In situ observation of wall effects in activated carbon filters by x-ray microtomography

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ABSTRACT

X-ray microtomography is a powerful non invasive visualization technique which can be advantageously used to get a better understanding of dynamic adsorption processes. In the present work, this technique is shown to be able to detect wall effects during the dynamic adsorption of methyl iodide on activated carbon filters. The analysis of transversal cross sections along the filter height clearly shows the existence of radial concentration profiles. These radial adsorption profiles are directly linked to velocity profiles due to a higher permeability at the wall. Obtaining such in situ information constitutes a real progress in order to validate simulation models allowing predicting reliable breakthrough times.

Keywords X-ray microtomography, activated carbon, adsorption, wall effect, radial profile

1 INTRODUCTION

Activated carbons are largely used to adsorb organic compounds in industrial processes as well as for civil and military protection. Their main advantages are: high specific areas, high affinity towards organic compounds and low cost. As organic vapours are potentially toxic, a correct estimation of filter breakthrough times is essential. Many authors have developed models to predict breakthrough times [1]. Most of these models are one-dimensional, use an average linear velocity for the gas and neglect radial velocity profiles within the carbon beds. Nevertheless, because of the higher porosity at the wall, the permeability is more important in that region, leading to gas flow through preferential channels [2]. Consequently, as the annular zone close to walls becomes rapidly saturated, a part of the gas stream leaves the filter before elimination of organic compounds and the breakthrough time decreases.

Even though the knowledge and the quantification of wall effects are crucial for a correct filter operation, they remained quite difficult to determine until the development of non intrusive visualisation techniques such as X-ray tomography. This technique has been extensively applied in chemical engineering for the last 10 years in order to determine the phase spatial distribution within large process vessels [3,4]. More recently, the development of microfocus X-ray sources and high resolution CCD detectors led to the commercialisation of microtomographs with improved resolution (around a few microns). These systems, dedicated to small objects (from the mm^3 to few cm^3) opened the way for many applications in domains such as material science [5-8], biomedical science [9,10], and food science [11,12]. In this work, X-ray microtomography is used to evidence the existence of

concentration radial profiles during adsorption of methyl iodide. The use of this non destructive imaging technique to follow a dynamic adsorption process within activated carbon filters has been recently validated [13,14].

The experiments were carried out with CH_3I. The selection of methyl iodide is justified by its low boiling point, 42°C, and by its adsorption behaviour which is the same as its radioactive counterpart [15,16]. Organic iodine is one of the three main radioactive forms that are produced during nuclear incidents, with molecular and particulate iodine (aerosols) [17].

2 MATERIALS AND METHODS

2.1 Activated carbon filters

Polyethylene cylindrical canisters with an internal diameter of 15 mm whose inlet and outlet consist of a metallic grid allowing gas circulation were used. Polyethylene was chosen because of its low X-ray attenuation, allowing obtaining good contrasted images of the carbon bed. The experiments were realised with BPL® 12×30 (Calgon Carbon Corporation) activated carbon the grain diameter of which ranges between 0.6 and 1.7 mm with an average of 1 mm. About 1.6 g of activated carbon were used for each trial, corresponding to an apparent density of 0.45 g/cm³. The adsorption tests were conducted in a classical breakthrough measurement system [18] with CH_3I in dry air at a temperature of 293 K. The gas flowrate was fixed at 5 l/min, producing superficial velocities of 47 cm/s. The filters were analysed by x-ray µ-CT before and after exposure to CH_3I.

Figure 1

2.2 X-ray microtomography

X-ray tomography is a non destructive technique allowing to visualise the internal structure of the investigated objects. Transversal cross sections are reconstructed from radiographs obtained at several angular positions, around 180 or 360°. The grey level of each pixel is related to the local X-ray attenuation. The attenuation mainly depends on the energy of the incident beam, on the material density and on its atomic number [19]. In this study, a "Skyscan-1074 X-ray scanner" (Skyscan, Belgium) was used. The cone beam source operated at 40 kV and 1 mA. The detector was a 2D, 768 pixels × 576 pixels, 8-bit X-ray CCD camera giving images with a pixel size of 41 µm. The rotation step was fixed at the minimum, 0.9°, in order to improve image quality, giving total acquisition times close to 10 min. After the filters were scanned, cross section images (Fig. 2a) separated by 205 µm were reconstructed along the plastic canister using a cone-beam reconstruction software based on Feldkamp's algorithm [20].

2.3 Image analysis

Figure 2

The analysis of cross sections is based on the fact that the grey level of a pixel full of carbon increases when adsorption takes place, due to the condensation of CH_3I within activated carbon pores. This is related to the increase of the local X-ray attenuation coefficient with density (carbon + adsorbed CH_3I). Image processing was realised as follows: a) a circular mask was applied to eliminate the filter walls; b) the average grey level was determined on

the whole cross section; c) the average grey level was determined in concentric rings, obtained by successive erosions of 10 pixels; d) by calibration, the grey level was converted to a percentage of the maximum mass uptake that can be reached under the used experimental conditions. For the sake of illustration, Figure 2b shows how the concentric rings are constructed when 30 erosions are applied between each of them.

3 RESULTS AND DISCUSSION

3.1 Qualitative approach

Figures 3a and 3b show the radiographs of a filter before and after exposure to CH_3I during 4 min. These images are represented in false colours, with level 255 corresponding to non attenuating material and level 0 corresponding to highly attenuating material. With this colour scale, X-ray attenuation increases when going from light blue to yellow. The red colour corresponds to pure activated carbon, while the yellow tones stand for the adsorbed CH_3I. The colour change linked to adsorption is clearly observed on Figure 3b: an axial profile develops within the filter. The adsorption front seems corresponding to plug flow with a weak axial dispersion.

Figure 3

Transversal cross sections corresponding to the filter shown on Figure 3b are represented on Figure 4, with the same colour scale. Three levels were selected, at the bottom, in the middle and at the top of the filter. On these images, the carbon grains appear in light blue. On the radiographs, red colour, i.e. higher attenuation level, was obtained because it corresponded to the superimposition of all the carbon grains located on the X-ray path. On the 2D cross-

sections, the colour of each pixel strictly refers to the local attenuation value. The dark blue zones correspond to CH_3I adsorption and clearly indicate the existence of a radial adsorption profile. At the bottom of the filter, the uniform light blue colour corresponds to virgin carbon. A comparison between cross sections b and c clearly shows that the radial profile is more marked when the section is close to the entrance of the gas flow.

Figure 4

3.2 Quantitative approach

The radial profiles obtained after 4 min. exposure to gas stream are plotted on Figure 5, for increasing depths. One can clearly observe the existence of radial adsorption profiles: the amount of adsorbed CH_3I increases from the centre to the wall. As the column-to-particle diameter ratio is rather low ($\cong$ 15), this kind of results was expected. Nevertheless, in our knowledge, this is the first time that in situ measurements of such profiles are realised using X-ray microtomography. This observation must be put in relation with a higher local permeability at the wall, and, consequently, with higher gas velocities [2,21]. The transport of the pollutant as well as its adsorption is thus favoured at the periphery of the filter. Moreover, the gradients at the wall are more important at the inlet of the filter. This experimental result is in agreement with the simulation of Valdes-Solis et al. [22] who found that the velocity profile, initially parabolic, tended to flatten in the flow direction.

Figure 5

To highlight the interest of X-ray microtomography to study adsorption processes, Figure 6 shows the corresponding axial profile, i.e. after 4 min. exposure to CH_3I. This axial profile can be considered as the mirror of the traditional breakthrough curve obtained by measuring

exit concentration. The same typical sygmoidal shape can be obtained by in situ measurements. The well known mass transfer zone (MTZ) can even be clearly identified on the curve.

Figure 6

4 CONCLUSIONS

In this work, X-ray microtomography was used to visualise wall effects during dynamic adsorption of CH_3I. This non destructive and non invasive technique allowed the quantitative determination of the radial adsorbate concentration profiles at different filter depths. The results show that it is important to take this phenomenon into consideration, especially for modelling purposes. These promising results open the way to more detailed experiments. Future work will focus on the influence of operating conditions (superficial gas velocity, temperature), of adsorbate nature, and of the shape of the adsorbant (grains with controlled granulometry, monolith, …) on preferential flow at the wall. A detailed *in situ* investigation of the effect of the column-to-particle diameter ratio will be particularly interesting. Image processing will be improved in order to characterise the structure of the activated carbon bed, i.e. internal porosity profiles. Finally, these data will allow the validation of simulation results (e.g. obtained by the lattice Boltzmann methodology), taking into account the coupling between gas flow pattern and adsorption.

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