to be $k_{lump}a' \geq 15\,\text{s}^{-1}$. The model simulations suggest that the adsorption rate is governed by gas film diffusion at zeolite loadings up to 10% and not by internal diffusion in the zeolite under the experimental conditions used.

NOMENCLATURE

a'	external surface area per unit volume of reactor, $\text{m}^2\,\text{m}^{-3}$
A	surface area, m^2
C	gas concentration, $\text{mol}\,\text{m}^{-3}$

f	dimension less concentration
h	height, m
H	enthalpy, $J\,mol^{-1}$
k	permeability, m^2
k_f	adsorption rate constant, $m^3\,mol^{-1}\,s^{-1}$
k_{lump}	lumped mass transfer coefficient, $m\,s^{-1}$
K	adsorption equilibrium constant, $m^3\,mol^{-1}$
m	mass of adsorbent, kg
N	number of tanks in series
P	pressure, Pa
P_i	partial pressure of species i, Pa
q	concentration of adsorbed species, $mol\,kg^{-1}$
Q	superficial flow rate, $m^3\,s^{-1}$
q_0	maximum adsorption capacity in dynamic equilibrium with C_0, $mol\,kg^{-1}$
q_{max}	maximum adsorption capacity, $mol\,kg^{-1}$

Greek symbols

ε	porosity of the reactor
θ	dimensionless time
μ	viscosity, Pa s
ρ	mass of fibres per unit volume of the reactor, $kg\,m^3$
τ	hydraulic residence time, s
τ_m	time required for saturating all adsorbent, s
ξ	dimensionless concentration of adsorbed species

Sub- and superscripts

b	bulk phase
j	tank j
s	surface (outer surface of the ZSM-5/fibre composite)

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