



Full Length Article

Effect of silica additive on the high-temperature fireside tube corrosion during the air-firing and oxy-firing of lignite (Xinjiang coal) – Characteristics of bulk and cross-sectional surfaces for the tubes



Iman Ja'baz^a, Juan Chen^b, Barbara Etschmann^c, Yoshihiko Ninomiya^b, Lian Zhang^{a,*}

^a Department of Chemical Engineering, Monash University, Clayton, Victoria 3800, Australia

^b Department of Applied Chemistry, Chubu University, 1200 Matsumoto-Cho, Kasugai, Aichi 487-8501, Japan

^c School of Geosciences, Monash University, Clayton, Victoria 3800, Australia

HIGHLIGHTS

- The pure CO₂ caused the oxidation of Cr and Fe.
- The use of silica additive is effective in alleviating the diffusion rate of oxygen.
- Formation of augite by reactions between free oxides in ash and carbon steel surface.
- Speciation of S varies with ash deposit and tube material.
- A slightly accelerated outward diffusion of Cr by the ash deposit.

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ABSTRACT

Detailed speciation analysis has been conducted on the corroded tubes coated with two different ash deposits collected from the combustion of a lignite, namely Xinjiang coal mixed with and without external silica in both air- and oxy-firing modes. The exposure conditions are of temperature of 650 °C, 50 h and the use of different flue gases (*i.e.* pure CO₂, air, air-firing flue gas and oxy-firing flue gas). Apart from the lab-based XRD used for the characterisation of the top surface, synchrotron XANES was employed to determine the oxidation states of Fe, S and Cr on both top and cross-sectional surfaces of the corroded tubes. The results indicate that, irrespective of the Cr content, the tubes exposed to pure CO₂ underwent oxidation, which is most likely due to the reaction CO₂ + M = CO + MO where M stands for Fe or Cr. Cr in the Cr-bearing tubes were oxidised more rapidly than Fe in the pure CO₂. The combined use of silica additive and Cr-bearing tubes with a minimal Cr content of 2 wt% (*i.e.* T23) is beneficial in inhibiting the tube surface oxidation as well the penetration of oxygen and sulphur. Except alkali sulphates, the free oxides in ash deposit can also trigger the reactions for the formation of new species such as augite ((Ca,Na)(Mg,Fe,Al,Ti)(Si,Al)₂O₆) on the carbon steel tube surface. The oxidation states of sulphur on the tube surface is highly tube-specific and dependent on ash deposit composition as well, with the abundance of pyrite (FeS₂) on the carbon steel tube coated with the raw coal ash deposits, whilst the enrichment of troilite (FeS) upon the coating of ash deposits derived from the combustion of coal mixed with silica additive, due to the shortage of free sulphur in ash deposit and a preferential scavenging of Fe into chromite. For the Cr-bearing tubes, sulphate is the only S-bearing species due to the inhibited inward diffusion of S by the Cr-oxide layer, irrespective of ash deposit type. In addition, spatial organisation of the oxidation state of Cr suggests a slightly accelerated outward diffusion of Cr by the ash coating, thereby leading to a dense Cr-O layer that is highly protective on the tube top outer surface.

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1. Introduction

Chemical speciation of metals on corroded tube surfaces is pivotal to understanding the mechanisms underpinning the interaction between tube surface and flue gas, as well as the solid-to-solid interaction related to hot corrosion at high temperatures. This

* Corresponding author.

E-mail address: lian.zhang@monash.edu (L. Zhang).

is particularly important for a new combustion process such as oxy-fuel combustion in which flue gas composition and ash deposit properties are altered remarkably compared to the conventional air - firing mode. To date, the knowledge is still sparse regarding metallic speciation on the corroded tubes that are exposed to the ash-laden oxy-firing flue gas [1].

To date, most of the knowledge with regard to the metallic speciation on the corroded tube surfaces was merely achieved from

studies based on conventional air - firing cases, where nitrogen is predominant (nitride formation is thus probable), and the contents of impure SO₂, steam and CO₂ are lean. However, upon employment of the oxy-firing mode, impurities accumulate due to the recycling of the dirty, wet flue gas that are supposed to enhance the formation of corrosive sulphates/sulphides under the same exposure time as under the air-firing case, and even result in the formation of new species. Table 1 summarises all the past studies

Table 1
Summary on the literature studied on tube corrosion during the oxy-fuel combustion.

Reference	Purpose	Testing methods	Coal	Key findings
G. Stein-Brzozowska et al., 2011 [26]	Investigate the influence of an oxy-fuel combustion of a hard-coal on the surface of selected super-heater materials	SEM-EDS	Dry lignite from Germany, hard-coal from South America	Increase in sulphur induced corrosion for tubes exposed to the oxy-combustion atmosphere
G. Stein-Brzozowska et al., 2013 [27]	Study the interactions between flue gas atmosphere, deposits and heat exchanger materials during oxy-fuel firing	SEM/ICP-OES/XRD	Bituminous coal	No Sulphur induced corrosion is detected on the alloy specimens exposed under real fly ash - and pure CaCO ₃ - deposits
G.R. Holcomb et al., 2013 [28]	Compare air-firing and oxy-firing conditions on the tube corrosions	OM/SEM-EDS/XRD	Synthetic ash	The observed corrosion behavior shows accelerated corrosion even with sulphate additions that remain solid at the tested temperatures
K. Natesan and Z.T. Zeng, 2011 [2]	Addresses the potential fireside corrosion issues in environments typical of USC boilers and oxy-fuel combustion system	OM/SEM-EDS/XRD	Synthetic ash	NaCl in the deposit led to catastrophic corrosion at 650 and 800 °C Corrosion rates for Ni-base alloys were significantly less than those of Fe-base alloys under the same exposure conditions
L. Fryda et al., 2010 [29]	Study ash formation and deposition of selected coal/biomass blends under oxyfuel and air conditions in a lab scale pulverised coal combustor (drop tube furnace)	SEM-EDS/ICP/thermodynamic calculation using FactSage software	South African coal blended with shea meal (cocoa)	Deposition propensity was higher under oxy-fuel mode, however ash chemistry was not changed
R. Abang et al., 2013 [11]	Investigate the fireside corrosion behavior of superheater alloys under the oxy-fuel combustion mode	OM/SEM-EDS/XRD	Lignite from Germany	No carbonation observed; although sulfidation was observed, high CO ₂ content in oxyfiring did not result in an increase in corrosion
S. Tuurna et al., 2011 [17]	Investigate the performance of superheater alloys under the oxy-fuel combustion conditions	OM/SEM-EDS/GDOES	85% CaCO ₃ + 15% CaSO ₄	The corrosion resistance increased when the Cr content increased; no severe corrosion under oxy-fuel than the air-firing conditions
A. Hjornhede et al., 2010 [30]	Corrosion test in both oxy-fuel and air-firing conditions	LOM/SEM-EDS		Found evidence for increased deposition rates of ash during oxyfuel combustion compared with air-firing; no significant difference between oxyfuel and air-firing corrosion rates or carburisation
A. Robertson et al., 2010 [31]	Identify the tube corrosion mechanisms under the oxy-fuel conditions, and determine the effect of sulphur on boiler materials	SEM-EDS	Low - S coal Med - S coal High - S coal	The oxy-fired corrosion rates were typically no worse, and often less than air-fired rates
G. Scheffknecht et al., 2009 [32]	Study the fireside corrosion behavior of different alloys	SEM-EDS	Synthetic ash	Carburization and oxide scale morphology differences were confirmed between air and oxy-firing environments for austenitic materials
Y. Tang et al., 2014 [33]	Investigate the corrosion behavior of pure Fe under a Na ₂ SO ₄ deposit in an atmosphere of H ₂ O + O ₂	SEM-EDS/XRD		Corrosion rate is accelerated of the pure Fe significantly under a Na ₂ SO ₄ deposit in atmosphere of H ₂ O + O ₂ at 500 °C
M. Montgomery et al., 2015 [34]	Study the corrosion resistance of various alloys in an oxy-fuel process	OM/SEM-EDS/XRD	Lignite	Corrosion rate decreases with increase in alloying elements. The oxy-fuel ash deposit is similar to that found in conventional air-firing plant
M. Mobin and S.K. Hasan, 2012 [35]	Study the reaction of Fe ₂ O ₃ with Na ₂ SO ₄ in the presence of SO ₂ (g) at 800 and 900 °C	SEM-EDS	UK Coal	Constituents in the reaction products were presented as distinct phase
A.U. Syed et al., 2012 [36]	Assess the effects of increasing heat exchanger surface temperature on the fireside corrosion	OM/SEM-EDS	Biomass/coal mix	A significant increase in metal damage when moving from 600 to 700 °C was confirmed for both alloys with and without ash deposit coverage
K. Natesan and J.H. Park, 2007 [37]	Evaluate the corrosion performance of Fe-base and Ni-base alloys in coal ash environments	OM/SEM-EDS		Ni-base alloys generally exhibited less corrosion than Fe-base alloys under the similar exposure conditions. However, localized pitting was confirmed on the Ni-based tube surface

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