



Full Length Article

Curtailing the generation of sulfur dioxide and nitrogen oxide emissions by blending and oxy-combustion of coals



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ABSTRACT

This research examined the reduction of combustion-generated sulfur dioxide and nitrogen oxide emissions from a pulverized high-sulfur bituminous coal by co-firing it with a lignite coal and by substituting air with O₂/CO₂ gases, of compositions pertinent to dry once through oxy-fuel combustion conditions. Oxygen mole fractions were in the range of 21–30%. The neat bituminous and lignite coals as well as a 50–50 wt% blend thereof were burned in a laboratory-scale electrically-heated drop-tube furnace under fuel-lean conditions. Coal particles were in the size range of 75–90 μm and the furnace was operated at 1400 K. Results showed that the SO₂ emissions from the lignite coal were drastically lower than those from the bituminous coal, whereas the NO_x emissions were only mildly lower. Co-firing the high-sulfur bituminous coal and the low-sulfur lignite coal reduced the SO₂ emissions, under both air and oxy-combustion conditions, to values well-below those predicted by linear interpolation of the respective emissions of the two neat coals. This observation, in conjunction with ash analysis shows that the alkali-rich ashes of the lignite coal acted as sulfur sorbents for the copious SO₂ emissions of the bituminous coal. This behavior was contrasted to SO₂ sorption when the bituminous coal was co-injected with calcium- and sodium-based sorbents at analogous alkali to sulfur ratios. Co-firing of the coals reduced the NO_x emissions only mildly, whereas the substitution of the background N₂ gas with CO₂ resulted in much more extensive NO_x reductions.

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1. Introduction

Coal is abundant, affordable and easy to transport, store and use. It is also insusceptible of geopolitical tensions and, as a result, it plays a vital role in electricity generation worldwide. Coal-fired power plants currently generate about 40% of global electricity [1], whereas in some countries, such as Australia, India, China, Kazakhstan, and Poland, coal generates more than 50% of their electricity needs [2]. In the US, 39% of the electric power in 2014 was generated by burning coal [3] which, however, produced 1553 million metric tons of CO₂ [4]. In addition to emitting carbon dioxide, power plants also emit air pollutants, such as particulates and oxides of sulfur (mostly SO₂ and small amounts of SO₃) and oxides of nitrogen (mostly NO and small amounts of NO₂, the combined amount of which is termed NO_x). SO₂ and SO₃ are health-hazardous gases and they contribute to the formation of submicron acidic particulates that can penetrate into human lungs and even be absorbed by the bloodstream. They also contribute to the

formation of acid rain, which is detrimental to forests and crops, and acidifies lakes. In conventional coal combustion effluents, SO₃ accounts for only a few percent (0.5–2.5%) of the primary emissions of sulfur oxides [5]. In oxy-coal combustion effluents the volume fraction of SO₃ is significantly higher (4–8%) [5]. NO_x are also health hazardous gases and cause photochemical smog, ground-level ozone and contribute to acid rain. In typical combustion effluents NO₂ accounts for only a few percent of the primary emissions of nitrogen oxides [6]. In recent years, annual SO₂ emissions from the electric power sector in the US amounted to approx. 4.3 million metric tons and those of NO_x amounted to approx. 2.1 million metric tons (EIA 2012 data [7]).

Among various methods for controlling the aforementioned pollutants in coal-based power generation is coal blending and oxy-combustion of coal. The former method has been proven effective in reducing SO₂ emissions of high sulfur coals and the latter method, while it primarily targets carbon capture and sequestration and it also reduces NO_x emissions.

Coal blending is designed to meet technical, environmental and economic objectives. The latter are aimed to produce energy at the lowest cost; in other words, blending is designed to take advantage

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Table 1
Chemical compositions (wt%) and energy contents (MJ/kg) of the two selected US coals.

Penn-state coal bank code → Rank/fuel source	DECS-24 Bituminous Illinois #6 Macoupin (I)	DECS-11 Lignite Beulah, North Dakota (B)
<i>Proximate analysis</i>		
Moisture (%)	13.2	33.4
Volatile matter (%)	35.4	37.4
Fixed carbon (%)	39.7	22.9
Ash (%)	11.6	6.4
<i>Ultimate analysis (dry)</i>		
Carbon (%)	66.1	66.2
Hydrogen (%)	4.6	4.0
Oxygen (%) (by diff.)	9.9	18.6
Nitrogen (%)	1.1	0.9
Sulfur (%)	5.5	0.7
Calcium (%)	0.53	1.8
Sodium (%)	0.11	0.66
Potassium (%)	0.18	0.05
Magnesium (%)	0.05	0.56
Ash (%)	13.4	9.6
Heating value (MJ/kg)	28.2	25.7

of the most available cost-effective fuel within the given constraints of an existing boiler, kiln, or process heat generator [8]. Coal blending has been shown to reduce SO₂ emissions [8,9]. Recent work in this laboratory [10,11] has shown that co-firing blends of high sulfur-content pulverized bituminous coals with low-sulfur high-alkali content low-rank coals (of the lignite and sub-bituminous ranks) resulted in beneficial synergisms. These synergisms reduced the SO₂ emissions beyond values that were predicted based on the blending proportions of the parent coals. Coal blending can also have a positive effect in reducing the NO_x emissions, depending on the combustion characteristics of the blends in particular boilers [8,10–12]; however, such reductions are far less pronounced than those of SO₂.

Burning coal in oxygen and dry recycled gases, which mostly contain CO₂, instead of air is termed oxy-coal combustion and it is considered to be a “clean-coal” technology as it facilitates capture and sequestration of CO₂ from power plants [13]. Abraham et al. [14] first suggested the idea of combustion in O₂/CO₂ environment to provide a CO₂-rich flue gas for use in the oil extraction industry. Burning coal in mixtures of O₂/CO₂ gases is different from burning it in air, as it affects the coal particle ignition and combustion characteristics, heat transfer and evolution of combustion-generated pollutants [5,15–20]. This is due to the different physical and chemical properties of the N₂ and CO₂ gases, such as density, heat capacity, molecular diffusivity, and radiative absorptivity [21].

Oxy-coal combustion can be performed either by recycling cleaned flue gases, upon removal of major pollutants (e.g., SO₂, NO_x and ash), or by recycling uncleaned or partially-cleaned flue gases (only ash removed) back into the furnace. Whereas the former approach is more costly, the latter is more problematic for the operation and longevity of a boiler. Much research has been done on the emissions of both NO_x and SO₂ from conventional and oxy-combustion of coal; however, pertinent reports on oxy-combustion of coal blends are hard to find.

When cleaned and dried flue gases are recirculated to the boiler and combustion of coal takes place in O₂/CO₂ environments (akin to once-through combustion [22]), then SO₂ emissions are not significantly different from those generated in air-fired combustion [15,16,23], even at different O₂/CO₂ ratios. Small SO₂ reductions under oxy-combustion conditions can be attributed to enhanced SO₃ formation and subsequent sulfur retention in ash [15]. When uncleaned or partially-cleaned flue gases are recirculated to the boiler then combustion of coal takes place in O₂/CO₂ environments in the presence of contaminants, in such cases it has been reported that SO₂ emissions can be reduced as compared to coal combustion

in air [5,24]. It has also been reported that the SO₂ emissions depend on the fuel and air proportions (expressed as the equivalence ratio, ϕ , which is defined in an ensuing section) and are almost independent of furnace temperature and O₂ concentration [25].

When cleaned flue gases are recirculated to the boiler and combustion of coal takes place in O₂/CO₂ environments, NO_x emissions are significantly lower than those generated in air-fired combustion [16,26]. Such a decrease in NO_x emissions in O₂/CO₂ is primarily due to the absence of nitrogen, which deactivates the “thermal-NO_x” generation by the Zeldovich mechanism [27]. Hence, in this case “fuel-NO_x” remains the foremost generation pathway. Increasing the oxygen partial pressure in oxy-combustion increases the combustion temperature [18,19,28] and has some effect on the “fuel-NO_x” formation [15,16].

When un-cleaned or partially-cleaned flue gases are recirculated to the boiler and combustion of coal takes place in O₂/CO₂ environments in the presence of contaminants, then NO_x emissions from the boiler can be further reduced [5] as compared to the previous case, because of re-burning and other beneficial mechanisms that take place in the boiler. It has been reported that the NO_x emissions depend on the fuel to air proportions (expressed as the equivalence ratio, ϕ) [23,25,29] and, also, depend on the furnace temperature and, thus, on the O₂ concentration [30]. Okazaki and Ando [31] mentioned three main reasons that the NO_x decreases in oxy-combustion with uncleaned recycled gases: (1) reduction of recycled NO in the combustion chamber, (2) the interaction between recycled NO and fuel-N and (3) the effect of increasing CO₂ concentration. Other researchers [22,32–38] also showed similar reduction trends in NO_x emissions from oxy-coal combustion.

In this work the techniques of coal blending and simulated dry and cleaned oxy-combustion were used to reduce SO₂ and NO_x emissions from burning coal. The oxy-combustion conditions were simulated in the laboratory by replacing the background nitrogen gas in a drop-tube furnace with carbon dioxide bottled gas. Whereas in previous work in this laboratory various coals were co-fired [10,11] to determine suitable combinations that lower SO₂ emissions, this work concentrated on a most effective set of coals: a high-sulfur and low-alkali bituminous coal and a low-sulfur and high-alkali lignite coal. Whereas atmospheric air was used as the furnace gas in previous work [10,11], herein bottled gases were used in all experiments to supply the furnace and ensure the comparability of results. The following mole fractions were used: 21% O₂/79% N₂, 21% O₂/79% CO₂ and 30% O₂/70% CO₂. Experimental SO₂ emissions were interpreted in light of thermody-

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