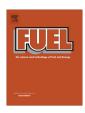


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Oxyfuel technology: NO reduction during oxy-oil shale combustion



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HIGHLIGHTS

- First investigation on NO reduction under oxy-oil shale combustion.
- Actual situation was simulated by injecting NO through the burner (SR NO).
- The SR NO reduction efficiency is affected by the media and mode of firing.
- The SR NO is reduced efficiently (60–70%) during unstaged combustion.
- The reduction of the SR NO is more efficient with staging (100%).

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ABSTRACT

Nitrogen oxides are one of the major environmental problems arising from fossil fuels combustion. Oxyfuel combustion is one of the most promising clean coal technologies for pulverized fuel-fired power plants to control and avoid CO₂ emissions; with this technology NO_x emissions are significantly reduced.

Due to the importance of oil shale utilization in Jordan and all over the world, this study continues with our previous work on oil shale combustion. Unstaged and staged air-firing and oxyfuel combustion were carried out using Jordanian oil shale collected from El-Lajjun area. The reduction of simulated recycled NO has been investigated in a 20 kW vertical reactor. The actual situation has been simulated by injecting NO in the reactor through the burner. It was found that the simulated recycled NO reduction efficiency is affected by the media and the mode of firing. The simulated recycled NO is reduced efficiently (60-70%) during unstaged oxyfuel combustion as well as air-firing of El-Lajjun oil shale. The reduction of the injected NO is more efficient with staging compared to unstaged combustion mode for both air-firing and oxyfuel combustion. The reduction of the injected NO for combustion in air ranges from 61% (without staging) to 100% (with staging) whereas for combustion in 27% O₂/73% CO₂, it ranges from 57% (without staging) to 100% (with staging). Moreover, El-Lajjun oil shale ashes are characterized as being high in CaO and SiO₂, low in Al₂O₃, SO₃, and P₂O₅ and with minor amounts of F₂O₃, MgO, Na₂O, K₂O, and TiO₂.

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1. Introduction

Oil shale is one of Jordans' future energy sources. As confirmed by geological surveys, the existing oil shale reserves cover more than 60% of Jordan's territory. Its large deposits are widely distributed all over the country, particularly in the central region [1]. Oil shale in Jordan and elsewhere faces many obstacles that hinder its utilization. Oil shale is employed in China, Estonia and Russia to yield shale oil through retorting processes and used for electric power generation by direct combustion. The inconsistency and the variety in composition of raw oil shale complicate the

mechanism of combustion. Moreover, the high content of inorganic materials, mainly carbonates, which could reach values greater than 55% of its content is one of these obstacles. The use of existing technologies for oil shale combustion creates serious environmental and health problems. For commercial use of oil shale deposits in the world, a significant improvement of waste removal and recycling system is needed [2].

Fossil fuel burning for power and heat generation and the resulting release of carbon dioxide into the atmosphere has been identified as the major reason for the man made climate change. One of the several technologies developed for ${\rm CO_2}$ capture and storage is the oxyfuel technology. The idea of oxyfuel technology is to combust the fuel in an enriched oxygen environment using almost pure oxygen diluted with recycled flue gas consisting of ${\rm CO_2}$ and/or ${\rm H_2O}$. Part of the flue gas consisting mainly ${\rm CO_2}$ is recycled

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back to the furnace to maintain the temperature and heat flux profiles, and to entrain the pulverised fuel. The concentration of carbon dioxide in the exhaust gas is significantly increased and the recovery of CO_2 becomes feasible with a lower-efficiency penalty [3]. Fundamentally, because of the variation in the oxidant and consequently the in-furnace gas environment, as compared to the conventional air-firing, oxyfuel combustion has impacts on the combustion process as well as other processes such as heat transfer.

An important secondary benefit of oxyfuel combustion is the reduction of nitrogen oxide (NO_x) emissions. During oxy-coal combustion, it is found that NO_x emissions generated per unit energy are reduced as it is re-circulated through the flame compared to air-firing [4-11]. Yamada et al. [12] reported that NO_x generated during oxy-coal combustion has a concentration one and half times higher than during air-firing. The amount of emission downstream the flue gas recycle location in mg/MJ (NO_x emission rate) is however approximately one third compared to air-firing. Croiset et al. [13] registered higher NO_x emission rate during air-firing when compared to oxy-coal combustion with oxygen concentration up to 35 vol.%. The reduction of recycled NO has also been observed during combustion with direct injection of O₂ [14]. Croiset et al. [15] observed a decrease in NO_x emission rate by 40–50% with flue gas recycling compared with once through O_2/CO_2 combustion. The reduction was associated with re-burning of NO_x recycled back into the furnace.

Park et al. [16] studied the fuel-N conversion during heterogeneous reaction of bituminous coal char with O_2 , CO_2 and H_2O over a broad range of conditions. They reported that char-N is converted entirely into N_2 when char reacts with CO_2 ; into N_2 and NO when char reacts with O_2 ; and into HCN, O_2 , into O_2 when char reacts with O_2 and O_2 when char reacts with O_2 and O_2 and O_3 when char reacts with O_3 and O_4 when char reacts with O_2 and O_3 and O_4 when char reacts with O_2 and O_3 and O_4 environments. NO was injected through the burner in a O_4 wertical reactor. It was found that the reduction of injected O_4 NO on Klein kopje char produced in O_4 environment is O_4 higher in O_4 combustion than in O_4 combustion.

In our previous paper [18] the feasibility of oil shale combustion under oxyfuel conditions was investigated using a 20 kW vertical reactor at a combustion temperature of $1200\,^{\circ}$ C. In addition, NO_x emission was also found to be lower and can be reduced efficiently by adopting staged combustion technology under oxyfuel conditions as well as air-firing; however, the oxyfuel investigations were carried out without flue gas recirculation. Since no NO_x recycling experiments was done at that stage, the motivation for this work is to investigate in more detail the reduction of recycled NO during oil shale combustion under oxyfuel conditions by simulating the actual situation. This work is the first of its kind handling oil shale combustion. Experimental results on a simulated recycled NO reduction with both air-firing and O_2/CO_2 combustion is reported.

2. Experimental

Oil shale sample used in this study was obtained from El-Lajjun area in Jordan. One hundred kg of the collected sample was crushed using a jaw crusher and then milled using an open hammer mill. Detailed analysis of the oil shale sample (batch 2013) was carried out at the Institute of Combustion and Power Plant Technology (IFK), University of Stuttgart. Proximate and ultimate analyses, heating value, and the size distribution of the sample are listed in Table 1. Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) was used to measure ash main (DIN 51729-11) and minor elements (DIN 22022-(1-6)), the data are reported in Tables 2 and 3, respectively.

Table 1Proximate, ultimate analyses and size distribution of El-Lajjun oil shale.

Proximate analysis Water (ar, %)¹ Ash (wf, %)²	1.08 54.20
Volatile (waf, %) ³	99.06
Fixed carbon (waf, %) ⁴	0.85
Ultimate analysis	
C (waf, %)	55.68
H (waf, %)	4.27
N (waf, %)	0.87
S (waf, %)	8.30
O (diff, %) ⁴	30.88
LHV (waf, kJ/kg)	19585.15
Size distribution	
$D_{10} (\mu m)^5$	1.25
D ₅₀ (μm)	9.86
D ₉₀ (μm)	35.3

- ¹ As received.
- Water free.
- ³ Water ash free.
- ⁴ By difference.
- ⁵ Di: represent that i% by volume of the particles that have a diameter equal to or less than Di.

Table 2 Ash main elements contents of El-Lajjun oil shale.

Ash main elements (oxide form)	(wf, %)
Al_2O_3	5.70
BaO	0.007
CaO	44.50
Fe_2O_3	1.65
K ₂ O	0.561
MgO	1.076
Na ₂ O	0.009
Mn_2O	0.193
$P_{2}O_{5}$	3.67
SO ₃	16.44
SiO ₂	25.85
SrO	0.151
TiO ₂	0.189

Table 3Ash trace elements contents of El-Lajjun oil shale.

Ash trace elements	(mg/kg)
As	11.5 ± 0.3
Ba	52.7 ± 1.5
Cd	49.5 ± 0.5
Cr	364 ± 1
Cu	73.5 ± 3
Mn	23.3 ± 2.8
Mo	235 ± 9
Ni	198 ± 3
Hg	n.b.(0.085 ± .016)
Pb	0.555 ± 0.181
Sb	10.2 ± 5.4
Se	34.6 ± 1
Sr	824 ± 6
V	167 ± 4
Zn	599 ± 18

El-Lajjun oil shale combustion was performed in a 20 kW vertical furnace. Details of the furnace had been described previously [18]. Here only essential and new information is presented. Experiments were conducted in a vertical, cylindrical, top fired furnace. The burner used in this study was a jet burner that was not optimized for low NO_x emissions.

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