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Optimization of leaching conditions for removing sodium from sodium-rich coals by orthogonal experiments

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HIGHLIGHTS

- Leaching treatments were used to selectively remove sodium by some mild agents.
- Leaching temperature was the significant factor in the W-leaching treatment.
- RES showed a volcanic relationship with the concentration of leaching agents.
- The optimal conditions were verified to be efficient and time-saving.
- The optimal conditions had a good universality among sodium-rich coals.

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ABSTRACT

The leaching pretreatment has been adopted to mitigate the ash-related problems encountered during gasification and combustion of sodium-rich coals. However, the wide-spread applications of this technology are limited to produce the ultra clean coal through remove all the ash by some aggressive agents. In the present study, some mild agents, such as deionized water, ammonium acetate solution and diluted hydrochloric acid, were employed to selectively remove troublesome sodium rather than all the ash. The primary leaching conditions such as particle size, temperature, residence time and concentration of agents were optimized by orthogonal experiments for the first time to achieve maximum removal efficiency of sodium (RES). Based on the range analysis, it was found that the significant factor was leaching temperature in the water-leaching (W-leaching) treatment and the RES showed a positive correlation with temperature. However, in both ammonium acetate-leaching (A-leaching) and diluted hydrochloric acid-leaching (H-leaching) treatments, the concentration of agents were found to play the dominant role and RES presented a volcanic relationship with it. Under optimal conditions, the leaching treatments were less time-consuming with improved RES (47.6% for W-leaching, 97.9% for A-leaching and 96.8% for H-leaching). Besides, the properties of coal samples after leaching were determined by a series of analysis methods. The results showed that the leaching treatments could significantly decrease the content of AAEMs and coal ash. Due to the removal of AAEMs, the ash fusion temperatures increased, while both gasification and combustion reactivity decreased obviously. The application to other three sodium-rich coals confirmed that the optimal leaching conditions had a good universality.

1. Introduction

Sodium-rich coals are widely distributed around the world, such as Montana (the United States), Victorian (Australia) and Zhundong (China), which are currently used as the feedstock for power generation [1,2]. However, the high level of sodium in coal has caused a series of excessive problems such as slagging, fouling and corrosion in the power plant boiler [3–6]. Nowadays, the sodium-rich coals can only be used in the specially designed boilers. Moreover, some promising gasification technologies like IGCC cannot directly use sodium-rich coals as well [2]. Thus, for the better utilization of sodium-rich coals, it is very essential to find out some alternative technologies.

A literature review shows that there are two major technological approaches to migrate or solve the sodium-related problems: minerals addition and solution leaching treatment [2,3]. The former is adding Si-Al rich minerals into boiler or gasifier to react with

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Nomenclature

RES	removal efficiency of sodium, %
AAEMs	alkali and alkaline earth metals
LTAs	low-temperature ashes
AFTs	ash fusion temperatures
W-leaching	water-leaching
A-leaching	ammonium acetate-leaching
H-leaching	hydrochloric acid-leaching
1#/R	1# raw coal
1#/W	1# coal after W-leaching treatment
1#/A	1# coal after A-leaching treatment
1#/H	1# coal after H-leaching treatment
k_i	mean value of the sum of RES in the same level, %
R_j	range of maximum and minimum value of *ki*, %
Y	value of RES in each trial of orthogonal test, %
C_A	concentration of ammonium acetate, mol/L
C_H	concentration of hydrochloric acid, mol/L
DTG	weight loss rate, %/min
DTG_{max}	maximum of weight loss rate, %/min
T_{max}	the corresponding temperature of DTG_{max}, °C
C_x	content of sodium in the leachate, mg/L
C_0	content of sodium in the blank experiment, mg/L
V_0	volume of leaching agent, L
$m_{Na,\ total}$	amount of sodium in raw coal, mg/g
m_0	amount of coal sample in each trial, g

sodium species at high temperatures and transform them to the less-corrosive and less-volatile forms [3,7,8]. The latter is adopting some leaching agents to remove the sodium species before coal conversion [2,9,10]. Considering that minerals addition has been widely investigated, the present work will pay more attention on the leaching treatment.

According to the relevant studies, most of them have focused more attentions on the upgrading of biomass [11–15]. It is well-known that raw biomass contains a high level of AAEMs (alkali and alkaline earth metals) by nature, which is considered as a severe threat for the normal operations of a boiler and gasifier. In view of this, prior to the gasification and combustion, some leaching treatments were adopted to remove a substantial fraction of troublesome elements [15,16]. After leaching treatments, it was generally believed that ash-related problems could be efficiently migrated due to the significant reduction of AAEMs and ash content, whereas the qualities of biomass for thermal conversion were greatly improved [12,14].

Compared with biomass, coal usually contains relatively less AAEMs; therefore, there have been only a limited amount of investigations to demonstrate the removal of AAEMs by using leaching treatment [1,9]. However, due to the great difference in the history of coal formation and local geological surroundings, some low-rank coals are featured by high content of sodium, up to 4% or even 20% in ash [2,17]. Therefore, a leaching pretreatment seems to be extremely necessary for the potentially troublesome sodium-rich coals. Nevertheless, the vast majority of studies have concentrated on the production of ultra clean coal (UCC) [2,10,18]. As reported, UCC is the coal-derived solid fuel with overall ash content ca. 0.1 wt%, the utilization of which instead of raw coal can completely estimate ash-related problems and flue-gas cleaning requirement. However, the leaching treatments for UCC production usually used some aggressive solutions such as strong acids (HCl, HNO_3 and HF) to ensure ash removal efficiency, which inevitably generated much acid-containing wastewater. Furthermore, due to the excessive consumption of acid, those processes are very uneconomical for industrial application [2]. It should be noted that, if only produce the clean coal for gasification and combustion, there is no need to remove all the mineral matters from coal. Most of mineral matters are not terribly harmful to boiler or gasifier. On the contrary, a certain amount of ash in coal is essential for the slag hanging in entrained-flow bed gasifiers.

Based on this conception, we have tried to remove the inherent sodium from coal since the abundant sodium has the most potential threat to boiler and gasifier. Several reports have verified that sodium is mainly presented as water-soluble (such as NaCl) and ion-exchanged (—COONa) forms, implying that they can be leached out by water and ion-exchanged agents [1,19]. Thus, several mild leaching agents can be employed to selectively remove sodium from coal with less cost and environmental influences. In this process, only part of ash compositions are removed, but ash-related problems could be efficiently migrated. Nevertheless, literature review has shown that only few researches have explored the feasibility of this leaching treatment, not to mention the optimization of process conditions [9].

Thus, in this work several leaching pretreatments were conducted by using some mild agents (deionized water, ammonium acetate solution and diluted hydrochloric acid) to remove sodium from those sodium-rich coals. The leaching conditions, such as particle size, temperature, residence time and the concentration of agents were well optimized by orthogonal experiments to achieve the maximum of removal efficiency of sodium (RES). Subsequently, under optimal conditions, the variation of RES along with residence time was investigated to examine the effectiveness of optimized results. Moreover, the effect of leaching treatments on the properties of coal were detailedly investigated by a series of analytical methods. Finally, three other sodium-rich coals were employed to examine the universality of optimal conditions. In short, as part of a broad research program, the objective of this research is to find out some alternative methods to solve the sodium-related problems during the utilization of sodium-rich coals.

2. Material and methods

2.1. Coal samples

Four typical sodium-rich coals labeled as 1#, 2#, 3# and 4# were selected as the testing samples. Among them, 1# and 2# coal were collected from Zhundong district of Xinjiang province, China. The 3# coal was collected from Yili district of Xinjiang province, China. The 4# coal was collected from the State of Victoria, Australia. All samples were dried at a room temperature, then pulverized and sieved to different size. Prior to leaching treatment, coal samples were dried at 110 °C for 2 h to eliminate the moisture disturbance. The proximate and ultimate analyses are listed in Table 1. The ash compositions are listed in Table 2.

2.2. Leaching treatment

Some mild solutions (water, ammonium acetate and diluted hydrochloric acid) were employed as leaching agents. The leaching treatment was performed on a water bath with a temperature controller and magnetic stirrer. Briefly, 1.0 g coal sample was initially immersed in 100 mL leaching agent with the continuous magnetic