



CO₂ capture by carbide slag calcined under high-concentration steam and energy requirement in calcium looping conditions



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HIGHLIGHTS

- O₂/H₂O combustion is proposed to provide heat for calciner in calcium looping.
- High-concentration steam in O₂/H₂O combustion lowers needed temperature of calciner.
- Carbide slag calcined under high-concentration steam retains high cyclic CO₂ uptake.
- Calcined carbide slag under high-concentration steam possesses porous structure.
- Calcination in high-concentration steam during calcium looping is energy-saving.

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ABSTRACT

In this work, calcination under high-concentration steam that can be implemented by O₂/H₂O combustion was proposed to replace high-concentration CO₂ that usually was implemented by O₂/CO₂ combustion in calcium looping. CO₂ capture performance of the carbide slag, an industrial solid waste from chlor-alkali plants, was investigated in a dual fixed-bed reactor under high-concentration steam calcination condition during the calcium looping cycles. Calcined under high-concentration steam (95%) atmosphere, the carbide slag can be completely and quickly decomposed at 800 °C, which is 150 °C lower than the calcination temperature under high-concentration CO₂ (100%) atmosphere. The carbonation conversions of the carbide slag calcined under high-concentration steam condition after 1 and 10 cycles are about 42% and 36% higher than those calcined under high-concentration CO₂ condition, respectively. This is because when the carbide slag is calcined under high-concentration steam, the relatively smaller CaO grains and more porous structure are generated, which are beneficial for CO₂ capture by carbide slag. Due to the low calcination temperature and the high CO₂ capture capacity of the carbide slag, the energy requirement in the calciner to capture per mole CO₂ under high-concentration steam condition is lower than that under high-concentration CO₂ condition. High-concentration steam calcination in place of high-concentration CO₂ calcination improves the CO₂ capture efficiency from 0.68 to 0.88 and saves a quarter of the energy consumption in the calciner for capturing per mole CO₂ in the calcium looping system, when the ratio of recycled carbide slag flow rate to CO₂ flow rate is 2 and the sorbent make-up ratio is 0.09. Thus, it is reasonable to consider O₂/H₂O combustion as an alternative energy-supply way of O₂/CO₂ combustion for the calcium looping of carbide slag.

1. Introduction

Excessive emissions of carbon dioxide (CO₂) are seen as main reasons resulted in serious climate changes [1]. As main CO₂ emission sources, coal-fired power plants are considered as one of the key points to mitigate CO₂ emissions [2]. Calcium looping is deemed as one of the most promising technologies for large-scale CO₂ capture due to its low

operation cost, lower efficiency penalty comparing to other post-combustion CO₂ capture technologies and possibility of integrating with other industrial processes [3–7]. Calcium looping includes a carbonation reaction between CaO and CO₂, where CO₂ in the flue gas is captured by CaO, and a calcination reaction in oxy-fuel combustion condition, where concentrated CO₂ is generated and the sorbent is regenerated. CO₂ (> 95%) released from the calciner can be directly

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Nomenclature

P_{eq}	CO ₂ partial pressure in the reactors (atm)
T	thermodynamic equilibrium temperature (K)
N	number of calcination/carbonation cycles
X_N	carbonation conversion of the sample after N cycles (%)
m_0	mass of the initial CS sample (g)
m_N	mass of the carbonated sample after N cycles (g)
$m_{cal,N}$	mass of the calcined sample after N cycles (g)
M_{CaO}	molar mass of CaO (g/mol)
M_{CO_2}	molar mass of CO ₂ (g/mol)
A	content of CaO in initial carbide slag (wt.%)
F_0	make-up flow rate of CS (mol/s)
r_N	mass fraction of carbide slag make-up after N cycles
F_R	flow rate of recycled sorbent (mol/s)
$F_{R,inert}$	flow rate of inert impurities in the recycled sorbent (mol/s)

X_{ave}	average carbonation conversion of CS in the carbonator
X_r	residual conversion of the carbide slag after infinite carbonation cycles
k	fitting constant in Eq. (5), which indicates the decay coefficient
E_{CO_2}	CO ₂ capture efficiency (%)
F_{CO_2}	CO ₂ flow rate entering the carbonator (mol/s)
$c_{p, CaO}$	heat capacity of CaO calculated according to Ref. [42] (kJ/mol K)
$c_{p, CaCO_3}$	heat capacity of CaCO ₃ (kJ/mol K)
$c_{p, inert}$	heat capacity of inert, by weighed average (kJ/mol K)
H_1	energy for heating cycling and fresh CS to the calcination temperature (kJ/mol)
H_2	energy for CaCO ₃ decomposition in the cycling and fresh CS (kJ/mol)
ΔH	decomposing heat of CaCO ₃ (kJ/mol)
H_{cal}	total energy consumption in the calciner (kJ/mol)

sealed or recycled as resources and the regenerated CaO is recycled during calcination/carbonation cycles [8]. Calcium looping demonstration plants such as a 1.7 MW and 1 MW pilot plants have been built in Spain [9] and Germany [10], respectively.

During the calcium looping process, the carbonation reaction of CaO is exothermic, but the calcination reaction of CaCO₃ is endothermic, thus additional energy is required for the calciner. The energy required in the calcination is supplied by O₂/CO₂ combustion of coal, so the flue gas from the calciner is almost pure CO₂ [8,11]. For complete and quick calcination of CaCO₃, the temperature of the calciner is usually higher than the thermodynamic equilibrium temperature, which depends on the CO₂ partial pressure in the calciner. The relation between thermodynamic equilibrium temperature and the CO₂ partial pressure was proposed by Baker [12] as follows:

$$\lg P_{eq} = 7.079 - \frac{8308}{T} \quad (1)$$

Usually, calcination temperature is above 900 °C (e.g. 950 °C) under high-concentration CO₂ atmosphere (e.g. 90%) [13]. However, such high calcination temperature and high-concentration CO₂ atmosphere intensify the sintering of calcium-based sorbents, which leads to a sharp decay in CO₂ capture capacity with the number of cycles [13–21]. Grasa et al. [13] found high calcination temperature intensified sintering of limestone, especially at 950–1000 °C, and the CO₂ capture capacity of limestone degenerated quickly. The additives such as manganese [14,15], magnesium [16,17], aluminum [18], zirconium [19], titanium [20] or other inert substances [21] are favorable for improving the sintering resistance of the calcium-based sorbent under the mild calcination condition, but they only show a small improvement in the sintering resistance under the severe calcination condition (the high temperature and high-concentration CO₂). Since the severe calcination condition including high temperature and high-concentration CO₂ intensifies the sintering of the calcium-based sorbent, it is necessary to seek a milder calcination condition for the calcium looping process. Decreasing the CO₂ partial pressure in the calcination is an effective method to reduce the calcination temperature and the sintering of the calcium-based sorbent.

Recently, O₂/H₂O combustion is gradually becoming a hotspot,

which is a kind of new oxy-fuel combustion technology for CO₂ capture from power plants [22]. The schematic of O₂/H₂O combustion process is shown in Fig. 1 [23]. The final main products of coal in O₂/H₂O combustion are high-concentration steam and low-concentration CO₂. Jin et al. [24] reported that O₂/H₂O combustion exhibited better performance in both thermodynamic and economic aspects than O₂/CO₂ combustion and they also found that the cost of electricity decreased by about 6.62%, and the net efficiency and the energy efficiency increased by about 0.90% and 1.01%, respectively. If O₂/CO₂ combustion is replaced by O₂/H₂O combustion in the calciner, the calcination temperature may be significantly reduced and the sintering of the sorbents is possibly alleviated due to the relatively low CO₂ concentration. In addition, it is very convenient to separate CO₂ from the mixed gases by simple condensation for subsequent storage and utilization. Therefore, O₂/H₂O combustion maybe appears promising to provide the required energy for the calcination step in the calcium looping cycles.

The fuel combusted under O₂/H₂O atmosphere in the calciner for the calcium looping will generate a high-concentration steam atmosphere. The effect of steam in the calcination process on CO₂ capture by the limestone has been reported. Donat et al. [25] and Champagne et al. [26] studied steam concentration less than 40% in the calciner and found the pore structure existed a shift from smaller to larger pores, which increased subsequent CO₂ capture capacities of the limestone. But the sintering also been observed with the steam injection [26,27]. Kavosh et al. [28] and Wang et al. [29] investigated the effect of high-concentration steam in the calcination in a fluidized bed reactor on CO₂ capture by limestone and they found high-concentration steam increased the rate of CaCO₃ decomposition and shortened the complete decomposition time [28,29]. Although they thought that high-concentration steam can decline the calcination temperature, they did not study CO₂ capture capacity of limestone at lower calcination temperature and the cyclic stability during the cycles. Since the calcination temperature may decrease under the high-concentration steam atmosphere, CO₂ capture performance of the calcium-based sorbent should be studied at this reaction conditions, which has been seldom reported.

Limestone is the most common CO₂ sorbent in calcium looping [4,28–30]. However, the excessive exploitation of limestone mine has already made destruction to natural environment. Carbide slag (CS), as

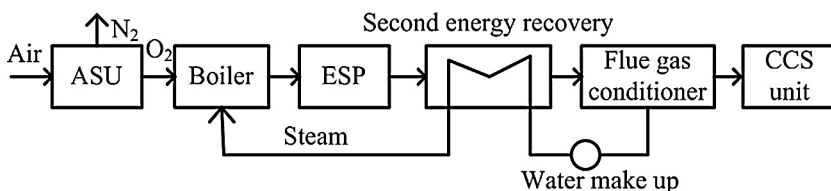


Fig. 1. Schematic of O₂/H₂O combustion process.

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