



Effects of calcium in ash on the corrosion performance of Ni-based alloys in simulated oxy-fuel environment

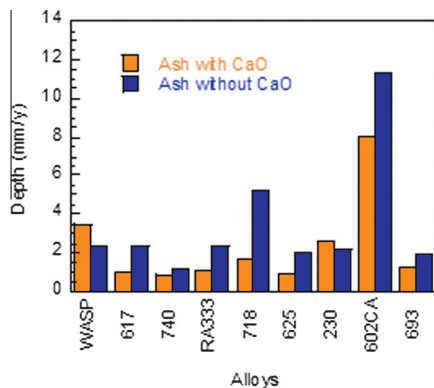


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GRAPHICAL ABSTRACT



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ABSTRACT

Increasing the efficiency of coal power plants requires raising the operating temperature above 650 °C. However, coal ash can severely attack alloy materials at high temperature. For example, the corrosion rates of commercial Fe- and Ni-based alloys are generally greater than 2 mm/year at 750 °C in the gas environment of oxy-fuel combustion. Thus, a critical study is needed to determine the effect of the constituents in the ash on corrosion and find an approach to reduce the corrosion rates in an ash-laden environment at high temperature.

The role of CaO in the ash (typical of U.S. Western coal ash) has been investigated in laboratory exposure environments with various structural alloys. Detailed results are presented on weight change, scale thickness, internal penetration, microstructural characteristics of corrosion products, and the cracking of scales for the alloys after exposure at 750 °C. The thermal stability of $K_3Al(SO_4)_3$ under the environment of oxy-fuel combustion was determined by thermogravimetric analysis and differential thermal analysis. The reaction of this low melting temperature salt with the CaO-containing ash is discussed. In addition, we performed synchrotron nanobeam X-ray analysis to study the phase and chemical composition of the oxide layers on the alloy surface. Results from these studies are used to address the role of CaO in ash in the long-term corrosion performance of alloys.

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1. Introduction

Energy production, particularly electricity generation, is expected to continue to increase globally due to population growth and a per capita increase in energy consumption. To meet the energy needs, fossil fuels (coal, oil, and gas) will play a major part in energy production, even with a projected increase in alternative renewable sources. However, to minimize greenhouse gas emissions, the current power plant systems emphasize the capture of carbon dioxide and subsequent sequestration. Oxy-fuel combustion systems (without the diluent nitrogen gas) would enable recycling of the carbon dioxide to the compressor, use of novel gas turbines, and advanced reuse of CO₂. The U.S. Department of Energy/Office of Fossil Energy is supporting the development of combustion systems that replace air with nearly pure oxygen, with the goal of achieving a coal-based power system with near zero emissions. For this purpose, turbines and combustor technologies that use pure oxygen in fuel combustion are being developed. The major advantage of combustion under pure oxygen is the potential for the separation and capture of CO₂ and for power system efficiencies in the range of 50–60%.

The presence of H₂O/CO₂ and trace constituents such as sulfur and chlorine in the gas environment and coal ash deposits, including alkalis at the operating temperatures and pressures, can have adverse effects on the corrosion and mechanical properties of the structural alloys in these plants [1]. The concentrations of CO₂, SO₂, and H₂O in oxy-fuel environments are higher than those under air combustion conditions, because of the absence of airborne N₂. Various investigators have studied the alloy corrosion behavior of steel materials in H₂O and CO₂ environments. Increasing steam content can lead to breakaway oxidation [2]. Carburization was observed when stainless steels were exposed to CO₂ at elevated temperature [3,4]. High CO₂ and H₂O concentrations also assisted in partially oxidizing SO₂ to SO₃ [5]. The concentration of SO₃ in the oxy-fuel environment is typically three or more times higher than that under the air combustion condition. This SO₃ can react with steam to form sulfuric acid. Subsequently, higher SO₃ and H₂O concentrations can lead to higher concentrations of corrosive H₂SO₄ under oxy-fuel conditions. Thus, the response of structural and turbine materials in simulated oxy-fuel environments needs to be evaluated to select materials that have adequate high-temperature mechanical properties and long-term environmental performance.

Sulfur can also be transported through oxide scales on alloys and increase their oxidation rates in coal plants. Radiometric analysis has indicated that the inward transport of sulfur is primarily due to the SO₂/S₂ molecular transport. Sulfides were observed in the inner scale layer [6]. However, the effect of sulfur on corrosion in an oxy-fuel system has scarcely been studied [7]. Coal ash can also lead to severe corrosion at high temperature [8–20].

The corrosion of alloys in the oxy-fuel and ash environments has been little studied [21–24]. While coal power plants are designed to be operated for a long time, recent studies usually

report only short-term test results regarding alloy performance. If the oxidation follows a parabolic law, short-term test results can provide useful information. However, if the oxide scale on the alloy breaks down after the incubation period, the corrosion rate can dramatically increase. Then, short-term test results below the incubation time will not be sufficient to establish the alloy performance. This paper is the result of a study of the long-term corrosion performance of alloy materials in oxy-fuel environments.

Ash corrosion is more severe than other types of corrosion in the environment of coal power plants, especially if the operating temperature is increased to improve the efficiency. Ash corrosion is also more complicated than other types of corrosion since it involves solid, liquid, and gas phases. In addition, it involves multiple corrosive gases: O₂, SO₂, SO₃, CO₂, H₂O, HCl, and Cl₂. Ash corrosion could also be localized, and this localization dramatically increases the uncertainty in determining the corrosion rate. Therefore, it is a great challenge to adequately characterize and explain the ash corrosion. In our previous studies [18,21–23], we performed corrosion tests to evaluate the performance of structural alloys in the presence of simulated U.S. Eastern coal ash, consisting of alumina, silica, and iron oxide along with sodium and potassium sulfates. Detailed results were presented on weight change, scale thickness, internal penetration, microstructural characteristics of the corrosion products, and cracking of scales after exposure of the alloys at 750 °C. Tests were also conducted in environments with and without steam in oxy-fuel gas atmospheres. Results from these tests were also used to address the role of steam in the long-term corrosion performance of alloys.

The U.S. Eastern coal is rich in iron oxide, while U.S. Western coal is rich in calcium oxide. The calcium content in ash is an important parameter that affects alloy corrosion. Typically, the calcium oxide contents in the ash of U.S. Western coal from the Powder River Basin (PRB) and Beulah-Zap coal zone are much higher than those in the ash of U.S. Eastern coal. It is thus necessary to understand the effect of the calcium content on ash corrosion. Moreover, CaCO₃ is also added to furnaces in coal plants to absorb SO₂ and reduce SO₂ emissions. It is also necessary to understand the reactions of CaO and CaCO₃ with alloy materials. For this paper, we investigated the effect of calcium on Ni-based alloy corrosion in ash with and without calcium oxide. The effect of calcium on Fe-based alloy corrosion in ash will be reported in another paper.

2. Experimental procedure

2.1. Alloy composition

The compositions of the alloys selected for the study are listed in Table 1. High-Ni alloys (333, 617, 625, 602CA, 230, 693, 740, and 718) were included in the study, especially for application in superheaters at temperatures above 700 °C. Apart from fire-side and steam-side corrosion resistance, the alloys selected for applications in steam superheaters and reheaters should possess adequate strength at elevated temperatures for the duration of service.

Table 1
Nominal composition (in wt%) of alloys selected for corrosion study.

Material	C	Cr	Ni	Mn	Si	Mo	Fe	Other
333	0.05	25.0	45.0	–	1.0	3.0	18.0	Co 3.0, W 3.0
617	0.08	21.6	53.6	0.1	0.1	9.5	0.9	Co 12.5, Al 1.2, Ti 0.3
625	0.05	21.5	Bal	0.3	0.3	9.0	2.5	Nb 3.7, Al 0.2, Ti 0.2
602CA	0.19	25.1	62.6	0.1	0.1	–	9.3	Al 2.3, Ti 0.13, Zr 0.19, Y 0.09
230	0.11	21.7	60.4	0.5	0.4	1.4	1.2	W 14, Al 0.3, La 0.015
693	0.02	28.8	Bal	0.2	0.04	0.13	5.8	Al 3.3, Nb 0.67, Ti 0.4, Zr 0.03
740	0.07	25.0	Bal	0.3	0.5	0.5	1.0	Co 20.0, Ti 2.0, Al 0.8, Nb + Ta 2.0
718	–	19.0	52.0	–	–	3.0	19.0	Nb 5.0, Al 0.5, Ti 0.9, B 0.002
WASP	0.02	20	Bal	0.5	0.75	4.3	2.0	Co 12–15, Ti 2.6–3.25, Al 1.0–1.5, Zr 0.1

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