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High-temperature tube corrosion upon the interaction with Victorian brown coal fly ash under the oxy-fuel combustion condition

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Abstract

This study has examined the corrosion of tubes exposed to Victorian brown coal fly ash under the oxy-fuel combustion mode. The temperature of 650 °C, flue gas composition (steam 30 vol%, O₂ 5%, SO₂ 3000 ppm, HCl 1000 ppm, N₂ 5% and CO₂ balanced), and an exposure time of 50 h were employed. Through the joint use of advanced instruments such as synchrotron X-ray adsorption spectroscopy and thermodynamic equilibrium calculation, it has been confirmed that, from both mass loss and penetration rate perspectives, the corrosion of low-Cr tubes under the oxy-firing mode is severe compared to the respective air-firing mode. Both flue gas-related and ash-related corrosions are responsible for tube corrosion. In contrast, the corrosion rate of high-Cr tubes (~20% for SUS304 and SUS347) is comparable between the two modes, due to the protective effect of Cr-oxide. The existence of fly ash on tube surface suppressed the chlorination reaction. Oxygen is the principal element permeating into tube under the air-firing mode, whereas both oxygen and sulphur penetrated into the tube under the oxy-firing mode, accelerating sulphation/sulphidation reactions for a rapid corrosion. Although Cr₂O₃ forms a protective layer under the air-firing mode, it broke down due to the accelerated corrosion attack caused by sulphates/sulphide under the oxy-firing mode. The interaction between ash and tube surface promoted the formation of pure oxides and oxide complexes with the involvement of Na and Ca as well. The presence of alkali sulphates in ash promoted the sulphidation reaction, the extent of which is however tube-specific and correlates little with Cr content. The use of advanced material SUS347 was found to be the most suitable candidate for heat-exchange tubes to be used in the oxy-firing boiler burning low-rank coal such as Victorian brown coal which is rich in alkali and alkaline earth metals. © 2016 by The Combustion Institute. Published by Elsevier Inc.

Keywords: Oxy-fuel combustion; Victorian brown coal; Tube corrosion; XANES Fe and S K-edges

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1. Introduction

Coal is the most abundant fossil fuel in the world and would expect to remain so for the rest of the century. The total amount of coal used is predicted to increase by 5% in 2040 [1]. Victoria, Australia is one of the largest brown coal reserves in the world [2]. Although bearing a very low amount of ash-forming elements in brown coal, the large amount of inherent moisture (up to 70 wt%) within it results in a notoriously high CO₂ emission rate, compared to black coal [3]. Deploying the low-emission technologies such as oxy-fuel combustion is pivotal for the sustainable use of brown coal in the carbon-constrained future.

Compared to conventional air-firing mode, the oxy-fuel combustion provides a different atmospheric environment in the boiler. Apart from the high-concentrations of CO₂ and steam derived from wet flue gas recycle, the trivial but corrosive components including SO₂ and HCl are also potentially accumulated in the furnace, due to the recycle of the uncleaned, dirty flue gas [4,5]. In nearly all of the pulverised coal-fired boilers in Australia, the downstream gas cleaning units do not exist. In light of this, the high-temperature tube corrosion in the heat-exchanger zone is supposedly altered remarkably [6], under the oxy-fuel combustion mode for Australian coals including Victorian brown coal. With regard to the high-temperature tube corrosion, coal quality is the major player on the fire-side [7]. In addition to the afore-mentioned corrosive gases resulting in the oxidation, chlorination, sulphation/sulphidation and even carburisation of tube material [8], the ash deposits also cause the wastage of tube surface, due to the formation of new species such as carbonates that are highly corrosive [9], and the promoted formation of other species such sulphates and chlorides that lower the melting point of tube material by forming new eutectics. As summarised in [9] and further discussed by the studies in an IEAGHG workshop [10], most of the previous studies on high-temperature tube corrosion focused on the effects of gaseous components such as CO₂, H₂O and SO_x. The ash-related corrosion, however has been much less examined. Even for the studies conducted to date, the conclusions achieved are far from generalised [11–14], due to the fact that the high-temperature tube corrosion is highly coal-specific. Victorian brown coal is different from German lignites [10,15] and American coals tested previously [10,16,17], due to the high contents of inherent moisture and sodium within it.

In this study, we presented the results for the ash-related tube corrosions in the typical gas environment encountered during both the air-firing and oxy-fuel combustion of Victorian brown coal. That is, apart from the accumulated SO₂, a steam content of 30 vol% in flue gas is also probable for the recycle of wet flue-gas and the burning of partially

dried coal. HCl was also added into the flue gas, to mimic the co-firing scenario in which biomass is blended with brown coal for a zero, and even negative carbon emission. The test temperature was fixed as 650 °C, at which the tube corrosion rate is maximum [9], and a test duration of 50 h have been conducted. Two typical fly ashes from Victorian brown coal-fired power plants were tested, and six tube materials with a chromium (Cr) content up to 20 wt% have been tested. After each test, apart from weighing the mass change of a tube, a variety of advanced analysis was also conducted, optical microscope (OM) for the cross-section observation to quantify the corrosion depth, scanning electron microscopy (SEM) coupled with energy-dispersive X-ray analyser (EDX) to map the penetration/diffusion of elements of interest through the ash-tube interface, X-ray diffraction (XRD) for the bulk surface, and synchrotron X-ray near-edge structure (XANES) for the oxidation states of iron (Fe), and sulphur (S) on both bulk and cross-section surfaces. Finally, thermodynamic equilibrium modelling was conducted to interpret the experimental observations. Such a study is expected to determine the suitability of and thus optimise the performance of tube materials. Ultimately, it aims to promote the deployment of oxy-fuel combustion for low-rank coals.

2. Experimental procedure

2.1. Tube corrosion test set-up and test conditions

The experimental set-up is illustrated in *Figs. S1* and *S2* in the supporting information (SI) document. A horizontal test furnace was used for tube corrosion test, which was first heated up to 650 °C. The fresh tube specimen, 2 cm × 2 cm × 0.2 mm, were prepared by a wire-electrical discharge machine. The oxidised outer surface was ruled out for testing. Photo of the fresh tube specimen is demonstrated in *Fig. S2* in the SI. Fly ash powder (~100 mg per run) was spread uniformly over the fresh tube plate surface to mimic the ash deposition on a real steam tube surface. The ash-laden tube specimen was then loaded on a quartz-made sample holder and pushed to the middle zone in the hot furnace. Subsequently, flue gas (as shown in *Table 1*) was fed at a flow rate of 300 mL/min continuously into the furnace. Each test lasted 50 h and two replicas were conducted for each condition. After the test, the particle-laden tube specimen was either scratched carefully to remove the particles to weigh its mass change, or quickly mounted (with ash particles together) and solidified into epoxy resin to avoid the surface oxidation. Photos for ash coating, and corroded tube surfaces are further illustrated in *Fig. S3*. Note that, such a testing procedure is consistent with the literature studies related to ash deposit-related tube corrosion [10,15–17].

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