



Influence of oxy-fuel combustion of Ca-rich oil shale fuel on carbonate stability and ash composition



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HIGHLIGHTS

- This study provides results of oil shale combustion in a batch reactor applying oxy-fuel conditions.
- Elevated CO₂ levels and inhibited carbonate decomposition rate have effect on SO₂ binding and stable anhydrite formation.
- Content of calcite in remaining ash increases with increasing CO₂ levels and decreased firing temperatures.
- Carbonate decomposition is delayed and composition of ash is changed in oil-shale firing at elevated CO₂ levels.
- Formation of secondary Ca–Mg silicate phases is directly proportional to firing temperature.

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ABSTRACT

Oil shale is an unconventional low-calorific-value fossil fuel, the usage of which is increasing due to increasing energy demand. Today, Estonia's oil shale usage is the largest in the world. Approximately 90% of the electricity consumed is produced from Ca-rich oil shale. Most of the oil shale, approximately 12 million tons yearly, is used for power generation utilizing pulverized combustion (PC) and circulating fluidized bed combustion (CFBC) technologies. As a result, Estonia has one of the world's highest CO₂ emission rates per capita. This study provides results of oil shale combustion in a batch reactor applying oxy-fuel conditions. The combustion experiments were performed at temperatures of 800, 850 and 900 °C while varying the N₂, O₂ and CO₂ ratios. In addition to different temperatures and air-composition variation, water vapor was injected into the gas flow entering the furnace to study the water vapor influence on the solid combustion products. Our results indicated that elevated CO₂ levels have a significant influence on the carbonate-decomposition extent and therefore on the SO₂-binding properties.

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1. Introduction

Global concern regarding climate warming and stringent limitations on greenhouse-gas emissions have made the oxy-fuel combustion of solid fuels an attractive potential firing technology. Oxy-fuel combustion is considered the most energy- and cost-efficient of the carbon-capture technologies, and it therefore may reduce the level of environmental harm that results from energy production [1–7]. Oxy-fuel technology applies fuel combustion in an O₂–CO₂ environment resulting in flue gas with a CO₂ concentration high enough for efficient CO₂ sequestration and subsequent storage [8,9].

Previous studies have suggested that the chemical and mineralogical composition of the ash forming during the combustion of

various coals and lignite do not differ significantly between air and oxy-fuel combustion conditions [10–12]; however, the ash deposition rate, particulate formation (grain-size) and proportions of minerals are reported to vary under oxy-fuel conditions [12–14].

The possible influence of oxy-fuel combustion on ash formation and composition would result from the high CO₂ partial pressures in the high-temperature mineral transformation processes [15–17]. In coal and/or biomass solid fuels, the inorganic (mineral) part of the fuel is low (<20 wt%) and dominated by (alumo)silicate phases (including quartz) that are typically thermally resistant/unreactive.

In contrast, oil shales that are low-quality, organic-rich sedimentary rocks [18] and contain a considerable amount of mineral matter (usually 30–90 wt%, e.g., [19]) can be more sensitive to the conditions present in oxy-fuel-combustion environments.

Oil shales are generally rich in bituminous organic matter [20] and are processed worldwide as an energy resource in a number

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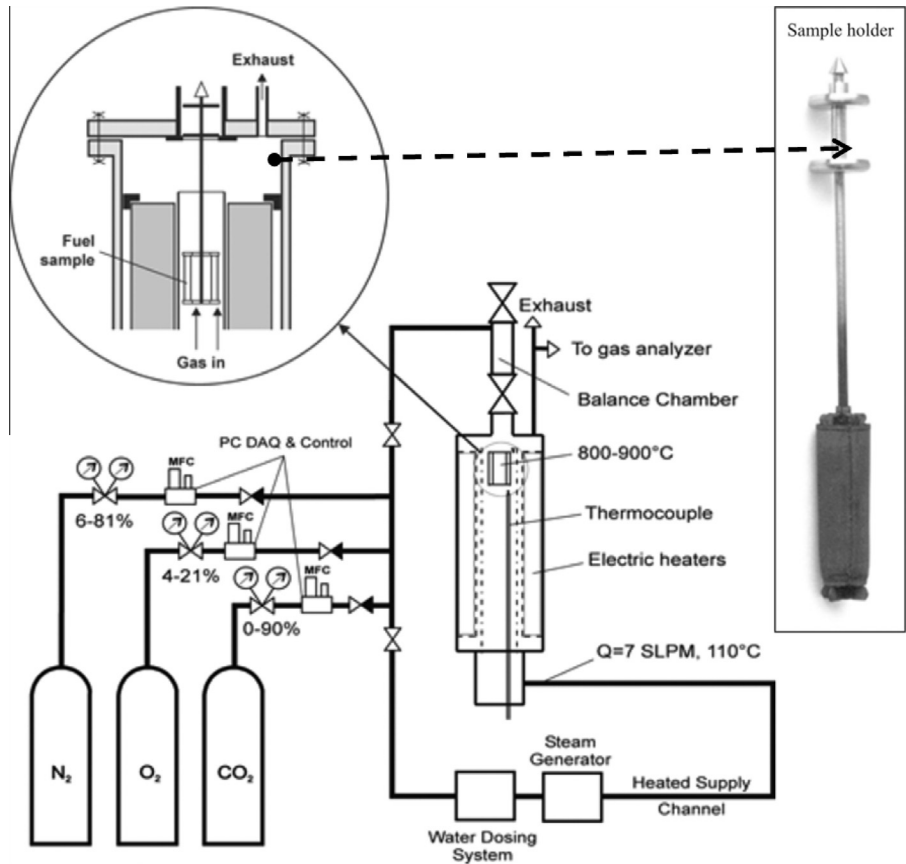


Fig. 1. Simplified schematic of the batch reactor and a sample holder.

Table 1
Firing temperatures and environmental conditions for the oil-shale combustion experiments, vol%.

Gas/temperature	800 °C	800 °C	800 °C	850 °C	850 °C	850 °C	900 °C	900 °C	900 °C
N ₂	79	81	6	79	81	6	79	81	0
O ₂	21	4	4	21	4	4	21	4	4
CO ₂	0	15	90	0	15	90	0	15	96
H ₂ O	0/8/15	0/8/15	0/8/15	0/8/15	0/8/15	0/8/15	0/8/15	0/8/15	0/8/15

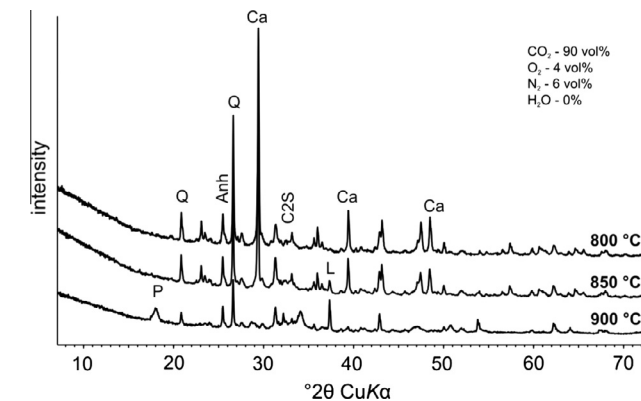


Fig. 2. Representative X-ray diffraction patterns of the collected ash. Experimental conditions are indicated in the figure. Anh – anhydrite, Ca – calcite, C2S – dicalcium silicate (belite), L – lime (CaO_{free}), and P – portlandite.

of countries, including China, Brazil, Jordan and Estonia, although large known reserves also exist in the U.S.A., Australia, Russia and elsewhere [21]. However, the increasing needs for energy

(esp. in developing countries), the depletion of easily accessible oil reserves and the concurrently increasing oil prices have significantly raised the interest in oil-shale mining and processing in the last few decades.

A significant drawback of oil-shale usage is its large CO₂ footprint, which is due to the low calorific value of the fuel as well as the composition of the mineral matter. A considerable portion of the oil-shale deposits worldwide are found in shallow marine or lacustrine sediments dominated by carbonate-mineral assemblages. The thermal dissociation of Na-, Ca- and/or Ca–Mg-carbonate phases liberates large amounts of CO₂ and oxidizes organic carbon. As a result, oxy-fuel combustion technology and consequent CO₂ sequestration would be specifically demanded for oil-shale processing. On the other hand, the thermal decomposition of carbonate minerals is sensitive to the CO₂ partial pressure [22], and in contrast to the case of coal-type fuels, combustion air enriched with CO₂ can influence the mineral-transformation pathways and rates in oil-shale combustion. Moreover, the CO₂ levels can influence the melting of silicate phases and the formation of secondary Ca-silicate minerals that have a significant influence on the physical and chemical properties of the ash, such as the pozzolanic properties of the fine fractions of ash, and therefore the potential usability of the ash waste as a binder/cement additive.

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