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Research article

Selective removal of sodium and calcium from low-rank coal – Process integration, simulation and techno-economic evaluation

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ABSTRACT

This paper has addressed the techno-economic feasibility regarding the selective removal of sodium (Na) and calcium (Ca) from low-rank sub-bituminous coal, aiming to reduce the ash slagging and fouling propensity in the pulverized coal-fired boilers. Four novel process integrations were proposed and simulated in Aspen Plus. Both the novel counter-current three-stage water washing process and an acid-water two-stage washing process have proven to improve the ash fusion temperature satisfactorily, reducing the mass fraction of Na_2O in ash from 4.32 wt% to 0.85 and 0.19 wt%, respectively. In addition, the use of acid-water washing removed 12.5% CaO and 19.5 wt% total ash. For the recycle and treatment of wastewater, the water gain is desirable for the use of an evaporator, owing to the dewatering of the initially high-moisture coal (25 wt%) in the centrifugal and the high water recovery rate from the evaporator. However, the good performance of evaporator was counteracted by the considerable capital cost caused by the huge heat transfer area requirement. Instead, the use of reverse osmosis (RO) resulted in a water loss up to 228.4 kg/t coal. Additionally, prior to the RO treatment unit, the recycle and reuse of the unsaturated water for maximum six times and four times for three-stage water washing and acid-water two-stage washing, respectively, was critical in reducing both the water and power consumption. The water consumption dropped to 38.1 kg/t coal and 48.1 kg/t coal for the three-stage water washing and acid-water two-stage washing process, respectively. Both are remarkably lower than 85.0 kg-water/t black coal. In terms of the power consumption, it decreased to ~9.4 kWh/t coal for the three-stage water washing process and further down to 5.8 kWh/t for the acid-water washing case, which was even slightly lower than 6.3 kWh/t for the black coal. Furthermore, the integration of acid-water washing and RO was also demonstrated to be economically viable by its high NPV, IRR and short payback period. Sensitivity analysis indicate that, the original Na content in raw coal is the most influential variable on the water and power consumption of the overall process, followed by the initial moisture content in the raw coal. For a low-rank coal containing > 2150–2520 ppm Na and/or < 19 wt% moisture, the washing process proposed would turn economically unviable compared to the existing black coal washing process. A minimum selling price of 136 RMB/t (−32% deviation) was also necessary to keep both NPV and IRR positive as well as the payback period shorter than the project lifetime.

1. Introduction

Low-rank coal, commonly referred to as brown coal and sub-bituminous coal, contributes to > 50% of the world's coal reserves [1]. It is abundant in regions such as Australia and China [2–3] and provides an economically attractive alternative to high-rank black coal (i.e. bituminous coal and anthracite) for electricity generation. The use of low-rank coal is becoming increasingly important with the ongoing depletion of high-rank bituminous coal. However, low-rank coal boilers are afflicted by severe slagging and fouling inside the boiler caused by its higher content of alkali and alkaline earth metal (Na, Ca, K and Mg) [3–5].

Conventional coal cleaning techniques are exclusively targeted at high-rank coal with a large portion of its inorganic metals present as discrete grains and separate from the coal matrix [6]. As the mineral matter has a larger density than the carbonaceous matrix, it can be removed physically [7]. Variations in surface properties can also allow for separation [8]. However, unlike black coal, sodium (Na) and calcium (Ca) in low-rank coal are deeply and chemically embedded within the coal matrix. Therefore, they cannot be removed using any physical

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approaches based on either density or surface property discrepancy between mineral matter and coal matrix [9]. Instead, chemical leaching or solvent extraction has been examined to remove these two elements from low-rank coal. In particular, research has been focused on the removal of all ash-forming metals to produce ultra-clean coals (UCC) or ash-free hyper-coal (HPC), from both high-rank and low-rank coals [10–13]. However, the corrosive acid/alkali reagents and/or high pressure and temperature employed in existing chemical leaching processes [10,14–16] raise severe environmental concerns, the harsh requirement for equipment, and high capital/operating cost, limiting their advance in practical applications. Solvent extraction is also inappropriate for low-rank coal because of the low carbon yield (< 30 wt %) resulting from its cross-linked carbonaceous structure [10]. For conventional coal-fired boilers, the removal of all ash-forming metals is also unnecessary.

In this study, instead of removing all of the ash-forming metals, only Na and Ca were selectively targeted, considering that these two metals are the most critical triggers for ash slagging and fouling in a pulverized coal-fired boiler [3–5,17]. Moreover, based on the high-water solubility of Na in low-rank coals, the existing water-washing process for black coal was adapted for the low-rank coal. That is, multiple-stage water washing, either with or without acid dosing was employed to wash the low-rank coal, whereas the resultant used water was repeatedly used before it is saturated and then sent to the wastewater treatment unit to remove the inorganic impurities [18]. The innovative characteristics of the new water-washing process are hypothesized from the following three perspectives, 1) the use of water can significantly be reduced by reusing it before it is fully saturated with sodium; 2) the mature existing waste water treatment and recovery techniques, once integrated efficiently with the coal washing process, can further help recover the water cost-effectively; and 3) the integration of acid and water on coal water-washing can significantly increase the extraction yields of both Na and Ca out of coal matrix, according to the following two equations [19–20] where R stands for organic moieties in coal. Additionally, since the original acid is neutralized by Na and Ca, its disposal causes little environmental issues.

$$R - COONa + H^+ \leftrightarrow R - COOH + Na^+ \tag{1}$$

$$Ca(R - COO)_2 + 2H^+ \leftrightarrow 2R\text{–}COOH + Ca^{2+} \tag{2}$$

In this paper, we have conducted process simulations to assess four different scenarios so as to optimize the integration of individual units to produce a washed low-rank coal that has a comparable content of Na and Ca with that in black coal. Furthermore, the ash in the resultant washed low-rank coal is expected to have an ash fusion temperature above the boiler operating temperature, at 1200 °C. Aspen Plus has been employed for the process flow-sheeting. Aspen Process Economic Analyzer (APEA) was further used to perform the economic evaluation based in the context of China. Finally, sensitivity analysis was undertaken to examine the robustness of the overall process including the water and power consumption, and economic feasibility. As far as the authors are aware, such a study has yet to be examined in the literature. The results achieved are expected to promote the deployment of low-rank coal washing in a large industrial and commercial scale in the future.

2. Methodology

2.1. Coal properties

A low-rank sub-bituminous coal, termed Xinjiang Zhundong coal was used for this study. The low-rank coal washing plant was assumed to be located beside a coal mine in Xinjiang, China, which is rich in low-rank coal [2]. Additionally, a reference black coal was tested for comparison. Their properties are tabulated in Table 1. The volatile content (30.2 wt%) in Xinjiang coal indicates that it belongs to sub-bituminous coal [21]. It is much higher than the value of black coal, at 7.8%. The sub-bituminous coal is also characterized by the high moisture of 24.5% and low ash content of 7.8%, compared with black coal reference. The gross heat value of 24.9 MJ/kg shows that this sub-bituminous belongs to the intermediate-heating-value coal [21]. With respect to the particle size distribution of the as-mined Xinjiang coal shown in Table 2, one can see that this coal is very fine with a mass percentage of 49% < 4 mm. One major reason is due to its low-ash content.

Table 1
Compositions of as-received Xinjiang low-rank coal and black coal.

Components	Low-rank coal (wt%)	Black coal (wt%)
Moisture[a]	24.5	9.68
Ash[b]	7.78	22.49
Volatile[b]	30.2	7.78
FC[b]	62.02	69.73
Chlorine[b]	0.09	–
Gross heat value[c]	24.89 MJ/kg	26.93 MJ/kg

[a] Air-dried.
[b] Dried basis.
[c] Dry ash free basis.

Table 2
PSD of as-received Xinjiang low-rank coal and black coal.

Low-rank coal		Black coal	
PSD/mm	wt%	PSD/mm	wt%
< 0.2	8	0–5	30
0.2–0.6	12	5–40	60
0.6–1.0	8	> 40	10
1.0–4.0	21		
4.0–8.0	15		
> 8.0	36		

Table 3 compares the ash compositions of the two coals, which are expressed as the most stable oxide of each element. It is observed that the contents of both Na_2O and CaO in low-rank coal are significantly higher than in black coal, which is in agreement with the past studies [3–5]. In terms of the mode of occurrence of Na and Ca in Xinjiang coal, it has been widely reported that more than half of Na was water-soluble, with the rest being organically bound with carboxylic acid that can be entirely washed away by ammonium acetate [2,10,16,22]. Conversely, the major forms of Ca in low-rank coal vary from one study to another, indicative of its dependence on the coal-forming environment. For instance, Wijaya et al. (2011) found that Ca was equally soluble in water, ammonium acetate and hydrochloric acid, yet no < 35% of Ca was water-soluble due to the presence of their chlorides. However, Ma et al. (2014) claimed that calcite and gypsum were the two dominated forms found in low-rank coal.

2.2. Low-rank coal washing process

The whole process consists of two major components, coal washing, and waste water treatment and recycling. The raw coal first undergoes sieving and crushing to obtain the particles < 4 mm in diameter, before being sent to the washing reactor by a belt conveyer. With respect to the coal washing unit, three options were proposed hereafter: the use of a single stage washing tank shown in Fig. 1(a), the use of three-stage counter-current flow of water against the coal stream in Fig. 1(b), and the combination of acid and water in Fig. 1(c) and (d). With respect to the waste water treatment and recycling, the mixture of washed coal and water from the washing step is firstly separated by a dewatering screen. Subsequently, the wet washed coal is transferred to a centrifuge to squeeze out the remaining water and make the final product that contains around 11% moisture. The resultant water from the centrifuge is mixed with coal fine-water slurry received from the dewatering