



Full Length Article

Investigation on the effects of different forms of sodium, chlorine and sulphur and various pretreatment methods on the deposition characteristics of Na species during pyrolysis of a Na-rich coal



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ABSTRACT

In this work the effects of different forms of sodium, chlorine and sulphur and various pretreatment methods on the evolutionary behaviors and deposition characteristics of Na species were systematically investigated. The pyrolysis experiments of Na-loaded coal samples (in which Na was loaded by NaOH and CH₃COONa) blended with various Cl- and S-bearing additives were conducted in a lab-scale pressurized fixed bed reactor at 1000 °C, and the transformation of Na was subsequently analyzed. Finally, the mechanism on the evolutionary behaviors of Na was proposed. Experimental results show that acid elution treatment could significantly elute corrosive elements from the coal and the absence of anions in gas phase could significantly reduce Na deposition even adequate Na was loaded into the acid washed coal. However, when Cl- and S-bearing substances were added to the Na-loaded coal, abundant NaCl and Na-S-O crystals were deposited on the probes and Cl is more competitive in enhancing Na-bearing compounds deposition, compared with S. During Zhundong coal pyrolysis, the crystalline process of Na species, morphologies of the deposits and Na retention in char are significantly affected by the interactions between Na-bearing compounds and coal matrix or other inorganic species. More importantly, the deposition of crystal NaCl was decreased with rising pretreatment temperature. While the addition of CaO in coal or char was effective in capturing volatile Cl atoms and/or HCl to generate CaCl₂, resulting in the absence of NaCl in the deposits during coal or char pyrolysis. Furthermore, the addition of CaO in char could also enhance the interactions between active Na species and coal matrix or minerals, resulting in the increased Na content in CaO-loaded char.

1. Introduction

Zhundong lignite with a predicted reserve of 390 billion tons will meet the rapid growth of energy consumption in China [1,2]. However, its inherent mineral species, especially the high contents of sodium and chlorine, in Zhundong coal could practically give rise to severe fouling, slagging and corrosion problems both inside the furnace and downstream processes, seriously restricting its large-scale utilization [3–5].

Reportedly, the water-soluble and organically bonded Na species are dominant in Zhundong coal [6,7]; while this highly active Na demonstrates a strong sublimation tendency [8,9]. Pyrolysis is the primary stage of coal thermal conversion processes, and during this process, abundant Na with high corrosivity could diffuse into the gas phase

[10–12]. Subsequently, the released Na-bearing compounds as well as Na atoms and/or ions in the aerosol could interact with the gaseous constituents and then generate the fouling precursor, deteriorating the slagging in equipment [13–15]. Previous studies largely focused on the quantitative and qualitative analysis of the released sodium [16–18] and on the characteristics of the residual char, fly ash and deposits generated during thermal conversion of Zhundong coal [19–21]. Recently, extensive fundamental efforts have also been performed on clarifying the evolutionary behaviors of Na during coal thermal conversion processes. In general, the diffusion of Na mainly relies on coal characteristics and thermal conversion conditions, e.g., the reaction temperature, atmosphere, pressure, coal rank and compositions, particle size and reactor type [22–25]. Specifically, the release of Na was

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significantly suppressed by the CO₂ atmosphere due to the formation of Na₂CO₃ during coal thermal conversion, while the inert or reducing atmosphere was conducive to the release of Na in the form of NaCl [26]. In addition, the high contents of Cl and S in coal could also enhance the emission of Na [27,28]. More importantly, the occurrence of sodium also has a significant effect on its diffusion behaviors. For example, water-soluble and organically bonded Na species in Zhundong coal have a high volatilization tendency during high temperature pyrolysis [6], while the insoluble Na is very stable in char [29]. However, due to the complexity of Na, Cl and S occurrence in coal, the diffusion and deposition properties of corrosive species with different forms in coal are still unclear.

On the other hand, various pretreatment methods, such as leaching and coal blending, have been tried to alleviate the release and deposition of Na-bearing substances during coal utilization [30,31]. However, those methods are practically costly and inefficient for reducing the emission of the more corrosive elements, such as Cl and S species. Generally, Cl and S species are supposed to be released during coal pyrolysis at relatively low temperatures, which would substantially mitigate the corrosion in equipment operated at high temperatures. Besides, suppressing the release of Cl and S could theoretically reduce the release and deposition of Na-bearing species. However, researches regarding the effect of various kinds of basic oxide, e.g., CaO known for efficient sulfur fixation, on inhibiting the emission of Na and Cl are very scarce.

As reviewed above, present literature mainly focused on quantitative conversion of sodium and coal ash chemical behavior. But few studies have explored the effects of anions on the evolutionary behavior of sodium. Getting insight into the evolutionary behavior of Na, Cl and S species would theoretically obtain more meaningful information on the formation mechanism of corrosive substances. Accordingly, this paper systemically reveals the effects of various forms of Cl and S on the initial diffusion and evolutionary mechanism of Na during pyrolysis of Na-loaded coal, and investigates the effects of different pretreatment methods on inhibiting Na emission via capturing and then analyzing the mineral aerosol generated from the evaporation or diffusion of volatile elements during coal pyrolysis in terms of chemical compositions and morphology. The fundamentals on the evolutionary behavior of corrosive species in Zhundong coal are beneficial for getting deeper insight into the roles of Na in inducing fouling and slagging and also provide supporting mechanism on new corrosion-control strategies of high-Na coal utilization.

2. Experimental section

2.1. Sample preparation and characterization

A typical high-sodium coal from Wucuiwan, Zhundong coalfield in Xinjiang province, (hereafter referred as W-coal), is used in this experiment. Prior to the experiments, the coal sample was ground and sieved to < 74 μm. In order to reveal the effect of specific elements on the evolution and deposition behaviors of Na and avoid introducing additional pollutant species, W-coal was successively washed with 0.2 mol/L dilute nitric acid (solid to liquid ratio = 1:50) three times for 12 h each round and subsequently rinsed with distilled water to eliminate the residual ions until no NO₃⁻ was detected in the leachate; the washed coal sample was then dried in the vacuum oven at 80 °C for 24 h. As a result, the obtained acid-washed coal (noted as AW-coal) contained a low content of Na that mainly existed as acid-insoluble forms (silicate and/or aluminosilicate) [32]. In order to differentiate the effect of various occurrences of sodium on its diffusion behavior, a certain amount of NaOH (based on the content of Na in W-coal) was loaded into the AW-coal via the impregnation method (details can be found elsewhere [33]) and this NaOH-loaded coal was termed as inNa-coal hereafter. Besides, sodium acetate (CH₃COONa) ion-exchanged coal (termed as orNa-coal) was also prepared in this experiment. In

Table 1
Analysis of samples.

Samples	W-coal	AW-coal
Moisture (wt% ad)	11.81	5.67
<i>Proximate analysis (wt% db)</i>		
Volatile matter	30.63	31.16
Fixed carbon	62.29	64.27
Ash	7.08	4.57
<i>Ultimate analysis (wt% daf)</i>		
C	79.07	75.58
H	4.05	4.06
N	0.79	0.52
S	0.51	0.35
O _(diff.)	15.58	19.49
<i>Elemental compositions of ash (wt%)</i>		
SiO ₂	25.56 ± 0.22	63.47 ± 0.24
Al ₂ O ₃	12.55 ± 0.17	24.82 ± 0.22
CaO	23.15 ± 0.21	1.22 ± 0.05
Na ₂ O	3.83 ± 0.10	0.31 ± 0.02
Fe ₂ O ₃	2.88 ± 0.08	2.57 ± 0.08
MgO	7.20 ± 0.13	1.04 ± 0.05
SO ₃	19.77 ± 0.20	3.25 ± 0.05
K ₂ O	0.36 ± 0.02	0.66 ± 0.03
Cl	0.96 ± 0.05	ND

Note: ad.: air dry basis; db.: dry basis; daf.: dry and ash free; diff.: by different; ND: no detect.

brief, 1 L of sodium acetate solution (0.2 mol/L) was mixed with 100 g AW-coal and the slurry was stirred at room temperature for 24 h and then filtrated and washed with adequate distilled water to eliminate the residual CH₃COONa, and dried at 80 °C for 24 h under vacuum. The proximate, ultimate and ash compositions analyses of W-coal and AW-coal are presented in Table 1. Clearly, the Na and Cl species in AW-coal was significantly eluted. Additionally, the content of Na in raw coal, AW-coal, inNa-coal and orNa-coal was 0.28 ± 0.0046, 0.049 ± 0.0015, 0.31 ± 0.0076 and 0.71 ± 0.013 wt%, respectively, on dry basis, which determined by microwave digestion (described elsewhere [34]) and inductively coupled plasma optical emission spectroscopy (ICP-OES). Meanwhile, the contents of various chemical forms of Na in W-coal, inNa-coal and orNa-coal were determined by sequential extraction method (the detailed description can be found elsewhere [34]), as plotted in Fig. 1. Clearly, Na exhibited significantly different occurrence characteristics in those Na-loaded coal samples. Specifically, the water-soluble and ammonium acetate-soluble Na was dominant in inNa-coal, which was, to some extent, in accordance with the content and occurrence features of Na in W-coal. However, the content of ammonium acetate-soluble Na in orNa-coal was significantly

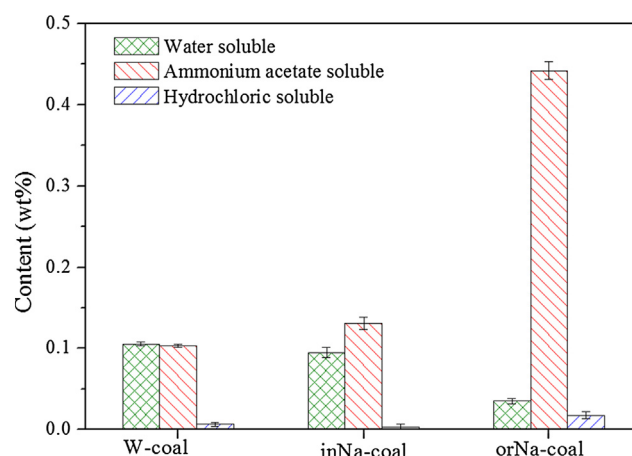


Fig. 1. The contents of various chemical forms of Na in W-coal, inNa-coal and orNa-coal.

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