Study on the cleanliness of super clean coal prepared by water-only cyclone

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Abstract: Maintaining clean and pristine nature is the key to the use of super clean coal (SCC) for coal-based materials or energy combustion. Herein, SCC is prepared by a new water-only cyclone method, and compared the surface characteristics with the SCC products obtained by conventional chemical deashing method. The results indicate that: FTIR analysis revealed that the chemical method changed the original functional group of coal; BET analysis revealed that the SCC products prepared by the water-only method maintained the micropore volume of the raw coal and yielded a uniform and concentrated distribution of pore sizes, however, the chemical method destroying the original pore structure in the coal; SEM-EDS analysis indicated that the surface of SCC particles that were deashed using the water-only cyclone method was smooth and neat, whereas those obtained using the chemical method were seriously corroded, had a rough surface, and the SCC particles were prone to acid residues and precipitates. This study opens an innovative, simple, and clean method for the preparation of SCC, which further expands and enhances the potential application value of SCC.

Keywords: cyclone separating, chemical deashing, super clean coal, surface characteristics, a comparative study

1. Introduction

Super clean coal (SCC) is defined as very low ash (< 3 wt%) coal; it features low amounts of ash, sulfur, and phosphorus, and high chemical activity, vitrinite content, and calorific value (Melikoglu, 2018; Li et al., 2020; Onel and Tanriverdi, 2020). SCC has extensive applications. As an energy source, SCC can improve product quality and reduce environmental pollution; its direct combustion can reduce the emissions of coal combustion pollutants such as particulate pollutants, SO_2, and NO_x, and thus reduce pollution. The use of SCC for the preparation of coal-water slurry, which is used to replace petroleum products, can reduce the dependence on petroleum (Strizhak and Vershinina, 2017), and the coke products obtained from SCC can be used in steelmaking to reduce the consumption of coke and improve the quality of steel. Additionally, SCC can be used to develop new coal-based composite materials to improve the quality of carbon fiber, carbon nanotubes, and activated carbon products, which have great application potential (Hong et al., 2013). Since China has proposed carbon peaking and carbon neutrality goals, research on SCC will become a hot topic (Qin et al., 2021; Wu et al., 2021; Liu et al., 2022).

Currently, the preparation methods of SCC mainly encompass chemical, physical (such as gravity separation), and physicochemical methods. Chemical methods include conventional acid-base, hydrofluoric acid (HF), and organic acid methods (Gulen et al., 2013; Rahman et al., 2017; Dikici and Hacifazlioglu, 2022). Additionally, the ash reduction of coal with high volatile content was investigated using HF and HNO_3 and achieved a reduction in the amount of ash from 7.90% to 2.60% and further down to 0.60% with continuous use of HF and HNO_3 (Steel and Patrick, 2003; Wu and Steel, 2007). Hacifazlioglu (2016) used HF, HCl, HNO_3, and citric acid aqueous solution to perform chemical leaching demineralization on Zonguldak bituminous coal with a particle size of -212 μm and obtained SCC with an ash content of 2.84%. Dikici and Hacifazlioglu (2022) explored an SCC preparation method with flotation followed by acid leaching. During flotation, sunflower oil was used as a collector and

eucalyptus oil as a frother, and the ash content of the pulverized coal was reduced from 29.67% to 6.10%; afterwards, the clean coal was soaked with citric and formic acid at 65 °C for 3 hours to produce SCC with an ash content of 2.92%-2.98%. This study revealed that SCC could be produced by a completely organic process; however, the use of excessive organic reagents was involved in the SCC preparation process, which hindered the further development of this method.

Although chemical methods perform well in ash reduction and are capable of reducing the ash content to a very low level, the preparation process requires the use of a large amount of strong acids, bases, and other hazardous chemical reagents, which not only poses a great safety hazard but also results in difficulties in the subsequent recovery of the chemicals and extremely serious environmental pollution. Even preparation processes that only use organic reagents consume a large amount of reagents, which also presents a major hurdle in the application of these methods (Behera et al., 2018; Dash et al., 2013; Sriramoju et al., 2021).

In addition, chemical methods alter the properties of coals, and thus greatly impact the quality and application of SCC products, further limiting the development and industrial application of SCC. Rubiera et al. (2002) analyzed samples using mass spectrometry and FTIR and concluded that the chemical demineralization process changed the chemical composition and organic structure of the samples, which were adulterated with nitro functional groups, with increased volatile matter, oxygen, and nitrogen content. Wang et al. (2021) examined the evolution of oxidation properties and microstructure of coal after acid treatment using gas chromatography, FTIR, and X-ray photoelectron spectroscopy (XPS). The experiment results showed that HCl and HNO_3 treatments increased the pore volume and porosity, enhanced the oxygen adsorption capacity, and accelerated the oxidation of coal. Xu et al. (2021) conducted erosion experiments on coal samples using HCl and NaOH and compared the erosive effects of HCl and NaOH on coal using XRD, FTIR, and XPS. Results from this research revealed that NaOH could not dissolve hematite and HCl could not dissolve quartz. Additionally, the alkali solution produced many new minerals in the dissolution process. HCl mainly acted on the carboxyl and oxidized parts in the coal, and sodium hydroxide mostly affected the hydroxyl and aromatic structures; both have little effect on aliphatic functional groups. HCl has a minor effect on surface morphological characteristics and can effectively remove particles from the surface of coal. Dissolution pores appear on the surface of coal after sodium hydroxide dissolution.

Physical gravity separation and physicochemical methods for separating SCC mainly include dense medium separation and flotation. Özbayoğlu and Mamurekli (1994) investigated dense medium separation and flotation techniques to produce SCC from Zonguldak bituminous coal, and the results showed that two-stage dense medium separation reduced the ash content of bituminous coal from 12.11% to 4.00%, and subsequent separation by flotation produced SCC with an ash content of 2.16%. Li et al. (2003) used a cyclo-microbubble flotation column, which combined cyclone separation and a flotation column, to recover SCC from 0.1 mm fine-grained coal. Both laboratory and pilot-scale experiments indicated that the process could produce SCC products with an ash content of 1.5% from a feed sample with an ash content of 9.8%. Hao et al. (2020) explored the mechanism of microbubble flotation for clean coal preparation using XRF and SEM-EDS analysis and found that a small amount of fine-grained clay minerals, such as kaolinite and illite, formed uneven agglomerates with coal particles and contaminated the clean coal during flotation. Furthermore, the presence of composite particles and coal slime coating was the main factor limiting the in-depth demineralization during microbubble flotation. He et al. (2016) mixed fine coal with magnetite powder to form a binary dense medium and obtained the optimum secondary air distribution layer height by verifying the uniformity of the bed density. Under these conditions, clean coal with a minimum ash content of 2.65% was prepared.

Compared with chemical methods, the conventional physical gravity separation method has the advantages of a simple process, low cost, and less damage to coal properties. However, the conventional physical separation method routinely requires the grinding of coal samples to form fine particles, therefore resulting in high energy consumption. Furthermore, the fine particle size makes the dewatering process of the products challenging; hence, further research on the dewatering technology of ultra-fine grain coal is required. Additionally, the flotation and dense medium separation processes are prone to retaining residues of chemicals and dense medium on the surface of the fine coal particles; these residues are hard to remove and pollute SCC products.

However, some coal preparation plants have produced coal slime with strong dissociative characteristics from washed raw coal, that is, some extraneous ash is dissociative with the cleaned coal particles, and only a very low content of inherent ash exists that naturally has a very low yield (Fan et al., 2015; Amini et al., 2016; Pyka and Wierzchowski, 2022). Recently, we (Wang et al., 2020; Liu et al., 2021) used a gravity separation method, a water-only cyclone, to obtain SCC products with an ash content of 2.74% and a yield of 10.56% from jigging overflow coal slime. The process was simple, efficient, low cost, and produced no pollution in the SCC products or the environment during the preparation process.

In the present study, we analyze the composition, BET surface area, porosity and surface morphology of SCC products prepared using the water-only cyclone and chemical methods using XRD, BET, SEM-EDS, and FTIR spectroscopy, based on previous studies. We further elucidate the influence of water-only cyclone and chemical methods on SCC products to provide reference information for the diversified usage of coal.

2. Materials and methods

2.1. Materials

The raw coal sample, characterized by a high ash content and heavy muddification, was sourced from coal slime produced from jig washers in a coal washing plant in Shanxi, China. The results of the particle size distribution, float and sink analysis are presented in Tables 1 and 2, respectively. The specific procedure for determining the ash content (Ad) is as follows: 1 g of air-dried coal sample was transferred into a muffle furnace. The furnace was slowly heated to 500 °C over a period of 30 min and then maintained at this temperature for 30 min. The temperature was then further increased to 815 °C and maintained for 1 h. During this process, the organic matter is completely combusted. The ash content was then calculated as the percentage of the sample's mass represented by the residue.

Table 1. Particle size distribution and ash of coal slime sample

Sizes (mm)	Yield (%)	Ash (Ad, %)	Cumulative (%)	
			Yield (%)	Ash (Ad, %)
＞0.500	0.54	8.05	0.54	8.05
0.25-0.50	2.47	8.21	3.01	8.18
0.125-0.25	16.18	16.29	19.19	15.02
0.075-0.125	12.04	28.67	31.23	20.28
0.045-0.075	13.62	34.23	44.85	24.52
＜0.045	55.15	39.39	100.00	32.72
Total	100.00	32.72		

Table 2. Float and sink analysis of coal slime sample

Density fraction (g/cm^3)	Yield (%)	Ash (Ad, %)	Cumulative (%)	
			Yield (%)	Ash (Ad, %)
-1.3	21.60	2.89	21.60	2.89
1.3-1.4	13.98	5.48	35.58	3.91
1.4-1.5	17.67	19.87	53.25	9.20
1.5-1.6	2.27	31.88	55.52	10.13
1.6-1.8	29.25	47.56	84.77	23.05
+1.8	15.23	85.32	100.00	32.53
Total	100.00	32.53		

The results of the particle size analysis showed that the coal sample had a wide range of particle sizes, and the content of individual particle sizes was unevenly distributed. Most of the fine coal slime had a particle size of less than 0.025 mm, and it was predominantly high ash content slime of particle size under 0.045 mm, with a yield of 55.15%. The overall ash content of the coal sample was 32.72%.

The results of the density analysis indicated that under a 1.3 g/cm³ density level, an SCC product could be recovered using the density separating process of the water-only cyclone, with a yield of 21.60% and an ash content of below 3%.

The dry ash-free volatile fraction of the coal sample was 40.72%, revealing that the sample was typical low-grade coal. The ash content of the air-dried base was 32.81%, indicating ash slime. The dry ash-free base contained a fixed carbon fraction at 57.28%, displaying a relatively high usage value.

2.2. Sample preparation

Figure 1 shows the flow chart of the water-only cyclone and chemical deashing process experiment. Water-only cyclone method: According to the properties of coal slime and the requirements of classification and separation, a two-stage cyclone (φ100 mm-φ75 mm) was used for the classification and separation processes. The diameter of the first stage of the cyclone was 100 mm, whereas the diameter of the second stage was 75 mm; the two stages were connected in series. The overflow products of the second-stage cyclone were screened and deslimed using a 0.125-mm mesh sieve, and the product was SCC. Herein, an ash content of 2.74%(Ad), accounting for 10.56% of the raw coal sample yield of SCC can be obtained over the sieve.
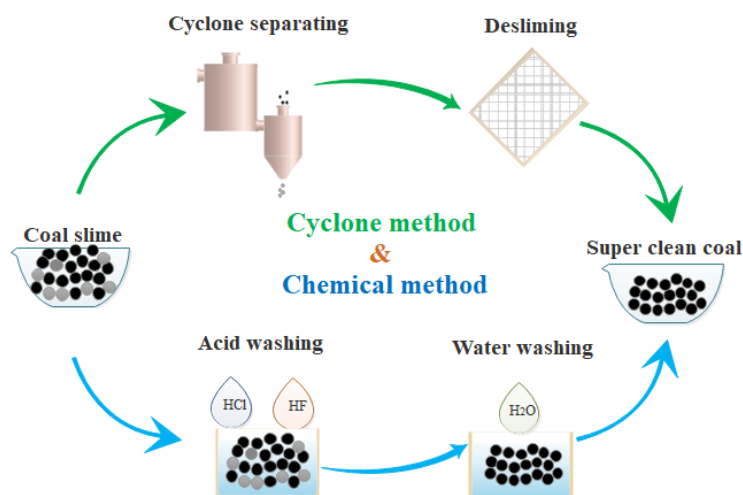

Fig. 1. Flow chart of the water-only cyclone and chemical deashing process experiment

Chemical deashing method: 6 g of coal sample was accurately weighed and put into a plastic beaker containing 40 cm³ of HCl (5 mol/dm³). The mixture was thoroughly stirred using a plastic rod ensuring that the sample was completely wetted, followed by heating in a water bath at 60 °C for 2 h. The mixture was filtered using a quantitative analysis filter paper and 40 cm³ of HF was slowly added. The acid-treated sample was re-heated in a water bath at 60 °C for 2 h and re-filtered using the same filter paper. 50 cm³ of concentrated HCl with a density of 1.19 g/cm³ was added to the sample until the ash content was reduced to 2.48%(Ad). The same heating and filtering procedure was repeated. Finally, the acid-treated coal sample was rinsed with distilled water until no Cl^{-1} ions were detected using an $AgNO_3$ solution. The treated coal sample was then vacuum-dried for 6 h and stored in a dryer for future use. The remaining weight after drying was 4.14 g, accounting for 69.00% of the yield of the raw sample.

2.3. Sample testing

The structure of the samples was analyzed using XRD with a Miniflex X-ray diffractometer made by Rigaku Corporation, Japan. A Nicolet FTIR infrared spectrometer (Thermo Fisher Scientific Inc.) was used, with KBr for sample preparation and a wavelength range of 400 cm⁻¹-4000 cm⁻¹ and resolution of above 0.5 cm⁻¹. A LabRAM HR800 Raman Microscope manufactured by HORIBA JOBIN YVON S.A.S. was used, with an excitation wavelength of 633 nm and a scanning range of 800 cm⁻¹-1800 cm⁻¹. TG/DTG analysis was carried out using an SDTQ 600 TGA-DSC analyzer, with a temperature ramping rate of 10 °C/min, N_2 flow rate of 100 mL/min, and a maximum temperature of 1000 °C.

3.   Results and discussion

3.1.   XRD analysis

Figure 2 shows the XRD patterns of the coal samples before and after deashing for both the physical and chemical methods. The XRD pattern of the raw coal shows sharp and strong diffraction peaks; the main mineral species in the raw coal were determined to be quartz, calcium carbonate, and kaolinite. The XRD pattern of the clean coal after the cyclone and chemical deashing processes only exhibited minor mineral matter diffraction peaks, and identification was not performed due to the low mineral matter content.
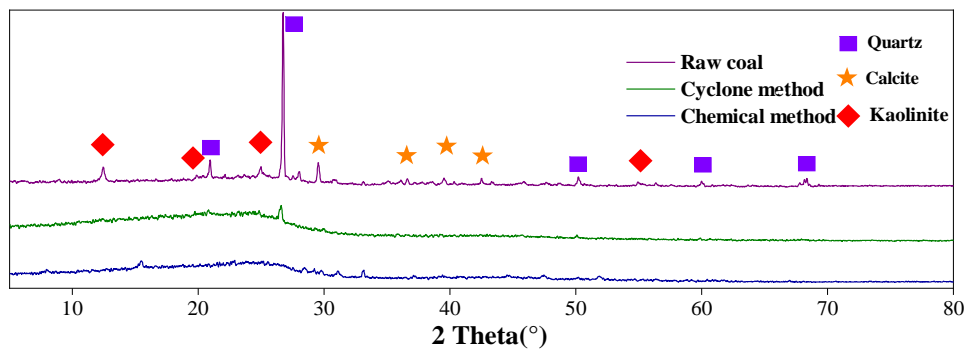
Fig. 2. XRD patterns before and after deashing

3.2.   FTIR analysis

Since the absorption peaks of the minerals in the coal masked most of those of the organic functional groups, the absorption peaks of organic matter were analyzed from the second derivative FTIR spectra. The results shown in Figs. 3 and 4 indicate that the absorption peak of free -OH appears at approximately 3670 cm^{-1} and the absorption peak near 3570 cm^{-1} is caused by independent hydrogen bonding with the -OH. The carbonyl group (C=O) showed a weak absorption peak at 1745 cm^{-1}-1630 cm^{-1}, and the absorption peaks at 1737 cm^{-1}, 1700 cm^{-1}, and 1675 cm^{-1} were from the stretching vibrations of aliphatic C=O (ester group, acids, ketones, and aldehydes), aromatic C=O (carbonyl/carboxyl groups), and C=O in a conjugated double bond, respectively. The peaks at 1644 cm^{-1} and 1650 cm^{-1} were highly conjugated absorption peaks of C=O. The absorption peaks at 1400 cm^{-1}, 1170 cm^{-1}, and 1140 cm^{-1} were caused by the stretching vibrations of C-O in carboxylic acids, alcohols, ethers, and esters, indicating the prevalence of these groups in the coal samples. The absorption peaks at 3000 cm^{-1}-2700 cm^{-1} were caused by the stretching vibrations of aliphatic functional groups, and those at approximately 2955 cm^{-1} were from the asymmetric stretching vibrations of -CH$_3$.
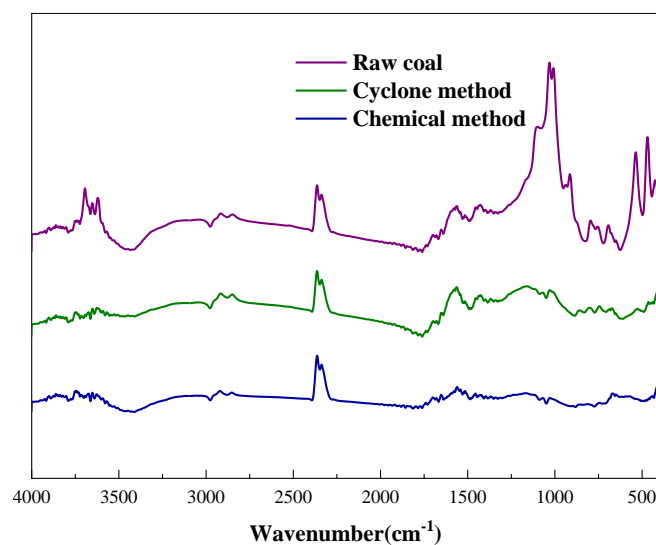

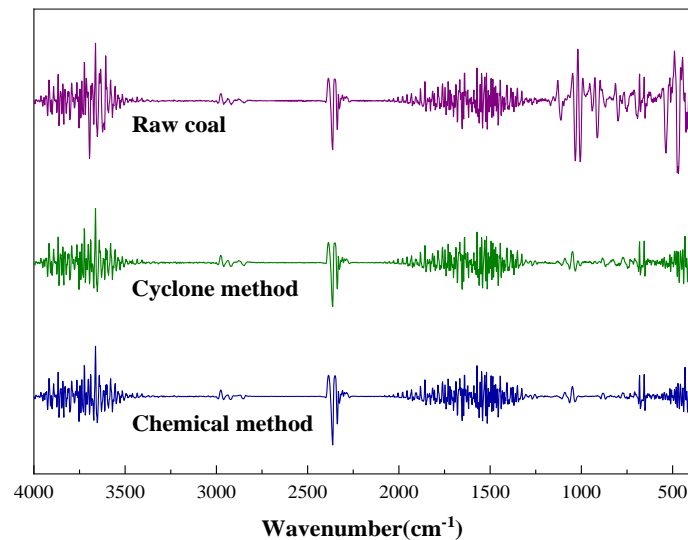Fig. 3. FTIR analysis of the samples

Fig. 4. Second derivative FTIR spectra

The peaks at 2918 cm^{-1} and 2845 cm^{-1} were from the asymmetric and symmetric stretching vibrations of -CH$_2$, respectively. In addition, 1453 cm^{-1} and 1368 cm^{-1} show the asymmetric and symmetric bending vibration absorption peaks of aliphatic -CH$_3$, respectively. The aromatic C=C has a stretching vibration absorption peak at 1600 cm^{-1}. The absorption peaks in the range of 900-700 cm^{-1} are caused by the out-of-plane bending vibrations of (C-H)ar; herein, (C-H)ar represents the bonding between the carbon atoms of the aromatic ring and adjacent hydrogen atoms in aromatic compounds. The bending vibration peak of the n-alkane side chain backbone (CH2)$_n$ appears near 720 cm^{-1}, indicating the presence of aliphatic side chains in the coal.

Compared with the raw coal samples, the characteristic FTIR absorption peaks of both kaolinite and quartz disappeared in the FTIR spectra of the samples after physical and chemical deashing, indicating the adequate performance of both methods. However, the disappearance of the absorption peaks of alkane side chains in the range of 900 cm^{-1}-700 cm^{-1} in the FTIR spectra of the chemically deashed samples reveals that chemical deashing impacted the side chains of the coal samples and changed the original surface structure of the coal.

3.3. Pore structure and specific surface area

Figure 5 shows that the adsorption on the desorption isotherm was always greater than that on the adsorption isotherm under a certain relative pressure. Because capillary condensation occurred in the fine pores in the coal, a hysteresis loop formed under moderate relative pressure, indicating the presence of abundant micropores and some mesopores in the coal samples (Hao et al., 2015; Xie et al., 2016). The sharp increase at p/p_0 = 1.0 in the N$_2$ adsorption/desorption isotherms in Fig. 5 indicates that the sample contains a certain amount of macropores. According to the classifications proposed by the International Union of Pure and Applied Chemistry, the adsorption isotherms of the three samples are of type IV with H$_3$-type hysteresis loops (Thommes et al., 2015).

The adsorption and desorption curves were roughly parallel throughout the relative pressure interval; the adsorption loops were small and appeared at relative pressures from 0.4 to 1.0, and none of the inflection points of the desorption loops were obvious. The adsorption/desorption curves were very stable until the relative pressure reached 0.8, after which they showed a clear upward trend, followed by a sharp increase in both curves under a relative pressure close to 1.0. Thus, we concluded that the pore types of the coal samples were not changed, and the dominant pores in the coal samples were impermeable pores closed at one end.

The pore size distribution analysis was conducted on the raw coal and SCC products obtained using the water-only cyclone and chemical methods. The variation of pore volume with pore size is shown in Figure 6. The figure shows that the pore volume of the micropores in the SCC prepared by the chemical method is close to 4.0×10^4 cm^3/g, which is larger than that of the samples prepared by the water-only

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cyclone method and the raw coal. Additionally, the microporous pore volumes of the water-only cyclone processed samples and the raw coal were the same, indicating that the chemical method substantially increased the microporous pore volume, whereas the water-only cyclone method maintained the microporous pore volume of the raw coal.
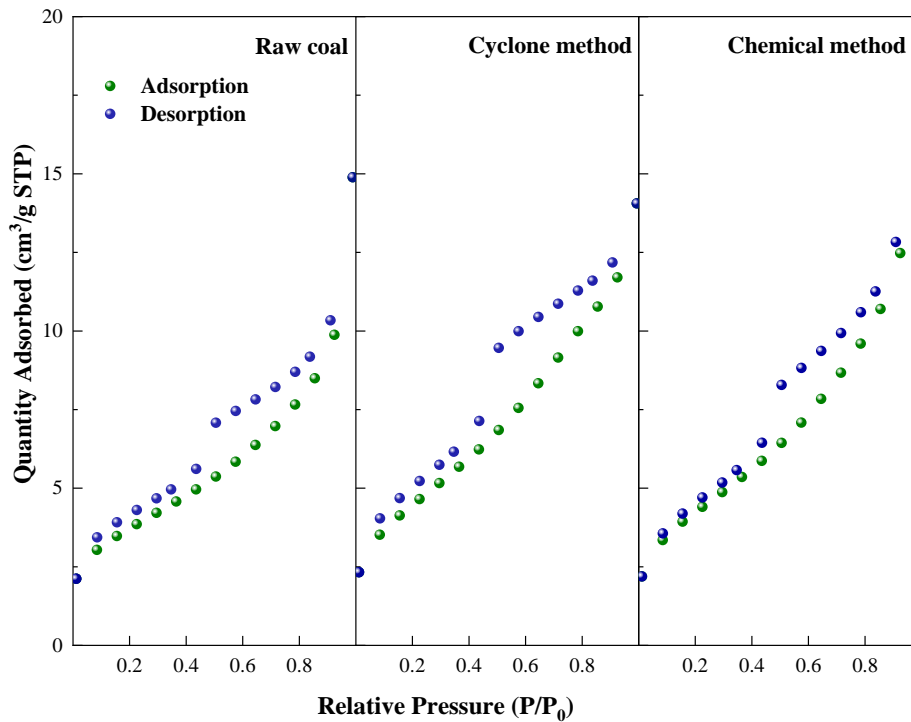

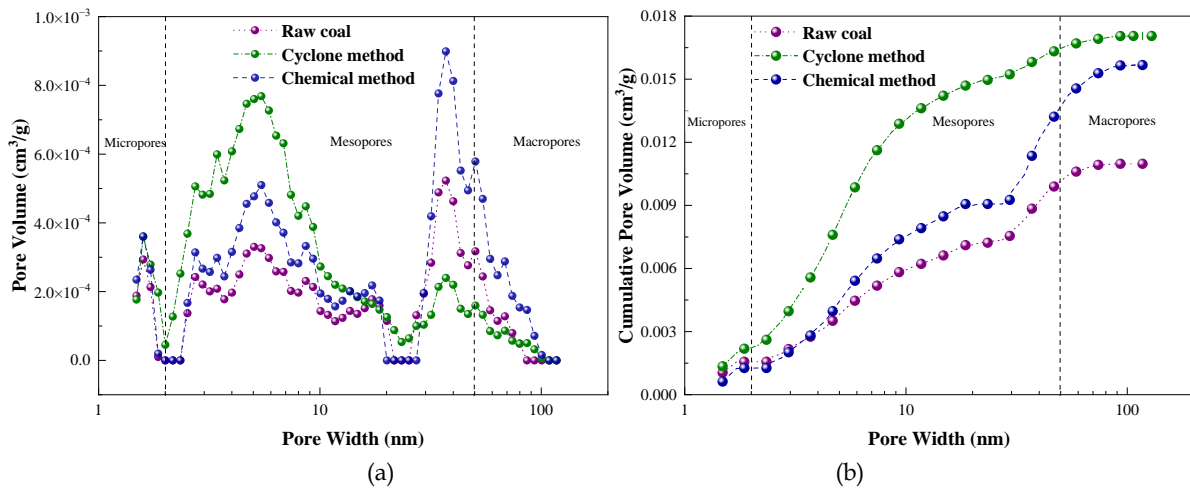Fig. 5. Comparison analysis of adsorption and desorption

Fig. 6. Pore size distributions: (a) pore volume vs width, and (b) incremental pore volume vs width

In the mesoporous region, when the pore size was less than 30 nm, the pore volume of the SCC prepared by the water-only cyclone method was substantially larger than that of the samples prepared by the chemical method and the raw coal, indicating that the coarse-grained SCC had a high mesoporous pore volume. When the pore size ranged from 30 nm to 100 nm, the pore volume of the SCC prepared by the chemical method was larger than that of the samples prepared by the water-only cyclone method and the raw coal, indicating that the chemical method increased the volume of pores with a pore size above 30 nm. Because chemical deashing was carried out using the reaction of chemical reagents with the minerals in the coal to produce water-soluble products, the pore size distribution of the coal was inevitably changed. Additionally, the HCl and HF used in acid deashing are highly

corrosive, and corrode the surface of the coal while dissolving the minerals, therefore destroying the pore structure of the coal and increasing the pore volume. When the pore size was between 30 nm and 100 nm, the pore volume of the samples prepared using the water-only cyclone method was smaller than that of the raw coal, indicating that the coarse grain-sized SCC prepared by the water-only cyclone method had a low pore volume when the pore size was larger than 30 nm.

Figure 7 shows that the contributions of pore size to the specific surface area are similar for raw coal and SCC products obtained using the water-only cyclone and chemical methods, and mesopores have more contributions to the specific surface area, mainly in the range of 2 nm -10 nm. Figure 7(b) shows that when the average pore size is larger than 2 nm, the accumulated pore specific surface area of SCC products obtained using the water-only cyclone is the largest, followed by chemical methods, and raw coal is the smallest. This confirms the increasing effect of the water-only cyclone and chemical methods on the specific surface area of the coal. According to the analysis in Fig. 7(a), the specific surface area of SCC prepared by the water-only cyclone method increased as the number of mesopores increased, with the pore size ranging from 2 nm to 10 nm. The chemical methods enhanced its specific surface area by increasing the number of mesopores in two intervals: 2 nm to 10 nm and 30 nm to 45 nm. In summary, compared to chemical methods, the pore size distribution of the water-only cyclone method samples were more uniform and concentrated, displaying a better product quality.
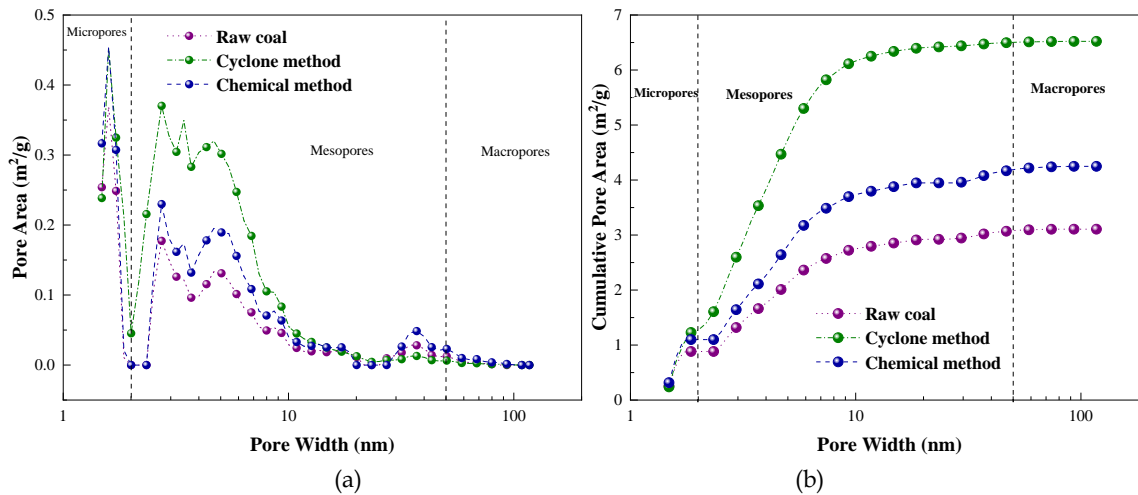

Fig. 7. Pore size distributions: (a) pore area vs width, (b) incremental pore area vs width

3.4. SEM-EDS analysis

The particle morphology, elemental distribution, and elemental compositions of raw coal and SCC samples prepared by the physical cyclone and chemical methods are shown in Fig. 8. Compared with the raw coal particles, the SCC particles prepared using the water-only cyclone deashing had a smooth and neat surface, free from contamination by fine-grained impurities; however, the chemically produced SCC particles were severely corroded, showing a rough surface with a large number of fine particles attached.

The EDS spectrum analysis shows that compared with raw coal, the C content of SCC products prepared by a water-only cyclone and chemical deashing significantly increased, while the content of O, Si, and Al substantially decreased. The Ca content of the chemically deashed product increased and Cl appeared. In the chemical preparation of SCC, HF reacted with the silica to generate insoluble CaF_2, which was deposited on the surface or in the pores of the coal particles. This was hard to remove and blocked the pore connections to a certain extent, thus impacting the adsorption performance of this SCC. Furthermore, during the chemical preparation, the Cl ions in the solution, cannot be completely cleaned up due to the large amount of HCl use, indicating that the clean coal particles are prone to acid solution residues.

The chemically prepared SCC showed the Si content was significantly reduced to 0.03%, whereas the Ca content increased from 0.80% to 4.11%, caused by the formation of insoluble CaF_2 from the reaction between the HF and silica. CaF_2 was deposited on the surface or in the pores of the coal particles

and was not easily removable. The chemically prepared SCC also contained 0.63% of Cl, indicating that the acid residue was commonly retained in the fine coal particles.
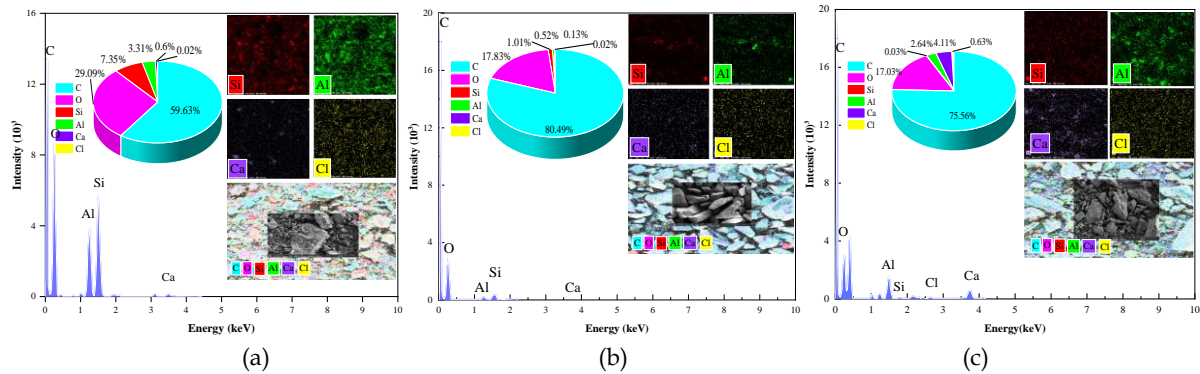

Fig. 8. Morphology and EDS spectrum analysis:(a) raw coal, (b) SCC after water-only cyclone deashing, and (c) SCC after chemical deashing

4. Conclusions

In this study, the effects of water-only cyclone and conventional chemical deashing on the features of SCC products were investigated using XRD, FTIR, BET surface areas, and SEM-EDS spectra. The achievement of high value use of coal slime was proposed through recovering SCC using a water-only cyclone. The SCC prepared by this method showed a relatively uniform and concentrated pore size distribution and a smooth and neat surface and was without additional mineral impurities and chemical contamination, revealing high potential in future applications.

Acknowledgments

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