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Mass balance and partitioning of trace elements under oxy-coal combustion: First experiences

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ABSTRACT

Mass balance and partitioning of inorganic trace pollutants have been evaluated at the largest oxy-Pulverised Coal Combustion (oxy-PCC) demonstration plant to date owned by Fundación Ciudad de la Energía (CIUDEN). Results reveal that the concentration of gaseous SO_2, B, Se, F, and HCl increases progressively in the CO_2-rich flue gas along the depuration train. This results in negative retention efficiencies of these types of pollutants and in the enrichment of trace elements in the oxy-fly ashes (oxy-FAs). These two factors affected the MB closure for highly volatile S, Cl, and F, and moderately volatile with high condensation potential B, As, and Se over the oxy-PCC and whole installation. This first experience has lead us to focusing further work on 1) elucidating the behaviour of inorganic trace pollutants in the CO_2-rich flue gas after a number of CO_2-rich flue gas re-circulations back to the boiler; and 2) investigating the speciation and leaching potential of inorganic trace pollutants, especially F, retained in the oxy-FAs in order to establishing remediation actions if required.

1. Introduction

Oxy-fuel combustion is a promising technology that may greatly contribute to the reduction of carbon dioxide (CO_2) emissions from power plants. The principle of oxy-fuel combustion is to increase the partial pressure of CO_2 in the flue gas in order to make its sequestration and compression easier and more cost effective [1]. This can be achieved by burning a fuel with pure oxygen instead of air as a primary oxidant, which results in higher flame temperatures. For this reason, the mixture is diluted with a portion of the resulting flue gas composed primarily of CO_2.

Fundamentally, due to the variation in the oxidant, and consequently in the in-furnace gas environment (as compared to conventional air-fired combustion), oxy-fuel combustion affects the combustion process of pulverised coal as well as related processes such as heat transfer [2]. As a result, most studies and research projects have focused on fundamental scientific and engineering issues (e.g. ignition, flame stability, heat transfer, recycled flue gas ratio, and combustion characteristics), and less attention to the formation, partitioning, speciation and fate of trace pollutants during oxy-fuel combustion.

Oxy-fuel combustion, in addition to the production of H_2O and CO_2, generates CO, SO_X, NO_X, and particulate matter (PM) and minor/trace levels of the gases HF, HCl, Hg, Br, As, Se, and B, among others. Significant quantities of the aforementioned pollutants in the CO_2-rich recycled flue gas stream may result in a 3–4 times increase of SO_X, HCl, HF, and Hg, with respect to the conventional pulverised coal combustion (PCC). This is not only due to the lack of flue gas dilution by airborne N_2 [3–5], but also due to the flue gas recycling [6]. Given that SO_X, HCl, and HF are potentially corrosive, this significant increase may cause problems in the boiler components and oxy-fuel combustion process. Sulphur trioxide (SO_3), formed by oxidation of SO_2, is known to be responsible for low temperature corrosion in cold parts of the flue gas train, while the acid gases NOx and SOx together with condensed water are highly acidic, and therefore, corrosive liquids in the CO_2 compression system [7]. High Hg concentrations in an oxy-fuel process may also have potential implications on corrosion of aluminum (Al)-alloys [8], which could be specifically problematic in the CO_2 processing unit of an oxy-fuel combustion power plant [9] where Al-alloys are applied, e.g. for heat exchangers [8].

Yang et al. [10], in a study at a 35 MWth large pilot boiler of oxy-fuel combustion with different flue gas recycle indicated the conversion of gaseous Hg to particulate associated Hg (Hgp) is enhanced in oxy-fuel combustion.

Oxy-fuel combustion is not a CO_2 capture selective method [8], but

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rather the CO_2 is enriched by recirculating the flue gas back to the combustion. Therefore, the CO_2-rich flue gas must be purified to meet the requirements of the pipeline transportation system and the constraints of its utilization and the proposed storage site. Further, the control of the non-CO_2 components from both the boiloperation and CO_2 processing/purification, transport, and storage is of paramount importance. With this in mind, a special focus must be given to the partitioning, behaviour and fate of inorganic trace pollutants during oxy-fuel combustion.

Recirculating a CO_2-rich gas stream with significant quantities of these types of pollutants may also enhance their content in the solid by-products, such as fly ash (FA), which could later lead to environmental leaching problems and reduce the reuse possibilities of this solid residue. The potential risk of contaminating the FA with excessive amounts of trace pollutants is thus very critical to the configuration of an oxy-fuel combustion power plant at an industrial scale. Accordingly, the aims of this work are to: *1)* establish the mass balance and the partitioning of inorganic trace pollutants at a 20 MWth oxy-Pulverised Coal Combustion (oxy-PCC) demonstration plant; and 2) evaluate the effect of the CO_2-rich flue gas re-circulated back to the boiler on the partitioning of inorganic trace pollutants.

2. Methodology

2.1. Oxy-fuel facility: first experiences

This project was conducted in 2014 at the largest oxy-Pulverised coal combustion (oxy-PCC) (20 MWth) demonstration plant to date owned by Fundación Ciudad de la Energía (CIUDEN). The 20 MWth oxy-PCC incorporates a fuel preparation unit where coals are ground, milled and dried; PC boiler that operates in both air and full oxy-model; an economiser where the heat of exhaust steam (30 bar and 420 °C) is used to raise the temperature of incoming water in the PC boiler; and a modular system for the treatment of flue gases (Fig. 1).

The fuel preparation comprises a number of sequential stages. Firstly, raw coal from 25 t-capacity trucks is stored, as received, in a hopper (42 m^3) to feed a coal crusher (15 t/h). Crushed coal is then stored in two coal silos (120 m^3) and transferred to a ball coal mill (5 t/h) to be ground. When the PC boiler is in operation, coal stored in the daily crushed coal silo feeds the mill-classifier where coal is pulverised and dried by means of a natural-gas generator. Pulverised coal is then transported suspended in the drying gases to a bag filter (47,000 m^3) where it is separated and stored in a CO_2 inertized silo (80 m^3) to prevent the risk of explosion. Pulverised coal is taken by screw conveyors and bucket elevator systems and individually fed to each burner, while part of the outlet gas from the bag filter system is returned to the process.

The 20 MWth PC boiler is a vertical water-tube (7.6 m × 4.5 m × 24 m) of natural circulation with draft balanced by forced and induced draft fans, with a selectable and adjustable furnace draft. The unit is equipped with four horizontal or wall firing burners, and prepared for two arch firing burners, which provide a very flexible design and wide range of operation conditions. The oxy-PCC process under which we conducted our research was the 4 wall firing design. This configuration entails the disposition of two wall firing burners (5 MWth) facing each other (Fig. 2). The oxidant stream required for the oxy-PCC is obtained from mixing oxygen with recirculation gas to improve the heat transfer and to temper combustion. At the CIUDEN's CO_2 capture TDP, the CO_2-rich recirculated flue gas (FGR) is divided into three oxidant streams: CB1, CB2, and CB3 (Fig. 1). The primary oxidiser (CB1) carries the pulverised coal into the 4 wall firing burners and supplies part of the oxygen necessary for combustion. The burner comprises a central nozzle from where pulverised coal and CB1 mixture flow. The secondary oxidiser stream (CB2) is divided into three lines CB2.1, CB2.2 and CB2.3 and provides a total and safe combustion of the pulverised coal in the furnace, being at the same time, the principal source of oxygen. The CB2.1, CB2.2 and CB2.3 are positioned through the burner in order to surround the central flame and complete the initiated combustion, achieving a global lower flame temperature (Fig. 2). The 4 wall firing burners have the capability to modify the fluid dynamic characteristics of the CB1 and CB2 streams to make possible the investigation of different flame configurations. Each pulverised coal burner has an integrated gas burner for starting-up and supporting and an ignition system that comprises high voltage electric igniters that ignite the gas. Additionally, the PC boiler operates with an over-fire air (OFA) system as part of an overall NOx reduction strategy by staging the combustion process. The OFA system is operative through the tertiary oxidant (CB3) stream which involves reducing oxygen availability early in the oxy-combustion process and re-introducing it later through ports located above the combustion zone

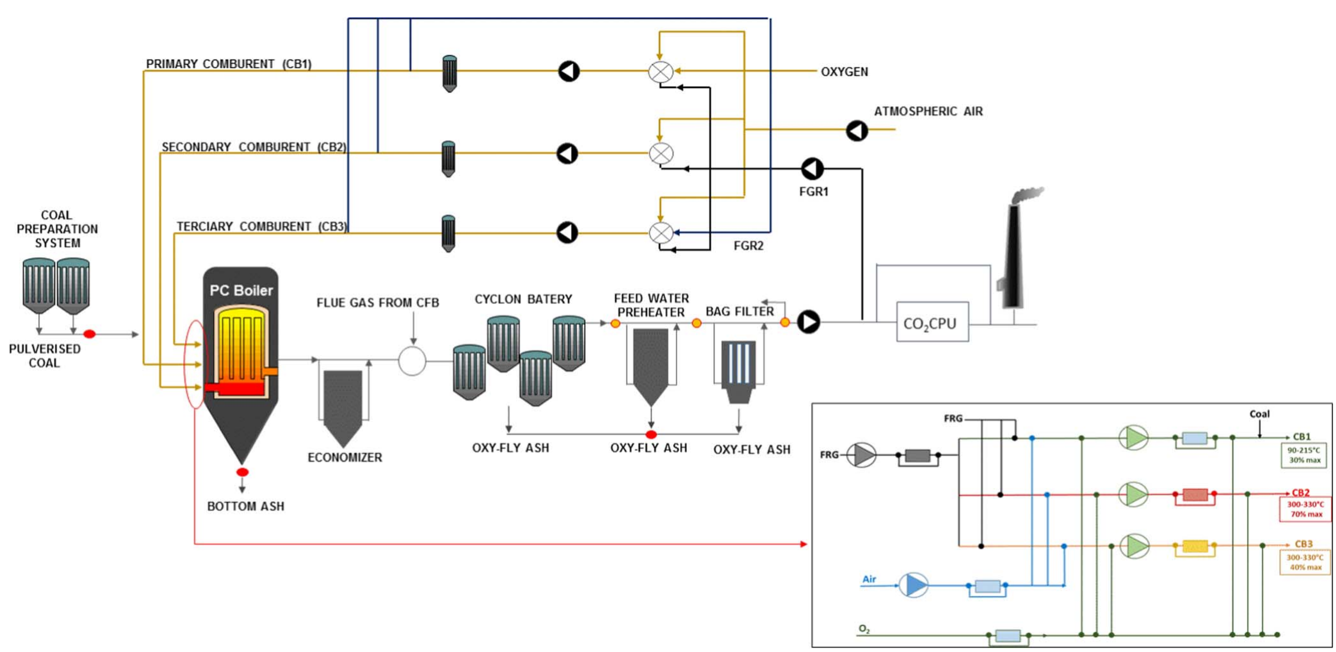

Fig. 1. Diagram and process of the oxy-PCC demonstration plant.