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The resource utilization of algae—Preparing coal slurry with algae

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A B S T R A C T

Nowadays, the occurrence of harmful algal blooms is increasing rapidly all over the world. However, the methods of resource utilization of algae are very few. In this study, we propose a new way to dispose algae, which is gasification of coal–algae slurry. Coal slurries prepared with algae were investigated, and gasification reactivity of coal–algae slurry was compared with that of coal–water slurry (CWS). The results showed that, anaerobic fermentation, chemical treatment, high-speed shearing and heating are effective pre-treatment methods on reducing the viscosity of algae, which could obviously increase the maximum solids concentration of coal–algae slurry. When the de-ionized water/algae ratio is 1:1, the maximum solids concentration could get to 62.5 wt.%, which is almost the same as that of CWS. All the coal–algae slurries exhibit pseudo-plastic behavior, and this type of fluid is shear-thinning. Compared with CWS, the stability of coal–algae slurry is much better, which could be no solids deposition after 70 h. The coal–algae slurry displays better gasification reactivity than CWS.

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1. Introduction

Algae are photosynthetic aquaticplants that utilize inorganic nutrients such as nitrogen and phosphorus [1]. In recent decades, the occurrence of harmful algal blooms has increased in global waters [2]. These events can cause harm to marine life [3] and human health, with associated detrimental economic impacts [4]. The accumulations of algal cells are fast and extensive. For example, a bloom of *Pseudonitzshia* extended from southern California to Washington in full 1991 [5] and a *L. polyedrum* bloom in 1995 extended from the Baja peninsula to the Monterey Bay [6]. In 2007, Taihu Lake which is the third largest lake in China, and provides drinking water for more than 2 million people, became a toxic nightmare because of cyanobacteria bloom [7].

The treatment of algal bloom in large lakes is a worldwide problem. Many countries try to control it with some ways, including pre-oxidation [8], coagulation and flocculation [9], and clarification either by dissolved air flotation [10] or sedimentation [11]. However, these methods inevitably result in environmental problems, or are not effective in short-time for a big water area. Collecting algae as physical method can operate soon without environmental pollution. In 2007, collecting algae played the key role in controlling Taihu Lake algal bloom. However, algae collected could get to thousands of tons everyday, and how to dispose these toxic algae is an urgent problem.

Coal occupies the primary position in Chinese frame of energy. Traditionally, the utilization of coal was also related to the increasing environmental pollutant, but the clean coal technology (CCT) proposed recent decades can give a promising way to solve this problem. Coal gasification has been a clean and effective way to produce sys-gas [12] from coal, which could be used to produce some chemical products, such as ammonia and methanol, etc., and also to generate power with gas turbine (named as IGCC). Now, the technology of coal gasification has developed so-called second generation technology widely such as Texaco Gasifier, OMB Gasifier, and Shell Gasifier [13–15]. Coal–water slurry [16–19] as the main feeding technology for coal gasification was widely studied by researchers. Compared with coal, biomass is renewable and more environmental friendly because of CO_2 abatement. Recently, many studies involving the utilization of coal and biomass have been conducted [20].

As energy resource, algae have some commonness with CWS, such as a certain caloric value, high water content and suspension system. Therefore, we proposed a new way to dispose algae, which is co-gasification of coal and algae. In this method, algae, coal and additives are milled to produce coal–algae slurry, which substitutes CWS in gasifier. Co-gasification of algae and coal can be achieved by gasification of coal–algae slurry. Because main solids in coal–algae slurry are coal, the effective sys-gas from gasifier also can meet the industrial criterion. However, compared with CWS, little information is available on the co-slurryability and co-gasification of algae and coal. In this study, four methods were used to pre-treat algae, and rheological properties and gasification characteristics of coal–algae slurry were investigated. Gasification of coal–algae slurry may provide an alternative way to dispose algae.

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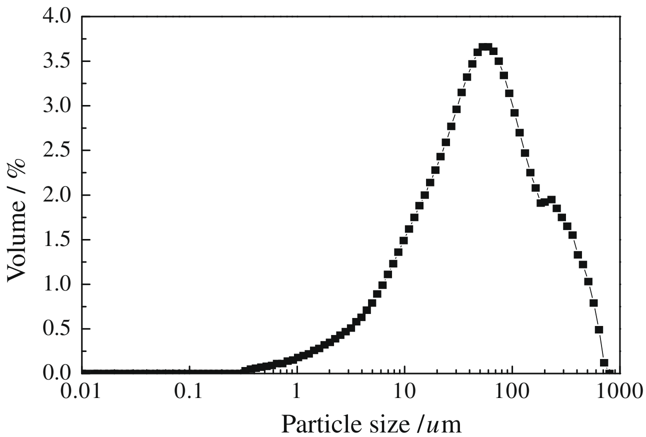


Fig. 1. The PSD of coal sample.

2. Experimental

2.1. Materials

Shenfu coal from Inner Mongolia, and algae collected from Taihu Lake in the end of May, 2007, both in China, were chosen for the study. The coal was first crushed to obtain sub-5 mm product, and then comminuted in a ball mill to produce an optimum particle size distribution (PSD). The PSD of coal sample is given in Fig. 1.

As observed in Table 1, algae, different to coal, have their own characteristics, such as high in moisture, volatile, ash and nitrogen and low in fixed-carbon and caloric value. Mixing of algae with coal to prepare coal–algae slurry can help to minimize these problems.

2.2. Pre-treatments on algae

Algae consist of cells ~5 μm wide and lined up side by side, thus form a bulk, heterogeneous system. The free space between algae forms a quite spacious uniform network of conducting intercellular water [21]. In order to destroy the network structure of algae and release intercellular water, we adopted four pre-treatment methods, which are anaerobic fermentation, high-speed shearing, chemical treatment and heating. The method of anaerobic fermentation means placing algae for a certain time under anaerobic conditions. High-speed shearing method is stirring algae at rotational speed of 1000 rpm by mechanical agitator blades for 20 min. Chemical treatment method is adding 10% NaOH (on dry algae basis) to the algae. The heating pre-treatment means the algae are heated in 50 °C thermostatic water bath for 30 min after chemical treatment.

2.3. Coal–algae slurry preparation

The milled coal and algae were mixed slowly in a vessel containing 1% (on coal basis) of the additive. If necessary, de-ionized water was added. In the experiments, sodium naphthalene sulfonate formaldehyde condensate was used as additive. The mixture was continuously stirred by a mechanical agitator at approximate 400 rpm for 20 min to ensure homogenization. The prepared slurry was allowed to stand for 5 min to release entrapped air before taking any measurements.

Algae contain nearly 4% solids, which cannot be ignored. When coal–algae slurry was being prepared, solids in algae contributed to the total solids content of the coal–algae slurry. It should be pointed out that all experiments in this study were carried out in double, and the reproducibility of the experiments was satisfactory. The data were the average of the two experiments.

2.4. Determination of rheological properties and stability of coal–algae slurries

Viscosity measurements were performed using rotating-type viscosimeter (model NXS-4C, Chengdu Instrument Factory, China). The viscosimeter consists of a cup centered on a turntable with a rotor concentrically suspended within it. When taking any determinations, the sample was placed in the gap between the inner rotor and outer cylinder. The temperature was controlled at 251 °C using a thermostatic water bath. The results, which were automatically recorded by computer, can reveal the relationships between the shear rate and the shear stress and apparent viscosity.

The changes between shear stress and shear rate can be fitted by Herschel–Bulkley model:

$$\tau = K\gamma^n \quad (1)$$

where, τ is shear stress, r is shear rate, and K and n are rheological constants, referred to as the fluid consistency coefficient and flow behavior index, respectively. For $n = 1$, this equation reduces to Newton's law of viscosity; hence, departure of n from unity indicates the degree of deviation from Newtonian behavior. The behavior is pseudo-plastic for $n < 1$ and dilatancy for $n > 1$.

The stability of CWS and coal–algae slurries were measured using rod drop method. The prepared CWS or coal–algae slurries were put into 50 ml measured cylinder. In order to prevent water evaporation, the slurry was sealed by a thin layer of paraffin wax (about 2 ml). The experiments with rod drop perform every four hours. When solids deposition appears, the rod can not fall to the bottom of the measuring cylinder. This time is defined as stability time [22].

2.5. Gasification procedures

Gasification reactivity measurements of CWS and coal–algae slurry were performed in a Termax500 thermogravimetric system manufactured by Thermo Cahn Corporation. The CWS and the coal–algae slurry (the coal–algae mass ratio of 3:2) are firstly dried at 105 °C until no weight loss, respectively. The particle size of coal is less than 100 μm. A weighed sample (about 15 mg) is placed on a crucible inside the furnace and heated under nitrogen stream up to reaction temperature in 25 K min^{-1}. When 1100 °C is reached, N_2 is replaced by CO_2 with the flow rate of 1500 ml min^{-1}. Gasification reaction is conducted at 1100 °C and 0.1 MPa until no evident weigh loss is observed. The system records automatically the

Table 1
Proximate analysis and ultimate analysis of Shenfu coal and algae.

Sample	Proximate analysis (%)				Ultimate analysis (%)				Q_d (MJ kg^{-1})
	M_{ar}	A_d	V_d	FC_d	C	H	N	S	
Shenfu coal	7.17	6.58	39.70	53.72	69.20	4.72	0.86	0.49	28.36
Algae	96.26	18.45	81.28	0.27	43.27	7.51	8.58	0.77	19.25

M_{ar} refers to moisture on received basis; A_d, V_d and FC_d refer to ash, volatile and fixed-carbon on dried basis; Ultimate analysis is also on dried basis; Q_d refers to higher caloric value.

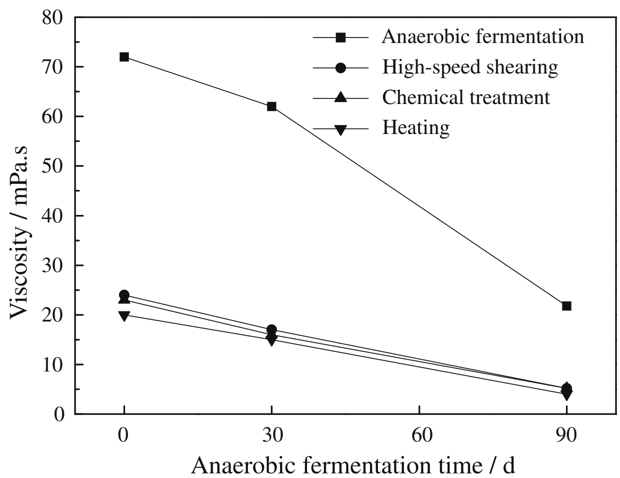


Fig. 2. Viscosities of algae processed by different treatments.

weight loss by a highly sensitive analytical balance located in the casing of apparatus. After the end of gasification reaction, system is cooled under nitrogen flow to room temperature. The purity of N_2 and CO_2 is 99.99% and 99.8%, respectively.

The time CO_2 flow replaced N_2 flow is set as the beginning time gasification reaction and the mass of sample at the time is thought as initial mass m_0. The conversion is defined as

$$x = \frac{m_0 - m}{m_0 - m_A} \tag{2}$$

Where m is instantaneous mass of sample (g) and m_A is ash mass (g). Gasification rate is defined as differential of conversion to gasification time.

$$r = \frac{dx}{dt} \tag{3}$$

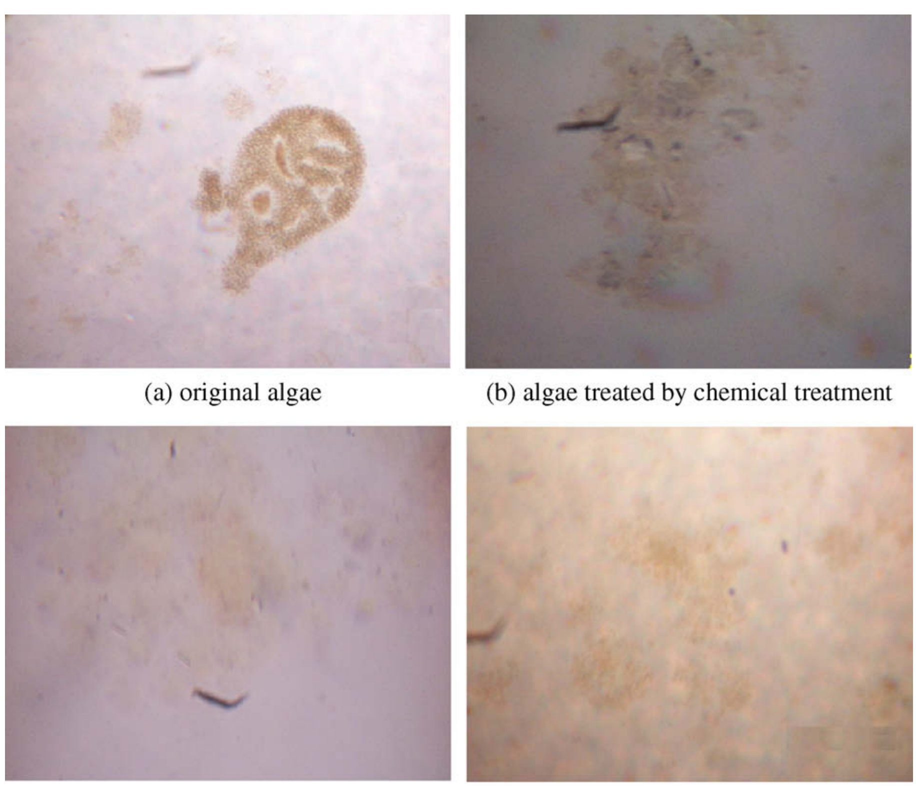
3. Results and discussion

3.1. Effect of treatments on algal viscosity

Three periods of 0 days, 30 days and 90 days were separated according to anaerobic fermentation time. At every time point, algae were treated by high-speed shearing, chemical treatment and heating, respectively. As heterogeneous system, algae exhibit non-Newtonian behavior. Viscosities at a shear rate of 100 s^{-1} were determined and depicted in Fig. 2.

It is observed from Fig. 2 that viscosities of algae processed by treatments significantly decline from original 72 mPa s. Among the methods of high-speed shearing, chemical treatment and heating, the last is the "best" method because of the lowest viscosity. Viscosity of algae processed by heating is 20 mPa s, and at 90 days time point of anaerobic fermentation, heating can make viscosity reduce to 4 mPa s.

The structures of algae observed by Nikon optical microscope (400 times) are shown in Fig. 3 An kind of network structure of free space between algae can be observed from Fig. 3a, and intercellular water is fixed by this structure. Algae have soft walls consisting of complex sugars and protein, and they are easy to be crushed [23]. After pre-treatment on algae, the original network structure which has been destroyed becomes as shown in Fig. 3b, c or d. Algal system collapses into tightly packed group of fragments, which was also found by Sokolowska [21]. A certain amount of intercellular and intracellular water becomes flowing, so the viscosity of algae decreases.



(a) original algae (b) algae treated by chemical treatment

(c) algae treated by high-speed shearing (d) algae treated by heating

Fig. 3. Structures of algae.

Table 2
The maximum solids concentrations of coal–algae slurries without de-ionized water.

	Pre-treatment method			
	Original algae	Chemical treatment	High-speed shearing	Heating
No anaerobic fermentation				
Concentration (wt.%)	48.0	52.0	54.0	55.0
Apparent viscosity (mPa s)	1006	998	1009	1067
Coal/algae ratio	46.0/54.0	50.1/49.9	52.2/47.8	53.3/46.7
30 days' anaerobic fermentation				
Concentration (wt.%)	52.5	55.0	55.5	57.0
Apparent viscosity (mPa s)	1101	1097	986	1034
Coal/algae ratio	50.7/49.3	53.3/46.7	53.8/46.2	55.3/44.7
90 days' anaerobic fermentation				
Concentration (wt.%)	57.0	57.5	57.5	58.0
Apparent viscosity (mPa s)	986	978	994	1002
Coal/algae ratio	55.3/44.7	55.8/44.2	55.8/44.2	56.4/43.6

3.2. The maximum solids concentrations of coal–algae slurries

It is different to CWS when using algae as substitutes for water to prepare coal slurries. The maximum solids concentrations of coal–algae slurries shown in Table 2 have been studied in the context of understanding the interactions of coal and algae. The maximum solids concentration is defined as solids concentration of slurry with apparent viscosity (1000 ± 100) mPa s at a shear rate of 100 s^{-1}.

It can be known from Table 2 that the maximum solids concentration of coal–algae slurry is 48.0 wt.%. When algae are previously processed by pre-treatment methods, the maximum solids concentrations obviously increase. The method of heating could make the maximum solids concentration of coal–algae slurry get to 55.0 wt.%, and at 90 days time point of anaerobic fermentation, that method could make the maximum solids concentration rise to 58.0 wt.%. By comparison Fig. 3 with Table 2, it is concluded that the lower viscosity of algae is, the bigger maximum solids concentrations of coal–algae slurries become.

Pre-treatments on algae increase the maximum solids concentrations of coal–algae slurries from 48.0 wt.% to 58.0 wt.%. But in contrast with 63.0 wt.% maximum solids concentration of CWS, the maximum solids concentration of coal–water slurry is also much lower. In order to further increase the maximum solids concentration, adding a certain amount of de-ionized water in the process of coal–algae slurries preparation is necessary. Fixing de-ionized water/algae ratio of 1:1, the maximum solids concentrations of coal–algae slurries are displayed in Table 3.

Table 3 shows that the maximum solids concentrations of coal–algae slurries are increased more than 4 percentage points comparing with de-ionized water before addition. Algae are diluted, and viscosity reduces. The method of 90 days' anaerobic fermentation could make the maximum solids concentration get to 62.5 wt.%, which is almost the same as that for CWS. At this time point, chemical treatment, high-speed shearing and heating as pre-treatment methods on algae, are ineffective on improvement the maximum solids concentrations of coal–algae slurries.

3.3. Rheological properties of coal–algae slurries

Two series of coal–algae slurries were selected to illustrate rheological properties. They are coal–algae slurries, whose concentrations are the maximum solids concentrations, and algae prepared coal slurries have been placed for 30 days under anaerobic conditions. Rheological properties of CWS and coal–algae slurries are displayed in Fig. 4.

CWS and all coal–algae slurries exhibit similar behavior. At shear rates ranging from10 s^{-1} to 100 s^{-1}, slurries exhibit non-Newtonian behavior, as characterized by a progressive decrease in viscosity with increasing shear rate. All values for n for coal–algae slurries deviate from 1.0, indicating pseudo-plastic behavior. The value of n is smaller for coal–algae slurries than for CWS. And the degree of deviation from Newtonian behavior increases.

This type of fluid behavior is typical for suspensions of microparticles [24,25]. Inside high-density coal–algae slurries, many interactions coexist, such as coal particle–coal particle, coal particle–algae, algae–algae, dispersant–coal particle and dispersant–algae. All the interactions inside coal–algae slurries lead to a kind of spatial structures formation. Different to the CWS, the algae fills to the interspaces among the coal particles, and the interactions of coal particle–algae and algae–algae become significant. When the coal–algae slurries are forced by shear rate, the spatial structure

Table 3
The maximum solids concentrations of coal–algae slurries with de-ionized water.

	Pre-treatment method			
	Original algae	Chemical treatment	High-speed shearing	Heating
No anaerobic fermentation				
Concentration (wt.%)	52.0	56.0	59.0	60.0
Apparent viscosity (mPa s)	975	1067	1028	1065
Coal/algae/de-ionized water ratio	51.0/24.5/24.5	55.2/22.4/22.4	58.2/20.9/20.9	59.2/20.4/20.4
30 days' anaerobic fermentation				
Concentration (wt.%)	60.5	61.5	61.5	61.5
Apparent viscosity (mPa s)	1003	941	921	907
Coal/algae/de-ionized water ratio	59.8/20.1/20.1	60.8/19.6/19.6	60.8/19.6/19.6	60.8/19.6/19.6
90 days' anaerobic fermentation				
Concentration (wt.%)	62.5	62.5	62.5	62.5
Apparent viscosity (mPa s)	1023	1001	1012	985
Coal/algae/de-ionized water ratio	61.8/19.1/19.1	61.8/19.1/19.1	61.8/19.1/19.1	61.8/19.1/19.1

was broken down resulting in shear-thinning. At the same time, the water in algae particles releases [26], which leads to the coal–algae slurry viscosity reducing more rapid than CWS.

3.4. Stability of coal–algae slurries

Coal–algae slurries belong to high concentration solid–liquid two phase dispersion system. Brownian motion, van der Waals or electrostatic force are not adequate to prevent the coal particles sedimentation. What can improve the stability is the spatial network inside slurries [22]. In order to show differences on stabilities between CWS and coal–algae slurries, the same samples as in Section 3.3 were also selected to represent all. Stability time of CWS and coal–algae slurries is depicted in Fig. 5.

From Fig. 5, it is obvious that stability time for coal–algae slurries is more than 70 h, much more than 4 h for CWS. For coal–algae slurries without de-ionized water addition, stability time increases

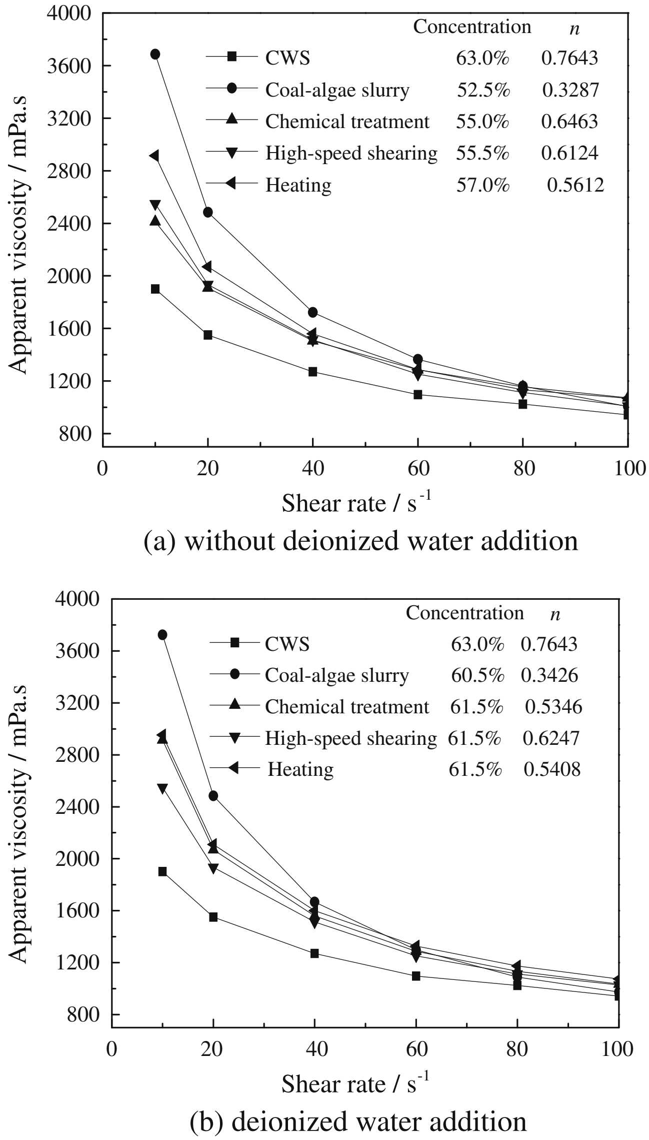


Fig. 4. Rheological properties of coal–algae slurries.

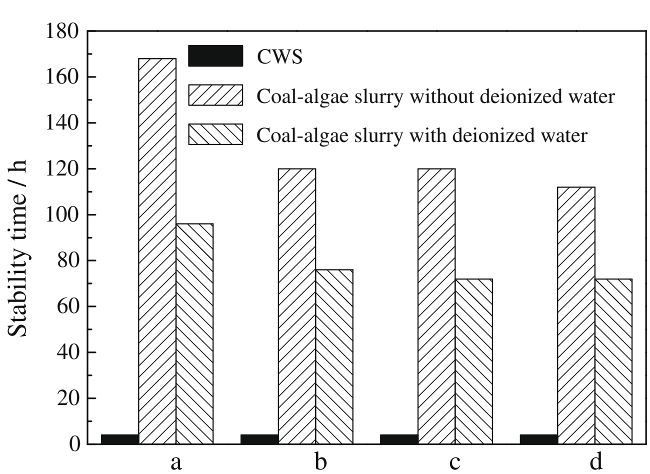


Fig. 5. Variation of stability of CWS and coal–algae slurries. (a) Original algae; (b) chemical treatment; (c) high-speed shearing; (d) heating.

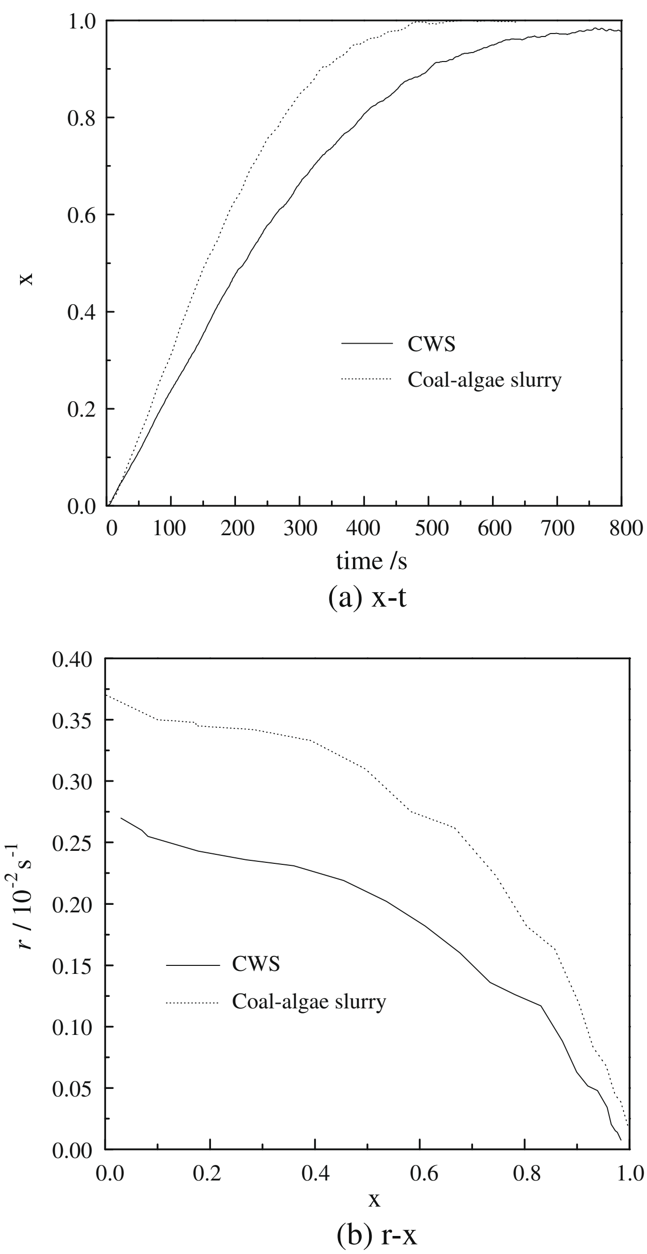


Fig. 6. Gasification curses of CWS and coal–algae slurry.

to more than 120 h. The results of the test show that algae have much greater effect on the stability of CWS.

Vlaski [11] reported that algae sedimentation relies on efficient coagulation and flocculation to produce flocs with good settling properties. Pieterse and Cloot [27] found that it is difficult to achieve such large, compact flocs from algae cells. Microalgae disperse inside slurry, and form a spatial network structure. Coal particles sedimentation is prevented by algal spatial structure.

3.5. Influence of algae on gasification reactivity of coal

Fig. 6a and b illustrate the x–t and r–x curves of coal slurries prepared with water and algae, respectively.

Gasification reactivity of coal with carbon dioxide has been widely studied. The coal gasification reactivity is usually quantified by a index R_s defined [28] as

$$R_s = \frac{0.5}{\tau_{0.5}} \tag{4}$$

where $\tau_{0.5}$ denotes the time (h) required to reach 0.5 of the fractional fixed-carbon conversion. It is calculated from Fig. 6a that R_s for CWS and coal–algae slurry are 8.4 and 11.4, respectively. Fig. 6b shows that gasification rate is higher for coal–algae slurry than for CWS at a certain fixed-carbon conversion. These results demonstrate the positive effect of algae on coal gasification. Walter Mulbry [29] reported that algae contain a certain concentrations of K, Ca, Fe and Mg, which can catalyze the gasification reaction of coal [30]. So, coal–algae slurry shows better gasification reactivity comparing with CWS.

4. Conclusion

Based on the results obtained in this investigation, a reliable method disposing algae by gasification of coal–algae slurry can be proposed. Anaerobic fermentation, chemical treatment, high-speed shearing and heating as pre-treatments on algae could obviously decrease viscosity that was favorable to improvement of the maximum solids loading for coal–algae slurries. The maximum solids loading could get to 62.5 wt.%, which was almost the same as that for CWS. Compared with CWS, the stability of coal–algae slurries was much better. All coal–algae slurries exhibited pseudo-plastic behavior. This type of fluid is shear-thinning and easy to transport. Algae can promote coal gasification rate, and gasification rate is higher for coal–algae slurry than that for CWS at a certain carbon conversion.

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