

# Mercury removal from aqueous solution and flue gas by adsorption on activated carbon fibres

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## Abstract

The use of two activated carbon fibres, one laboratorial sample prepared from a commercial acrylic textile fibre and one commercial sample of Kynol<sup>®</sup>, as prepared/received and modified by reaction with powdered sulfur and H<sub>2</sub>S gas in order to increase the sulfur content were studied for the removal of mercury from aqueous solution and from flue gases from a fluidized bed combustor. The sulfur introduced ranged from 1 to 6 wt.% depending on the method used. The most important parameter for the mercury uptake is the type of sulfur introduced rather than the total amount and it was found that the H<sub>2</sub>S treatment of ACF leads to samples with the highest mercury uptake, despite the lower sulfur amount introduced. The modified samples by both methods can remove HgCl<sub>2</sub> from aqueous solutions at pH 6 within the range 290–710 mg/g (ACF) which can be favourably compared with other studies already published. The use of a filter made with an activated carbon fibre modified by powdered sulfur totally removed the mercury species present in the flue gases produced by combustion of fossil fuel.

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## 1. Introduction

Mercury is well known as one of the most toxic pollutants with serious impact in the food chain due to potential bioaccumulation. Mercury and its compounds act as dangerous and insidious poisons and can be adsorbed through the gastrointestinal tract and also through the skin and lungs [1]. After adsorption, mercury circulates in the blood and is stored in the liver, kidneys, brain, spleen and bone which can lead to several health problems such as paralysis, serious intestinal and urinary complications, disfunction of the central nervous system and, in more severe cases of intoxication, death. Soluble compounds of mercury are particularly toxic because their adsorption is very fast. When ingested a dose less than 0.5 g can prove to be fatal.

Mercury is still used worldwide in applications such as barometers, thermometers, pumps, lamps and in many others. Industries mainly responsible for the dispersion of mercury are the chlor-alkali, paint, oil refining, rubber processing and fertilizer industries [2] and also all those that use fossil fuels. This fact and the contamination effects of mercury bioaccumulation in soil and sediments that can persist for decades give relevancy to the studies that involve the removal of this pollutant from the environment, such as the one that we report in this paper.

The increasing necessity for the reduction of mercury discharges leads to the need for research and development of better methodologies that can provide effective and total mercury removal from liquid and gas effluents. In general the currently available technologies include sulphide precipitation, ion exchange, alum and iron coagulation, electro deposition and activated carbon adsorption.

The adsorption of HgCl<sub>2</sub> on activated carbons in the liquid phase may take place by two mechanisms, i.e. adsorption of the HgCl<sub>2</sub> specie and/or reduction to Hg(I) by the surface

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functional groups [3]. This adsorption is dependent on the type of the sulfur and oxygen functional groups and on the porous structure of the adsorbent. On the other hand, the adsorption of mercury from the gas phase is mainly dependent on the elemental sulfur present in the activated carbon, on the material porosity and, to a lesser extent, on the activated carbon organic sulfur content [4].

During the last decades a number of studies concerning mercury adsorption in carbon materials from the liquid phase [2,5,6] and from the gas phase [7,8] have been published. Of particular interest are the papers published about the use of sulfur-impregnated carbons [3,8,9]. However, the development of better materials is still needed. Activated carbon fibres (ACF) are a comparatively modern form of porous carbon material with a number of significant advantages over the more traditional powder or granular forms. These include high surface area and adsorption capacity, as well as very high rates of adsorption from the gas or liquid phase. This high rate of adsorption can be advantageous in some industrial applications, particularly when using the direct injection method, where the contact time is reduced and the adsorbent quickly removed from the flue gas. There are only a few reported studies that use this kind of carbon material for mercury adsorption from the gas phase [4,10,11].

## 2. Experimental

### 2.1. Materials

The precursor used for the production of the laboratorial ACF sample was an acrylic textile fibre provided by Fisipe (Barreiro, Portugal). According to the manufacturer the fibre had been polymerised from acrylonitrile (~90 wt.%) and vinyl acetate (~10 wt.%) monomers. For the production of the ACF about 12 g of fibre and a horizontal tubular furnace were used. Stabilisation of the fibres was carried out by heating to 300 °C at a rate of 1 °C min<sup>-1</sup> under a constant N<sub>2</sub> flow of 85 cm<sup>3</sup> min<sup>-1</sup> and maintaining for 2 h. The fibre was then carbonised by raising the temperature at a rate of 5 °C min<sup>-1</sup> to 800 °C and maintaining at that temperature for 1 h. The carbonisation yield, in relation to the initial mass of unstabilised fibre, was 52 wt.%. Activation was carried out by raising the temperature again by 15 °C min<sup>-1</sup> to 900 °C and then switching to a CO<sub>2</sub> flow of 85 cm<sup>3</sup> min<sup>-1</sup>, maintaining for 5 h in order to obtain 53% burn-off, switching back to the N<sub>2</sub> flow and allowing to cool to below 50 °C before removing the ACF from the furnace and storing in a sealed sample flask. This sample shows basic characteristics, indicated by the point of zero charge value of 9.6 [12], and is designated as FA.

In this work, we also used the commercial ACF-1603-15 activated carbon fibre from Kynol<sup>®</sup> (designated as KACF) produced by physical activation directly from the Kynol Novolid<sup>®</sup> fibre made by polymerization of phenol and formaldehyde. This material has slightly basic properties as indicated by the point of zero charge (pzc) value of 7.3 [13].

### 2.2. Samples modification

#### 2.2.1. Sulfur introduction with S (s)—method A

The mixture of ACF and solid sulfur (1:3) was put into the centre of an electric furnace silica tube in a ceramic boat and heated to 600 and 800 °C under a 1 dm<sup>3</sup> min<sup>-1</sup> helium gas flow. The samples were kept at the final temperature for 1–4 h and cooled to 25 °C and were designated KACFS<sub>x</sub>Y h and FAS<sub>x</sub>Y h,  $x = 6$  or  $8$  for samples treated at 600 and 800 °C  $Y = 1, 2, 3, 4$  for 1, 2, 3, 4 h of thermal treatment.

#### 2.2.2. Sulfur introduction with H<sub>2</sub>S (g)—method B

0.3 g of ACF in a ceramic boat was put into the centre of an electric furnace silica tube and heated to 600 and 800 °C under a 1 dm<sup>3</sup> min<sup>-1</sup> nitrogen gas flow and when the sample reached the target temperature the flowing gas was changed to H<sub>2</sub>S, which was produced by chemical reaction. The samples were kept at the final temperature for 1 h and cooled to 25 °C under a nitrogen atmosphere and were designated KACFH<sub>2</sub>S<sub>x</sub>1 h and FAH<sub>2</sub>S<sub>x</sub>1 h,  $x = 6$  or  $8$  for samples treated at 600 and 800 °C.

### 2.3. Mercury adsorption studies

The samples before and after the sulfur introduction were tested for mercury removal from aqueous solutions by preparing several suspension with 50 mL of 1500 mg L<sup>-1</sup> HgCl<sub>2</sub> solution and 0.1 g of activated carbon fibres at pH 6.0. After 24 h of stirring in a thermostat bath at 25 °C the suspensions were filtered using Whatman#1 and the mercury amount measured by ultraviolet spectrophotometry at 230 nm. Measurement details are given elsewhere [5].

The mercury removal from flue gases originated by the combustion of a fuel prepared from a 50% weight mixture of coal Carbocol, from Colombia, and the commercially available sewage sludge Biogran<sup>®</sup> were performed on an atmospheric fluidized bed combustor (FBC) with 5 m height. An Advanced Hg analyser LECO AMA 254 was used to measure mercury concentration in the samples collected. More details about the FBC, gas sampling set-up and the measurements can be found in Ref. [14].

### 2.4. Physical and chemical characterisation of the activated carbon fibres

Nitrogen adsorption isotherms at 77 K were determined using a CE Instruments Sorptomatic 1990 after outgassing the samples at 400 °C to a residual vacuum of  $5 \times 10^{-6}$  mbar. We must note that these outgassing conditions did not affect the samples composition, as indicated by thermogravimetric analysis (not shown here). Elemental analysis of carbon, hydrogen, sulfur, nitrogen and oxygen was carried out using a Eurovector EuroEA elemental analyser. FT-IR spectra were recorded with a Perkin-Elmer model Paragon 1000PC spectrophotometer, using the KBr disc method, resolution of 4 cm<sup>-1</sup> and 100 scans.

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