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## Evaluation of the Variables that Influence Mercury Capture in Solid Sorbents

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### ABSTRACT

Newly obtained results on mercury retention in fly ashes and activated carbons during coal combustion are compared with previous findings. The influence of different variables on mercury capture, including the composition and nature of the sorbents, the composition of the flue gas and the mercury species in gas phase is investigated.

Coal-fired utility boilers are currently the largest single-known source of mercury emissions. According to the USEPA's Information Collection Request (ICR), on average, only 40% of the mercury entering a coal-fired power plant is captured, while 60% is emitted into the atmosphere. Several solid materials, including activated carbons, are considered to be effective sorbents for Hg control in flue gases from coal combustion. However, more economical alternatives to retain mercury such as the optimization of conditions for improving mercury capture in fly ashes in the installation also need to be studied.

In this work, the mercury retention capacity of different fly ashes was compared with retention by commercial activated carbons in different experimental conditions. As might be expected, the results obtained indicate that the quantity of mercury captured depends on the characteristics of the fly ashes and on the mode of occurrence of mercury in gas phase. The retention of elemental mercury in the fly ashes, unlike activate carbons, was greatly influenced by the gas composition. Although the efficiency of the mercury retention was higher in activated carbons than in fly ashes, some activated carbon showed a lower retention capacity for elemental mercury than some fly ashes in the simulated flue gas.

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### 1. Introduction

Coal fired power plants are one of the main sources of mercury emission to the environment (U.S. EPA, Mercury; European Pollutant Emission Register). Although various control technologies have been investigated, until now no cost-effective or efficient control process has been developed for mercury removal in coal power stations. Sorbents, such as activated carbons, are considered to be effective sorbents for mercury control in flue gases from coal combustion (Felsvang et al., 1994; Ghorishi and Gullett, 1998). However, economical alternatives need to be developed. Growing interest in developing such systems has encouraged several

research groups to determine the maximum level of retention that can be achieved by by-products originated in combustion plants. Fly ashes are made up of fine particles of aluminosilicate glass, quartz, mullite, hematite, ferrite, rutile in various stages of transformation, depending on their origin (Ward and French, 2006). They may also contain a variable, but small, proportion of organic matter (unburned) with a relatively high surface area compared to that of the inorganic fraction.

Several studies have shown that fly ashes are able of capturing some of the volatile species generated from coal combustion (Sakulpitakphon et al., 2000; Díaz-Somoano and Martínez-Tarazona, 2002; Dunham et al., 2003; López-Antón et al., 2006). This type of matrix is susceptible not only to trace element condensation on the finest particles (Díaz-Somoano and Martínez-Tarazona, 2002) but also to gas-solid surface reactions in a

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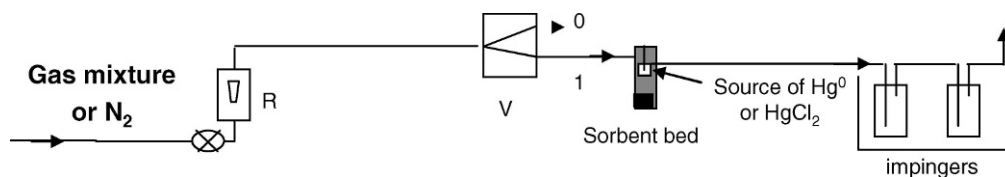


Fig. 1. Schematic diagram of experimental device.

medium that contains metal species in gas phase. Examples of these reactions include the chemically controlled surface deposition of Pb on particles of fly ashes containing S (Clarke and Sloss, 1992) and reactions of As on the surface of particles containing Ca (Mahuli et al., 1997). Recent research has shown that certain fly ash materials have an affinity for Hg (López-Antón et al., 2007a) and that the unburned material present in fly ash shows considerable retention capacity (Pavlish et al., 2003; Li et al., 2002). However, there are a number of variables that may influence mercury capture by fly ashes (López-Antón et al., 2007a; López-Antón et al., 2007b; Goodarzi and Hower, 2008), and the exact nature of Hg-fly ash interactions and the influence of the process conditions need to be investigated more thoroughly.

The aim of the present work is to review the variables that have an effect on the retention of mercury in fly ashes taken from power stations which burn coals of different rank and nature. The mercury retention capacity of these fly ashes was compared with retention in commercial activated carbons. The three main variables studied were: i) the nature and characteristics of the sorbents (raw fly ashes with a small amount of unburned particle content, fly ashes enriched in unburned particles, and activated carbons) ii) the mercury species in gas phase ( $\text{Hg}^0$  and  $\text{HgCl}_2$ ) and iii) the gas atmosphere (nitrogen and a simulated flue gas).

## 2. Experimental Section

Four fly ash samples (CTA, CTSR, CTES, and CTP) and two activated carbons (RBHG3 and RB3) were used as mercury sorbents. CTA, CTSR, and CTES were obtained from different pulverized coal power plants (PCC). CTA was taken from a coal power plant in which the coal blend contained a high rank coal. CTSR and CTES were sampled from two PCC plants in which the coal blends contained bituminous and subbituminous coals respectively. The fourth fly ash (CTP) was taken from a fluidized bed combustion plant (FBC) that burns mixtures of coal and coal wastes with a high mineral matter content and uses limestone in the bed. The commercial activated carbon, Norit RBHG3, is prepared by impregnating Norit RB3 with S compounds, with the S content of 6.07 and 0.47 wt%, respectively. The fly ashes were used in their original sizes (12–57  $\mu\text{m}$ ) and the activated carbons were ground to a particle size of 0.2–0.5 mm. Fractions of fly ashes enriched in unburned carbon particles were separated on the basis of a previous size fractionation study (López-Antón, 2004). Thus, besides the original fly ashes, three more samples were tested: CTA > 150  $\mu\text{m}$ , CTSR > 80  $\mu\text{m}$ , and CTES > 200  $\mu\text{m}$ . In the case of CTP, the unburned particles were homogeneously distributed between the different sizes and for this reason only the raw CTP sample was used in this study.

A schematic diagram of the experimental equipment is shown in Figure 1. The  $\text{Hg}^0(\text{g})$  and  $\text{HgCl}_2(\text{g})$  present in the gas atmosphere were obtained by evaporating  $\text{Hg}^0$  and  $\text{HgCl}_2$  respectively. The mercury concentrations in gas phase for  $\text{Hg}^0$  were 0.4  $\mu\text{g ml}^{-1}$  in

the combustion atmosphere and 0.1  $\mu\text{g ml}^{-1}$  in the inert atmosphere. In the case of  $\text{HgCl}_2$ , the concentration was 0.4  $\mu\text{g ml}^{-1}$  in both atmospheres. These higher concentrations were used with the aim of reaching a high retention capacity in a reasonable time period. Although this fact influences the absolute retention capacity for each one of the materials tested, it is valuable to differentiate among the materials tested as sorbents in similar conditions. A synthetic gas mixture was passed through the reactor. The simulated gas flue contained 15%  $\text{CO}_2$ , 9.2%  $\text{O}_2$ , 0.2%  $\text{SO}_2$ , 6.6%  $\text{H}_2\text{O}$ , and was balanced with  $\text{N}_2$ . HCl was not included in the synthetic atmosphere, as is well known, HCl may influence mercury oxidation and, therefore, in this way we can contribute to understand the influence of other variables in such oxidation. This mixture carried the element compound in vapor phase through the sorbent bed at a flow rate of 0.5  $\text{L min}^{-1}$ . The temperature of the sorbent was 120°C. The element that could not be retained in the sorbent bed was captured in impingers containing 4%  $\text{KMnO}_4$  + 10%  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$  0.5N. The sorbent bed was prepared by mixing 1 g of fly ash or activated carbon with 3 g of sand. The bed was 2.5 cm in diameter and 1.1 cm in height. The amount of mercury retained was determined by analyzing the sorbents post-retention by means of cold vapour atomic absorption (CV-AA) after mercury extraction using 60% (v/v)  $\text{HNO}_3$ . An Automatic Mercury Analyser (AMA) was used to determine the mercury content of the solid directly.

BET surface area was determined by volumetric adsorption of nitrogen at 77K. Atomic absorption spectrometry (AAS) was used to determine the elemental composition, including total CaO. Free CaO was determined by the Standard method UNE-EN 451-1

## 3. Results and Discussion

Many parameters may affect mercury capture in solid sorbents during coal combustion. These include gas composition, the chemical properties of the sorbents, the process conditions, mercury speciation, etc (Pavlish et al., 2003). Three important variables are reviewed in this work in the following sections. Two parameters were used to evaluate mercury retention in fly ashes and activated carbons: i) Maximum retention capacity (MRC), which was determined as the maximum amount of mercury retained per g of sorbent and ii) efficiency (E), which was calculated as the percentage of mercury captured. The confidence limit of the results is given as the standard deviation. The results are summarized in Table 1.

### 3.1. Influence of the composition and characteristics of the sorbents

Numerous studies at laboratory scale have been performed to evaluate mercury capture in fly ashes and activated carbons. Although several studies have been carried out to understand the mechanism of mercury retention (Padak and Wilcox, 2009), the effect of the inorganic ash constituents and organic matter on the

**Table 1**

Maximum mercury retention capacity (MRC) and efficiency (E) in fly ash and activated carbon samples.

| Sorbent  | Hg <sup>0</sup>           |         |                           |         | HgCl <sub>2</sub>         |         |                           |         |
|----------|---------------------------|---------|---------------------------|---------|---------------------------|---------|---------------------------|---------|
|          | combustion                |         | N <sub>2</sub>            |         | combustion                |         | N <sub>2</sub>            |         |
|          | MRC<br>mg g <sup>-1</sup> | E<br>%  | MRC<br>mg g <sup>-1</sup> | E<br>%  | MRC<br>mg g <sup>-1</sup> | E<br>%  | MRC<br>mg g <sup>-1</sup> | E<br>%  |
| CTA      | 12±1                      | 11±2    | 0.3±0.0                   | 2.7±0.8 | 2.5±0.3                   | 14±1    | 2.5±0.2                   | 16±4    |
| CTA>150  | 13±1                      | 15±2    | 0.3±0.0                   | 2.6±0.9 | 2.2±0.3                   | 17±3    | 2.5±0.3                   | 14±2    |
| CTSR     | 25±1                      | 8.1±3.0 | 3.7±0.2                   | 16±2    | 12±1                      | 21±1    | 3.6±0.4                   | 14±1    |
| CTSR>80  | 27±1                      | 10±2    | 3.8±0.2                   | 15±1    | 16±2                      | 16±8    | 4.3±0.8                   | 11±2    |
| CTES     | 1.5±0.2                   | 1.5±0.3 | 0.1±0.0                   | 0.1±0.0 | 3.2±0.4                   | 2.1±1.2 | 1.7±0.3                   | 2.3±1.0 |
| CTES>200 | 9.3±0.5                   | 2.5±0.2 | 0.2±0.0                   | 0.2±0.0 | 7.0±0.5                   | 5.6±1.0 | 6.5±0.3                   | 5.8±0.9 |
| CTP      | 0.7±0.1                   | 3.2±1.1 | 0.6±0.0                   | 2.2±1.1 | 5.0±0.5                   | 19±1    | 5.6±0.4                   | 23±2    |
| RBHG3    | *                         | 55±6    | *                         | 46±3    | 11±1                      | 30±4    | 11±1                      | 32±5    |
| RB3      | 7.5±0.8                   | 21±8    | 7.5±0.9                   | 12±4    | 10±1                      | 24±6    | 10±1                      | 35±5    |

\* 150mg g<sup>-1</sup> value supplied by NORIT

retention of mercury has still not been fully assessed. Some inorganic components of fly ashes, such as iron oxide, aluminosilicates, and calcium oxide promote mercury oxidation and hence a better capture (Norton et al., 2001; Norton et al., 2003; Ghorishi et al., 2000). The composition of the inorganic components of the fly ashes and of the activated carbons is shown in Table 2. From an evaluation of the ash composition of the original fly ash samples, the most remarkable difference was found to be the higher CaO free content in CTP. Ghorishi and Sedman (1998) suggested that other components in high-Ca ashes may scavenge Hg<sup>0</sup>, but the exact mechanisms are unclear (Senior et al., 2000). In fact, CTSR fly ash showed the maximum retention capacity (Table 1) but the highest CaO free content was found in CTP (Table 2). Since interactions between CaO particles and HgCl<sub>2</sub>(g) are possible (Gullett and Ragnunathan, 1994), the CaO free content should be considered as an important parameter for a satisfactory ash composition evaluation (Table 2) and will be also considered in the section 3.2. The mineral composition of RBHG3 activated carbon is very similar to the composition of RB3. In these cases, no relation was found between the compositions of the samples and mercury capture (López-Antón et al., 2007a; López-Antón et al., 2007b).

With respect to the role of fly ash carbon content, mercury capture is often but not always enhanced by high levels of unburned carbon (high loss of ignition [LOI]) (Butz and Albiston, 2000; Hassett and Eylands, 1999; Hower et al., 2006). A summary table (Table 3) of the ash content, LOI, surface area and mercury content in different sorbents is shown in this study. Activated carbons contain more than 90% wt of carbon material and the fly ashes enriched in unburned carbon studied in this work have LOI

values between 18 and 54% wt. In the study previously carried out by Lopez-Anton et al. (2007a) and here extended to include another type of fly ash, CTES, no clear relation between the quantities of carbonaceous material (Table 3) and mercury retention (Table 1) was observed. However, a close examination of the retention by each fly ash and its corresponding fraction shows that retention by the fraction enriched in carbon is greater in most cases, especially in the case of fly ash (CTES) taken from the power station that burns subbituminous coals.

Another sorbent characteristic that affects the retention of mercury is the surface area. The amount of mercury retained generally increases with the surface area of the fly ash (Serre and Silcox, 2000). However, in some fly ashes which are more porous and have less surface area, mercury capture is enhanced (Yudovich and Ketris, 2005). In this study, the highest values for surface area were found in CTES>200 and CTSR>80 (13.4 m<sup>2</sup>g<sup>-1</sup>; 17.6 m<sup>2</sup>g<sup>-1</sup>) but not in all cases was the mercury retention capacity higher (Tables 1–2). Thus, a larger surface area does not necessarily imply enhanced mercury capture. These values are not obviously significant compared to the activated carbons (RBHG3: 868 m<sup>2</sup>g<sup>-1</sup>; RB3:1183 m<sup>2</sup>g<sup>-1</sup>). However, RB3 showed a lower retention capacity for Hg<sup>0</sup> than some fly ashes in the simulated flue gas (Table 1). As might be expected, RBHG3, a commercial activated carbon impregnated with sulfur, was the best adsorbent because the mechanism responsible for mercury capture is chemical adsorption (López-Antón et al., 2002). Efficiency was found to be higher in the activated carbons than in the fly ashes (Table 1). This suggests that surface area is not the main factor affecting retention, even though it may control the kinetics of the process.

**Table 2**

Elemental composition for the different fly ash samples.

| % (db)                         | CTA  | CTA>150 | CTSR | CTSR>80 | CTES | CTES>200 | CTP  | RBHG3 | RB3   |
|--------------------------------|------|---------|------|---------|------|----------|------|-------|-------|
| SiO <sub>2</sub>               | 53.3 | 43.2    | 55.3 | 24.2    | 58.5 | 41.0     | 52.1 | 1.87  | 1.70  |
| Al <sub>2</sub> O <sub>3</sub> | 25.6 | 20.4    | 22.7 | 14.4    | 23.7 | 25.7     | 21.9 | 0.30  | 0.21  |
| Fe <sub>2</sub> O <sub>3</sub> | 5.87 | 5.14    | 4.84 | 3.49    | 10.5 | 8.38     | 5.97 | 0.27  | 0.19  |
| MgO                            | 1.82 | 1.27    | 1.51 | 0.87    | 1.03 | 0.67     | 1.39 | 0.63  | 0.35  |
| Na <sub>2</sub> O              | 0.72 | 1.87    | 0.62 | 0.19    | 0.88 | 0.41     | 0.63 | 0.19  | 0.15  |
| K <sub>2</sub> O               | 3.37 | 3.18    | 2.42 | 1.92    | 1.54 | 1.05     | 2.98 | 0.36  | 0.23  |
| TiO <sub>2</sub>               | <1   | <1      | 0.74 | 0.39    | 0.66 | 0.33     | <1   | <0.05 | <0.05 |
| SO <sub>3</sub>                | 0.31 | 0.17    | 0.46 | 0.27    | 0.83 | 0.62     | 5.12 | 1.00  | 0.49  |
| CaO total                      | 2.09 | 1.25    | 2.77 | 1.40    | 0.32 | 3.53     | 6.36 | 0.47  | 0.34  |
| CaO free                       | 0.11 | 0.04    | 0.22 | 0.20    | 0.30 | 0.20     | 1.06 | –     | –     |

**Table 3**

Ash content, LOI, BET surface area and mercury content of different sorbents.

| Sample   | Ash content (%) | LOI (%) | Surface Area ( $\text{m}^2\text{g}^{-1}$ ) | Hg ( $\mu\text{g g}^{-1}$ ) |
|----------|-----------------|---------|--|-----------------------------|
| CTA      | —               | 5.7     | 1.6  | 0.39                        |
| CTA>150  | —               | 22      | 4.2  | 0.40                        |
| CTSR     | —               | 7.2     | 9.4  | 1.80                        |
| CTSR>80  | —               | 54      | 17.6                                       | 4.95                        |
| CTES     | —               | 2.0     | 1.9  | 0.04                        |
| CTES>200 | —               | 18      | 13.4                                       | 0.10                        |
| CTP      | —               | 3.8     | 6.7  | 1.10                        |
| RBHG3    | 5.5             | —       | 868  | —                           |
| RB3      | 6.0             | —       | 1183                                       | —                           |

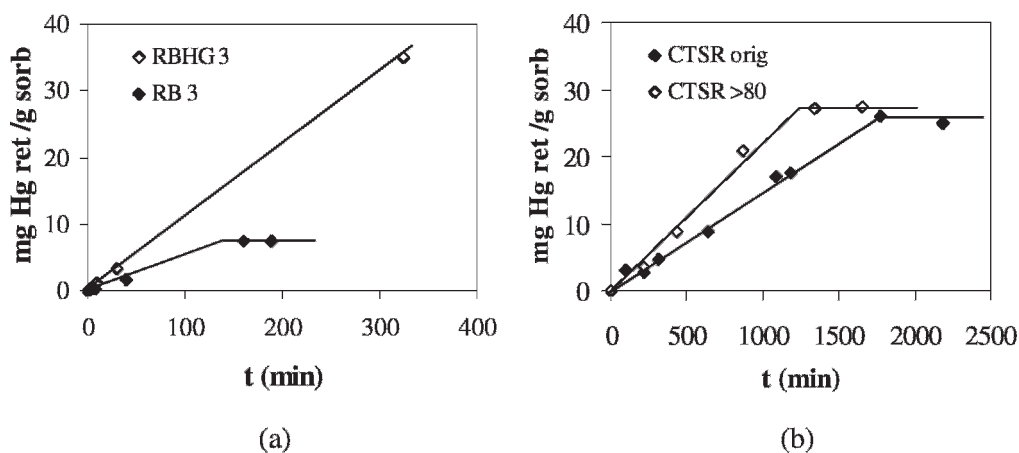
### 3.2. Influence of mercury species in gas phase

During coal combustion, the mercury is transformed into three species: (i) particle-bound mercury ( $\text{Hg}_p$ ); (ii) vapour-phase elemental mercury ( $\text{Hg}^0$ ) and (iii) vapour-phase oxidized mercury ( $\text{Hg}^{2+}$ ), primarily in the form of  $\text{HgCl}_2$ . Retention has therefore been evaluated depending on whether Hg or  $\text{HgCl}_2$  is the mercury species in gas phase. According to a theoretical assessment of the thermodynamic equilibrium data obtained at  $120^\circ\text{C}$  (temperature of the sorbent bed), if  $\text{Hg}^0$  were the source of the element in the combustion and inert atmospheres,  $\text{Hg}(\text{g})$  would be the main species present in gas phase, whereas  $\text{HgCl}_2(\text{g})$  would be the main

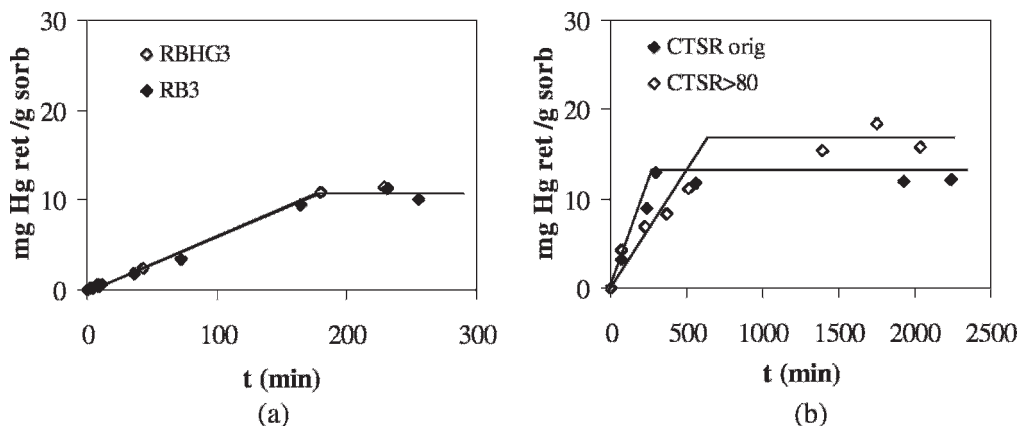
species if the evaporated solid were  $\text{HgCl}_2$  (López-Antón et al., 2007a).

The results clearly demonstrate that fly ashes have a different retention capacity for these two species and that this capacity may be modified by the gas atmosphere (Table 1). In the first study carried out with CTA and CTSR fly ash samples and different mercury species in gas phase (López-Antón et al., 2007a), it was observed that MRC in the case of  $\text{HgCl}_2$  was similar or higher than in the case of  $\text{Hg}^0$  in an inert atmosphere. This has been verified with the fly ashes included in this work (CTP and CTES). However, in the combustion atmosphere, retention capacities for  $\text{HgCl}_2$  are similar or lower than for  $\text{Hg}^0$  with the exception of those attained by the CTP fly ash (Table 1). Considering that interactions between CaO particles and  $\text{HgCl}_2(\text{g})$  are possible (Gullett and Ragnunathan, 1994; Krishnan, et al., 1997), the higher retention of  $\text{HgCl}_2$  than  $\text{Hg}^0$  in CTP may be due to its higher CaO content (Table 2). However, no linear correlation between the CaO content and  $\text{HgCl}_2$  retention was observed.

In general, the mercury retention capacity in the case of  $\text{Hg}^{2+}$  is better than in the case of  $\text{Hg}^0$  in activated carbons. In this study, the MRC of RB3 carbon for  $\text{HgCl}_2$  is also slightly higher than for  $\text{Hg}^0$  (Table 1). The retention of elemental mercury can be improved if the activated carbons are impregnated with sulphur or iodine (Liu et al., 2000; Karatza et al., 2000).  $\text{Hg}^0$  retention by RBHG3 is much higher than for  $\text{HgCl}_2$ .



**Fig. 2.** Retention of  $\text{Hg}^0$  in activated carbons (a) and in fly ash samples (b) in the combustion atmosphere.



**Fig. 3.** Retention of  $\text{HgCl}_2$  in activated carbons (a) and in fly ash samples (b) in the combustion atmosphere.

The retention of  $\text{HgCl}_2$  in the commercial activated carbons, which may involve physical adsorption, is higher than in most fly ashes, with the exception of CTSR. This may be related to the common surface properties of these carbons. This study confirms that the CTSR fly ash, as well as its fraction enriched in unburned particles  $\text{CTSR} > 80$ , continues to be the best sorbent for both species in most of the conditions. The retentions of  $\text{Hg}^0$  and  $\text{HgCl}_2$  by CTSR fly ash and by RB3 and RBHG3 activated carbons in the combustion atmosphere are compared in Figure 2 and Figure 3.

### 3.3. Influence of the gas atmosphere

In general, the mercury sorption capacities of these sorbents in a nitrogen atmosphere were found to be similar. However, the experimental results may be influenced by the flue gas constituents, which often have a greater effect on the surface reactions than the properties of the solid sorbents (Pavlish et al., 2003). In the absence of acid gases, Miller et al. (2000) showed that a lignite-based activated carbon accounted for only 10–20% mercury capture of  $\text{Hg}^0$ . When the sorbent was exposed to  $\text{SO}_2$ , the capture improved slightly and when HCl, NO, or  $\text{NO}_2$  were added, the mercury captured amounted to 90–100%. The presence of acid gases causes a rapid oxidation of the mercury (Miller et al., 1998). López-Antón et al. (2007b) compared the retention of  $\text{Hg}^0$  in some original fly ashes and in activated carbons in different atmospheres. MRCs for fly ashes were found to be higher in the simulated flue gas ( $\text{CO}_2$ ,  $\text{O}_2$ ,  $\text{SO}_2$ ,  $\text{H}_2\text{O}$  and  $\text{N}_2$ ) than in the nitrogen atmosphere in contrast with activated carbons where MRCs were similar in both atmospheres. This was attributed to the possible effects of the inorganic components of the fly ashes on mercury oxidation. This suggests that the species retained in the fly ashes in the combustion atmosphere was not  $\text{Hg}^0$ , but probably an oxidized mercury compound. The CTES fly ash samples included in the present study showed the same behaviour as the other fly ashes (Table 1).

The behaviour of  $\text{HgCl}_2$  was also studied by López-Antón (2007a) in two fly ashes (CTA and CTSR) in different atmospheres. In the present study, this has been complemented by the inclusion of the CTES and CTP fly ashes and the RB3 and RBHG3 activated carbons. The MRC was found to be similar in most of the cases in both atmospheres. Therefore,  $\text{HgCl}_2(\text{g})$  may be assumed to be the species retained in simulated flue gas and inert atmosphere when  $\text{HgCl}_2$  is the source of mercury.

## 4. Conclusions

A comparison between the results obtained from the fly ash and activated carbon samples indicates that the retention of  $\text{Hg}^0$  is not only influenced by the carbon content and characteristics but also by the gas atmosphere, even in the absence of HCl. Although the efficiency of the mercury retention was higher in the activated carbons than in the fly ashes, some activated carbon showed a lower retention capacity for  $\text{Hg}^0$  than some of the fly ashes in the simulated flue gas. This suggests that in the experimental conditions studied, surface area is not the main factor affecting retention, even though it may control the kinetics of the process. The behavior of mercury changes depending on the mercury species. The retention of  $\text{HgCl}_2$  in the commercial activated carbons, which may involve physical adsorption, is higher than in most fly ashes, with the exception of CTSR and also depends on

the gas atmosphere. The fly ashes obtained from PCC plants are more efficient for mercury retention than those obtained from the FBC studied.

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