



Impact of oxy-fuel combustion gases on mercury retention in activated carbons from a macroalgae waste: Effect of water



M.A. Lopez-Anton, N. Ferrera-Lorenzo, E. Fuente*, M. Díaz-Somoano, I. Suarez-Ruiz, M.R. Martínez-Tarazona, B. Ruiz

Instituto Nacional del Carbón (CSIC), Francisco Pintado Fe, 26, 33011 Oviedo, Spain

HIGHLIGHTS

- Activated carbons obtained from algae industry waste may retain mercury.
- Mercury retention is favored by the presence of O₂ in absence of other gases.
- High percentage of mercury oxidation but no retention in oxy-combustion conditions.

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ABSTRACT

The aim of this study is to understand the different sorption behaviors of mercury species on activated carbons in the oxy-fuel combustion of coal and the effect of high quantities of water vapor on the retention process. The work evaluates the interactions between the mercury species and a series of activated carbons prepared from a macroalgae waste (algae meal) from the agar-agar industry in oxy-combustion atmospheres, focussing on the role that the high concentration of water in the flue gases plays in mercury retention. Two novel aspects are considered in this work (i) the impact of oxy-combustion gases on the retention of mercury by activated carbons and (ii) the performance of activated carbons prepared from biomass algae wastes for this application. The results obtained at laboratory scale indicate that the effect of the chemical and textural characteristics of the activated carbons on mercury capture is not as important as that of reactive gases, such as the SO_x and water vapor present in the flue gas. Mercury retention was found to be much lower in the oxy-combustion atmosphere than in the O₂ + N₂ (12.6% O₂) atmosphere. However, the oxidation of elemental mercury (Hg⁰) to form oxidized mercury (Hg²⁺) amounted to 60%, resulting in an enhancement of mercury retention in the flue gas desulfurization units and a reduction in the amalgamation of Hg⁰ in the CO₂ compression unit. This result is of considerable importance for the development of technologies based on activated carbon sorbents for mercury control in oxy-combustion processes.

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1. Introduction

Oxy-coal combustion is a promising technology in the field of CO₂ capture and sequestration (CCS). The basic principle behind this technology is to burn coal in a mixture of O₂ and recycled gas so that the resulting flue gas is mainly CO₂ and H₂O which simplifies the process of separation of CO₂ from the flue gas for further storage or beneficial use. Although this gas also contains small quantities of O₂, N₂ and minor concentrations of SO_x and NO_x, the CO₂ can be relatively easily separated before its capture

(Buhre et al., 2005; Wall et al., 2013). The new oxy-combustion conditions may lead to certain changes with respect to conventional combustion conditions, which might involve modifications not only in the composition of the reactive gases (Normann et al., 2009; Liémans et al., 2011; Stanger and Wall, 2011; Ting et al., 2013), but also in the fate of the trace elements (Roy and Bhattacharya, 2014). Of the trace elements, mercury is the one of greatest concern because coal combustion is one of the major anthropogenic sources of mercury to the environment (Zhuang et al., 2011b; Roy et al., 2013).

Mercury is a highly toxic element whose environmental impact is widely recognised, which is why governments and several organizations have introduced measures to bring it under control (USEPA, 2011). In particular, one of the UNEP (United Nations

* Corresponding author. Tel.: +34 985 119090; fax: +34 985 297662.

E-mail address: enriquef@incar.csic.es (E. Fuente).

Environment Programme) targets is to protect human health from the release of mercury and its compounds (UNEP, 2013), and a priority in this field, is the mercury released from coal combustion. However, in the case of oxy-combustion, mercury is not only an environmental problem but also a technological one since Hg^0 can accumulate in the CO_2 compression units and damage the aluminum heat exchangers due to amalgamation of mercury with aluminum (Santos, 2010).

There are basically two ways to capture mercury during coal combustion processes. One of them is to install purpose-designed systems for mercury capture in power stations, and the other is to make use of already installed systems intended for the capture of other contaminants for the co-removal of mercury. A large number of the purpose-designed systems for capturing mercury species in gas phase are based on solid sorbents. Of these, activated carbons that are capable of physically or chemically adsorbing this element, are the most common (Sjostrom et al., 2010). The effectiveness of activated carbons, which have been evaluated not only at laboratory and pilot level, but also at industrial scale (Pavlish et al., 2010), depends on particle size, surface area, porosity, etc. (López-Antón et al., 2002), and gas composition, since reactive gases can influence mercury speciation and mercury sorption mechanisms.

Numerous studies have already been performed on the application of activated carbons for mercury capture in gases from combustion, and to refer to all of them is outside the scope of this paper. However, the knowledge accumulated by these studies constitutes a solid basis to address the emerging problems associated with oxy-combustion that, given the current state of the art, are far from being understood. To achieve a solution to these problems it is first necessary to identify the mechanisms of heterogeneous mercury oxidation on the carbon surface and to determine how the gas composition may modify these mechanisms (Liu and Vidic, 2000; Miller et al., 2000; Granite and Presto, 2008). To achieve these goals, the effects of reactive gases need to be considered.

Although the effect of SO_2 on mercury adsorption is of great importance, it has not yet been fully clarified even in conventional combustion plants. Some authors (Uddin et al., 2008; Diamantopoulou et al., 2010; Morris et al., 2012) have concluded that SO_2 favors mercury adsorption, particularly in the presence of O_2 , due to the formation of HgSO_4 on the surface. Other studies (Presto and Granite, 2007; Granite and Presto, 2008; Sjostrom et al., 2009; Morris et al., 2010; Zhuang et al., 2011a; His and Chen, 2012), however, suggest that the SO_3 in flue gases leads to the formation of H_2SO_4 on the surface of the activated carbon which leads to pore blockage, inhibiting the adsorption of Hg^0 . The role of water in mercury retention by activated carbons during conventional combustion is another variable whose influence on the adsorption mechanism has been explained in various ways. However, in the case oxy-combustion the role of water needs to be fully clarified because in this particular process, water vapor can be expected to be present in significantly higher concentrations. Apparently, the presence of water in the flue gas, even in the range of 5–10%, seriously impedes the removal of mercury (Cudahy and Helsel, 2000; Richards, 2005; Presto and Granite, 2007), probably due to the adsorption of water molecules into the micropore structure of the activated carbon, which reduces the number of the active sites available for mercury (Zheng et al., 2012). However, Li et al. (2002) found that moisture on activated carbon surfaces favors Hg^0 adsorption at room temperature.

To stress the importance of flue gas composition for mercury retention on activated carbon is not to imply that the characteristics of the activated carbon itself are not important. Typical activated carbons may capture Hg^{2+} , but in general they are not good sorbents for Hg^0 . The retention of Hg^0 can be significantly

enhanced by using activated carbons impregnated with sulfur, chlorine, bromine or iodine that chemically interact with Hg^0 (Asasian and Kaghazchi, 2013; Rupp and Wilcox, 2014). The limitation that is often attributed to these impregnated materials is the cost of the preparation (Pacyna et al., 2010). This explains why the use of solid wastes as precursors for the preparation of activated carbons has emerged as a competitive way to produce these materials (Klasson et al., 2010). Satisfactory results for mercury retention have been obtained using renewable sources of carbon such as bamboo charcoal, poultry manure, waste tires and chicken waste (Skodras et al., 2007; Yaji et al., 2008; Tan et al., 2011). Although some of these sorbents have been tested at industrial scale (De et al., 2013), most of the studies have so far been focused on the development of good textural and chemical properties of the activated carbons with very little attention being paid to the effect that the gases in a full coal combustion atmosphere have on mercury retention. In addition, the results may be very different in an oxy-composition atmosphere and in one of air combustion due to their different gas compositions.

In view of these considerations this work focuses on the interactions between mercury species and a series of activated carbons prepared from a waste from the agar–agar industry in an oxy-combustion atmosphere, paying special attention to the role that the high concentration of water in the flue gases can play in the retention of mercury.

2. Experimental

The activated carbons used in this work were prepared from two types of precursor (i) a macroalgae waste (algae meal) from the agar–agar industry (AM) and (ii) a char (AMP) obtained by pyrolysis of AM at 750 °C. The conditions for obtaining both samples have been previously described (Ferrera-Lorenzo et al., 2014a). These precursors (AM and AMP) were chemically activated with KOH, in precursor/KOH proportions of 0.5:1 and 1:1. The activation temperatures used were 750 and 900 °C and the heating rate and N_2 flow rate were 5 °C min^{-1} and 150 mL min^{-1} , respectively (Ferrera-Lorenzo et al., 2013, 2014a). The activated carbons obtained from AM and AMP were labelled AMA and AMPA followed by the precursor/KOH proportions and the activation temperatures used (Table 1). These materials were washed with HCl 5 M and deionized water, and dried before testing. A commercial sulfur-impregnated activated carbon developed for Hg^0 retention (Norit RBHG3), was employed as a reference material (López-Antón et al., 2002).

The moisture content of the macroalgae waste was obtained following the UNE 32002 norm, on the basis of weight loss at 105 °C over a period of 1 h, and the ash content was determined by calcining the sample in a muffle at 815 °C for 1 h in the presence of oxygen, according to the UNE 32004 norm. The carbon, hydrogen and nitrogen contents of the samples were determined using LECO CHN-2000 equipment. The sulfur content was measured on a LECO S-144-DR equipment. The chlorine concentration was mea-

Table 1
Activated carbons obtained in the experimental conditions used.

Sample	Activation agent/precursor	Activation temperature (°C)	N_2 flow rate (mL min^{-1})
AMA0.5:1-750	0.5:1	750	150
AMPA0.5:1-750	0.5:1	750	150
AMPA0.5:1-900	0.5:1	900	150
AMPA1:1-750	1:1	750	150
AMPA1:1-900	1:1	900	150

Nomenclature: AMA: Activated carbon from algae meal (AM); AMPA: Activated carbon from algae meal carbonized (AMP).

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