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# Effect of shear-induced breakage and reflocculation on the floc structure, settling, and dewatering of coal tailings

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**Abstract:** Flocculation is crucial for the treatment of coal tailings in industries. In this paper, the effects of shear-induced breakage and reflocculation of the floc, settling, and dewatering of coal tailings were investigated. The results show that as shear strength increases, the settling velocity of flocculated tailings decreases. A shear rate of 200 rpm (170.6 $s^{-1}$) leads to the loss of half the settling velocity. However, at high dosage cases, 200 rpm-300 rpm shear could improve the clarity of the supernatant. Small particles are flocculated preferentially, especially for particles below 10 µm. With the increase in dosage, the critical particle size for the occurrence of flocculation increases. The chaos index proposed can quantitatively reflect the degree of flocculation or reflocculation of coal tailings. At high dosage conditions, shear could enhance the dewatering performance of flocs by reconstructing the filter cake. Controlling the structure of flocs by dosage and shear strength can help obtain appropriate settling, clarifying, and dewatering performance of coal tailings.

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***Keywords:*** coal tailings, floc structure, shear, settling, dewatering

## 1. Introduction

In coal preparation plants, wet washing and screening processes produce large amounts of tailings (Hansdah et al., 2017, Li et al., 2020, Lu et al., 2019). Flocculation-based solid-liquid separation is widely employed in the treatment of coal tailings (Ma et al., 2018, Zhang et al., 2017). In industrial production, flocs are always in a dynamic balance of flocculation, breakage and reflocculation under the action of flocculants, hydraulic shear and even mechanical shear in transportation(Jarvis et al., 2005, Ofori et al., 2011). The flocculants and shear strength determine the floc characteristics and further have a profound impact on the treatment effect of tailings (He et al., 2018).

Several previous works have reported some results concerning the effect of shear rate on floc characteristics, breakage and reflocculation performance. For example, Yu et al., (2010) conducted researches on the formation, breakage, and regrowth of flocs formed by kaolin and aluminum sulfate. They found that the residual turbidity and particle number after breakage and regrowth both decreased as the applied shear increased up to 250 rpm. Higher breakage shear, such as 400 rpm, gave higher residual turbidity. He et al., (2012) investigated the effect of low shear rates on the flocculation of kaolin. They found that when G=11-16 $s^{-1}$, the decrease in floc size was caused by the irreversibility of PAC-floc breakage. Jung et al., (1996) found that low shearing rates (60-200 rpm) induced only the restructuring of the iron hydroxide flocs but that a high shearing rate (1500 rpm) induced both breakage and restructuring of the flocs. Zhang et al., (2019) figure out that an appropriate shear rate ($9s^{-1}$) produced more desirable flocs with better settling performance for Chlorella vulgaris flocs. Ghobaeiyeh, (2013)investigated the effect of shear on the flocculation of fine tailings of oil sands using an anionic flocculant. The results showed that shearing reduced the floc size, increased the floc density and compacted the floc structure. Additionally, the results suggest that controlled shearing and subsequent reflocculation can improve the dewatering and consolidation properties of flocculated fine tailings. The results indicated that breakage of flocs flocculated by a coagulant are reversible, while flocculation by a

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polymer is irreversible(Cheng et al., 2010, Moruzzi et al., 2017). Shear breakage and reflocculation are of great significance for controlling the structure, settling, and dewatering of flocs (Jarvis et al., 2005, Li et al., 2017, Ofori et al., 2011, Xu and Gao, 2012). However, most previous studies have focused on the flocs of clay(Wang et al., 2018), wastewater(He et al., 2018), soil sludge (Jarvis et al., 2005), oil sands tailings (Wang et al., 2014), or other minerals (Yin et al., 2011). Relatively little research has been conducted on the floc of coal tailings. And the relationship between floc characteristics and setting/dewatering effect is also insufficient.

The objective of this work was to provide basic research on the effect of shear-induced breakage and reflocculation on the floc structure, settling, and dewatering of coal tailings. The knowledge gained may contribute to the utilization of chemical additives and shear conditions to effectively control the characteristics of flocs and the settling/dewatering process of coal tailings to achieve desired economic and environmental benefits.

## 2. Materials and methods

### 2.1. Materials

The coal tailings sample is collected from the Sihe Coal Preparation Plant of Jincheng Coal Industry Group in Shanxi Province, China. Table 1 lists some characteristics of the sample, including the solid content, pH, zeta potential, etc. Fig. 1 shows the particle size distribution determined by a Microtrac S3500 Laser Particle Size Analyzer (America). The size of particles in the coal tailings are below 0.5 mm, and the $d_{50}$ is 59.95 μm.

Table 1. Characteristics of the coal tailing sample

| Sample | Solid content | pH | Conductivity | Zeta potential | Ash content |
|---|---|---|---|---|---|
| Coal tailings | 50 g/L | 7.8 | 1.98 ms/cm | -28.76 mV | 47.29% |

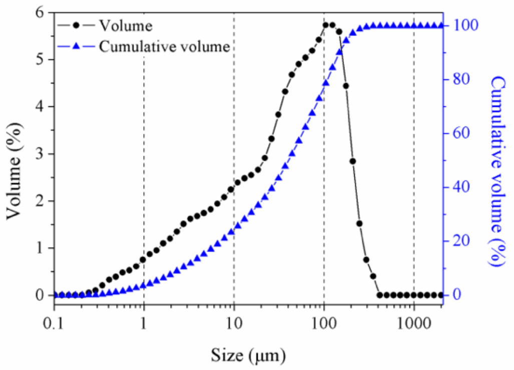


Fig. 1. Particle size distribution of the coal tailings

The X-ray diffraction (XRD) analysis of the coal tailing is shown in Fig. 2. There are many narrow, sharp and symmetrical diffraction peaks of minerals in the XRD spectrum. The mineral crystallinity is high. The baseline is low, indicating a high ash content. Compared with the diffraction peaks of the standard phase, kaolinite, quartz, calcite, illite and muscovite are found to be the main minerals in the coal tailings.

A commercial high molecular weight anionic polyacrylamide was used as the flocculant in the tests because of its domination in tailing processing of coal preparation plants all over the world, including in China, America, India, Turkey, African countries, etc. (Alam et al., 2011, Ciftci and Isık, 2017). The solution concentration of flocculant was prepared as 0.1%.

### 2.2. Experimental section

The tailings were first flocculated with the required amount of flocculant in a 250 mL glass graduated cylinder. The cylinder was inverted five times to ensure that the particles and flocculant were well mix-

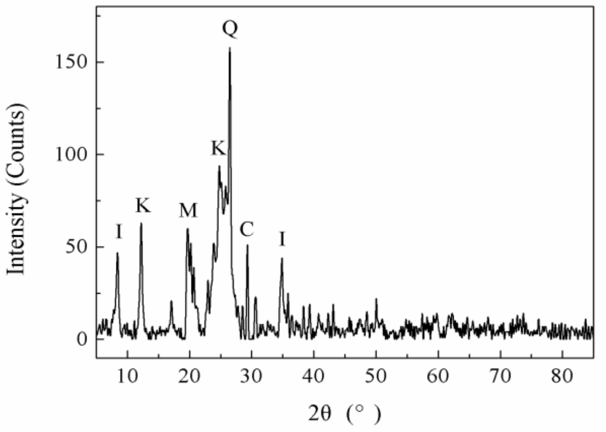


Fig. 2. XRD spectrum of the coal tailings (I-illite, K-Kaolinite, M-muscovite, Q-Quartz, C-Calcite)

ed. Then, the cylinder was laid down, the time and height of the interface between the supernatant and the particle-rich sediment were recorded to calculate the initial settling velocity.

We then transferred the mixture into a 450 mL glass beaker and stirred the mixture for 4 min at the required stirring speed. After being sheared, the mixture was put back into the graduated cylinder for reflocculation and resettling. The transmissivity of the supernatant was measured using a JH721 vis spectrophotometer (Shanghai, China). The floc size was tested three times using a Microtrac S3500 Laser Particle Size Analyzer (America).

The dewatering test was conducted in the laboratory using self-made vacuum filtration equipment. The vacuum pump pressure was set at 4kPa. The filtration time was 10 min for all samples. A more detailed description was in our previous works(Fan et al., 2015). The time and corresponding filtrate volume were recorded to calculate the filtration velocity. The shear action was produced by a two-bladed paddle. The relationship between the stirring speed and the shear rate is shown in Table 2.

Table 2. Shear rate as a function of stirring speed

| Stirring speed, rpm | Shear rate, $s^{-1}$ |
|---|---|
| 50 | 21.5 |
| 100 | 61 |
| 200 | 170.6 |
| 300 | 313.4 |
| 400 | 476.9 |
| 500 | 666.5 |
| 600 | 865.7 |
| 700 | 1090.9 |
| 800 | 1316.5 |

## 2.3. Image analysis

To obtain the morphology of flocs, we absorbed and carefully dropped the flocs on an enamel plate using a pipette with a diameter of 1 mm. Then, we used USB microscopy to capture photos of the flocs at 400x magnification. Inspired by (Droppo et al., 2008), a chaos index (CI) was defined to quantitatively evaluate the degree of flocculation of the particles: the larger the chaos index, the worse the flocculation effect.

$$CI = 100 \times OAF/PAF \tag{1}$$

where CI is the chaos index, OAF is the outline area fraction, and PAF is the particle area fraction.

We used ImageJ software to process the images captured by adjusting the threshold to obtain a binary image of high similarity with the original image. In the image, the black area represents particles or flocs (Fig. 3(a)); then, we calculated the area fraction of particles – PAF (Fig. 3(b)). Then, we extracted the outline of the black area using a black line with a proper width and calculated the outline area fraction – OAF – the line occupied (Fig. 3(c)). Finally, the chaos index (CI) was calculated according to Equation (1). Fig. 3 gives an example of the calculation of the chaos index of images of raw coal tailings; the CI was calculated as 59.73%.

The image of a filter cake was captured by the method above. The SEM images were obtained using a TESCAN MIRA3 LMH (Czech Republic).

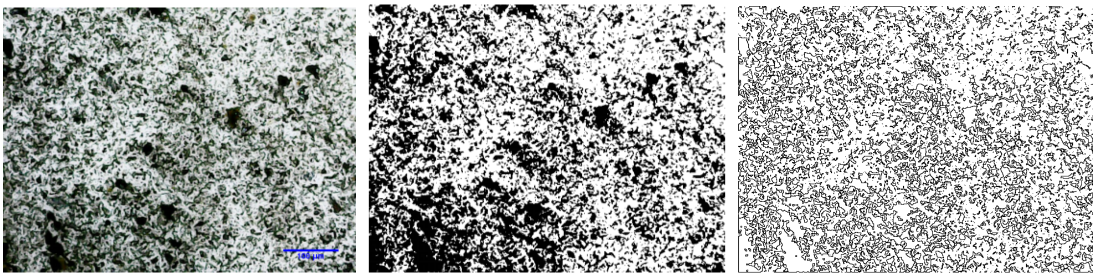

(a) Raw coal tailings
(PAF=40.577%)
(b) Binary image
(OAF=24.236%)
(c) Outline image

Fig. 3. Calculation example of the chaos index

## 3. Results and discussion

### 3.1. Initial settling velocity

Fig. 4 present the initial settling velocity as a function of dosage and stirring speed. The results show that dosage and shear strength have great influence on the settling velocity. Increasing dosage results in the increase of setting velocity, while increasing stirring speed results in the decrease of setting velocity. For the case of dosage 5 g/t, the settling velocity is low at 4.54 cm/min. After shearing, the settling velocity decreases slightly; In the case of dosage 10 g/t, the settling velocity increases to 15.14 cm/min. After shearing by 200 rpm, the settling velocity decreases by approximately 50%, and by 400 rpm, it decreases by 75%. For the case of dosages 20 g/t, 25 g/t and 35 g/t, the settling velocity is high enough to meet the need of actual production, but the settling velocity still decreases by approximately 50% after stirring by 200 rpm.

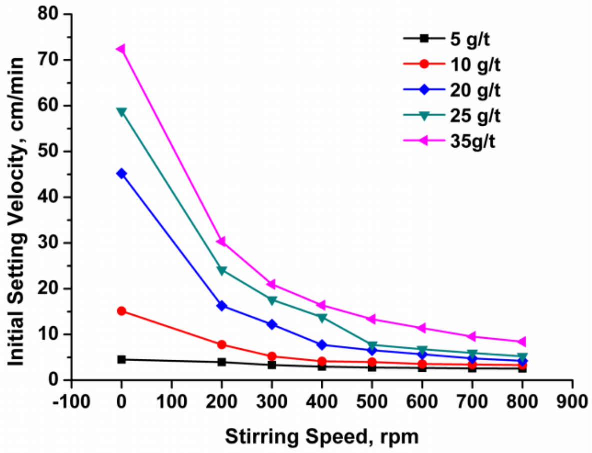


Fig. 4. Effect of stirring speed and dosage on initial settling velocity

Generally, we found that the shear strength of 200 rpm (170.6 $s^{-1}$) can lead to a loss of half the settling velocity. At different dosages, after stirring at 800 rpm, there is a deadly loss of 90% in settling velocity. At the low dosage of 5 g/t, when the stirring speed is higher than 400 rpm, the initial settling velocity in the reflocculation stage is close to that of raw coal tailings without the addition of a flocculant, which means the flocculation effect of polyacrylamide has almost disappeared. However, at a high dosage, such as 25 g/t, even after shearing at 800 rpm, the settling velocity of flocs is still higher than that of flocs at the dosage of 5 g/t without shear. These results indicate that a high dosage could play a compensation effect on velocity loss when shear breakage exists.

### 3.2. Transmissivity

Transmissivity is another important parameter to evaluate the solid-liquid separation performance, which reflects the amount of residual coal particles in the supernatant. Fig. 5 illustrates the different effects of shear strength on setting velocity and clarity. For the case of dosage 5 g/t, the transmissivity is only 14.4%, meaning there are lots of fine residual coal particles in the supernatant. After shearing and reflocculating, transmissivity decreases. The clarity of the supernatant in case of dosage 10 g/t is better than that in case of dosage 5 g/t. The transmission also decreases with the increasing stirring speed. A dosage of 20 g/t shows much better clarity, and a stirring speed of 200 rpm impairs the efficiency slightly. However, when the dosage is up to 25 g/t or 30 g/t, something different happens. At the initial flocculation stage, transmissivity become smaller compared with that in case of dosage 20 g/t. This is a common phenomenon because of an overdose of flocculant (Sabah and Erkan, 2006, Alam et al., 2011). However, transmissivity increases at the beginning and then decreases as the stirring speed rises from 200 rpm to 800 rpm. The turning point occurs at 300 rpm and 500 rpm for the dosages of 25 g/t and 35 g/t, respectively. These results mean that, though mechanical shear causes a detrimental effect on the settling velocity of coal tailing flocs, for clarifying, it facilitate matters in high dosage cases.

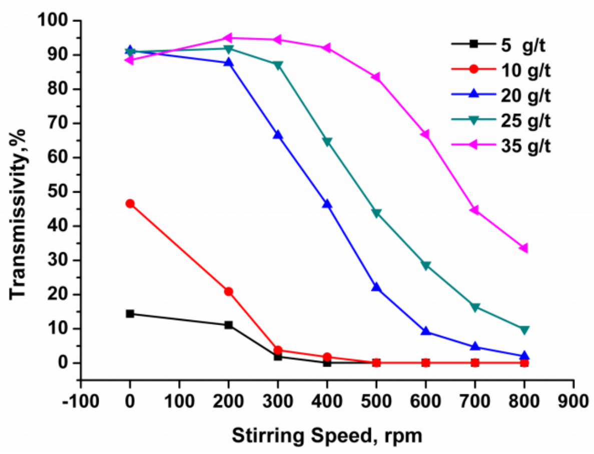


Fig. 5. Effect of stirring speed and dosage on transmissivity

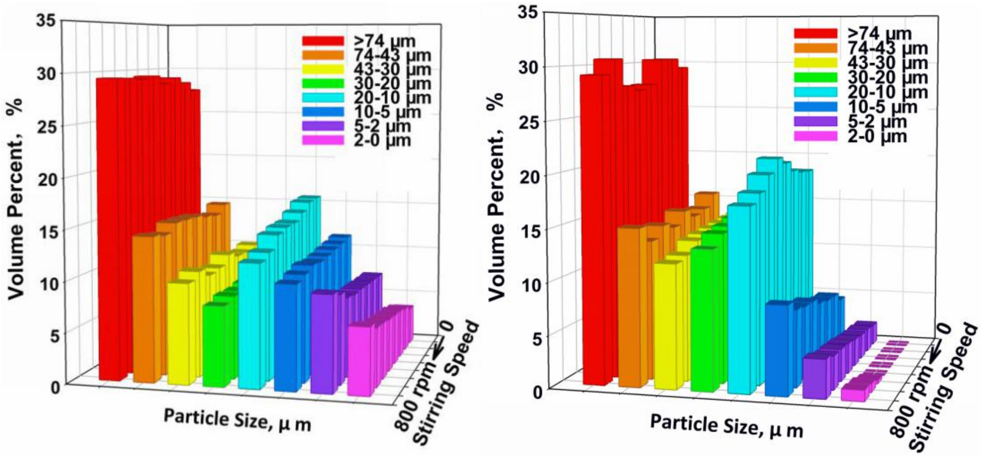
### 3.3. Floc size distribution

The floc size distribution at low (10 g/t), medium (20 g/t), and high (35 g/t) dosages with stirring speeds ranging from 200 rpm to 800 rpm were measured as shown in Fig. 6. For raw coal tailings, the volume percent of particles of sizes 0-2 µm and 2-5 µm is 7.60 % and 9.71 %, respectively. Fig. 6(a) shows that when the dosage is low (10 g/t), the volume percent of particles of sizes 0-2 µm and 2-5 µm decreases compared with raw coal tailings, and the volume percent of particles of 10-20 µm increases obviously. After shearing and reflocculating, flocs above 5 µm continue to break with the increase in stirring speed from 200 rpm to 800 rpm. Eventually, the size distribution is close to that of raw coal tailings. This indicates that, for the case of dosage 5 g/t, 5 µm is a critical size for the occurrence of flocculation. Initial flocs are mainly of sizes 5-30 µm, and after shearing, the flocs are totally broken and unrecoverable.

For the case of medium dosage (20 g/t), particles below 2 µm disappear completely, and the amounts of particles sizing 2-10 µm decreases greatly. In contrast, the volume of 10-43 µm particles increases greatly as shown in Fig. 6(b). After reflocculating, flocs of diameter 20-43 µm break into flocs of 5-20 µm below 500 rpm. When the stirring speed reaches 500 rpm, flocs of diameter 10-20 µm start fracturing, and the volume of particles of 5-10 µm in size increases. 10 µm becomes a critical size for the occurrence of flocculation in this case. Particles below 2 µm appear only when the stirring speed is up to 600 rpm. This means a medium dosage (20 g/t) can enhance the flocculation and reflocculation efficiency of fine particles, as well as the critical size value for the occurrence of flocculation.

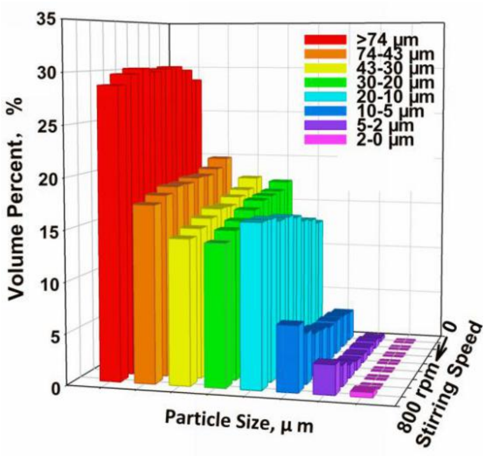
For the case of high dosage (35 g/t), there is a large decrease in the volume of particles below 10 µm, and particles below 5 µm almost completely disappear at the initial flocculation stage, as shown in Fig.

6(c). The size of flocs obtained is mainly 10-74 µm. After breaking and reflocculating, the volume percent of the 20-72 µm size range decreases. 20 µm is a critical size value for the occurrence of flocculation. Even when the stirring speed is set as 800 rpm, the volume of particles below 2 µm is very small after reflocculation.



(a) Dosage 10 g/t

(b) Dosage 20 g/t



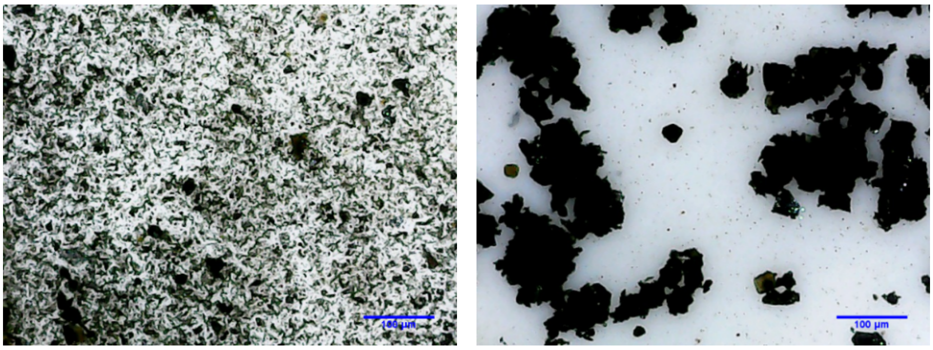
(c) Dosage 35 g/t

Fig. 6. Effect of dosage and stirring speed on floc size distribution (Note: 0 rpm represents flocs without any shear)

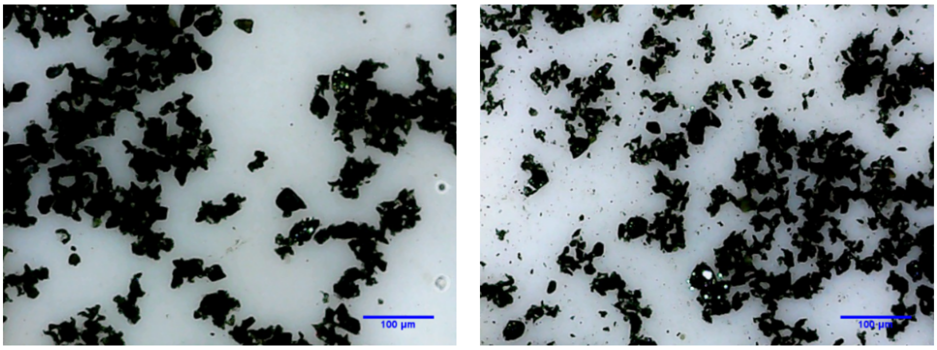
The generally accepted view contains two models of floc rupture, that is surface erosion and large-scale fragmentation caused by different stresses (Jarvis et al., 2005, Yeung and Pelton, 1996). Research findings in this paper show that large flocs are very fragile and break preferentially, mainly through large-scale fragmentation. Only when the flocs are very small and shear strength is quite high may the surface erosion model of floc rupture happen. Small particles are flocculated preferentially. With the increase in particle size, the ability to flocculate continues to weaken. Especially for particles above 10 µm, their ability to flocculate and reflocculate significantly decreases. A high dosage can facilitate a strong flocculation effect, and the critical size value for the occurrence of flocculation will increase, as well as floc size.

## 3.4. Floc morphology

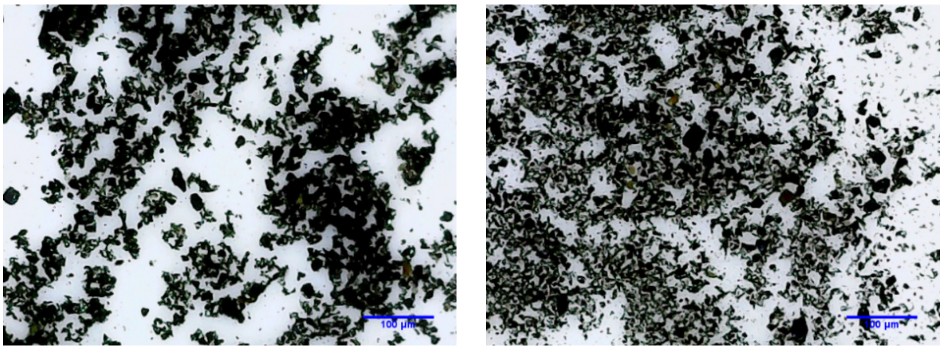
The above analysis shows that a dosage of 20 g/t can help to obtain an appropriate settling velocity and transmissivity. It is suitable for practical production. Thus, the floc morphology formed in case of dosage 20 g/t was studied by a microscopic analysis technique. Fig. 7(a) shows that raw coal tailings contain many fine particles. Its chaos index is up to 59.73. At the initial flocculation stage, fine free particles aggregate together to form large flocs (Fig. 7(b)), and the chaos index drops to 11.34. After stir-



(a) Raw coal tailings (b) Initial flocs (20 g/t)



(c) Reflocculating flocs after 200 rpm shear (d) Reflocculating flocs after 400 rpm shear



(e) Reflocculating flocs after 600 rpm shear (f) Reflocculating flocs after 800 rpm shear

Fig. 7. Images of flocs under different conditions at a dosage of 20 g/t

ring, the reflocculation effect becomes poor. In the case of 200 rpm shear (Fig. 7(c)), large flocs are broken into small flocs, with the chaos index increasing to 18.53. However, there are still no free particles. When the stirring speed rises to 800 rpm (Fig. 7(f)), flocs are badly broken, and lots of small flocs are generated. The chaos index is 43.19. The chaos index could well reflect the degree of flocculation or reflocculation of coal tailings quantitatively, and it is proportional to the stirring speed, as shown in Fig. 8.

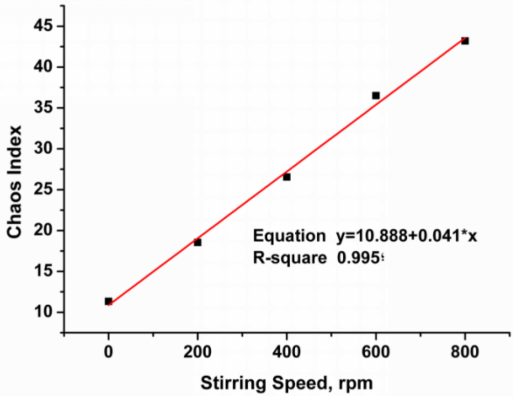
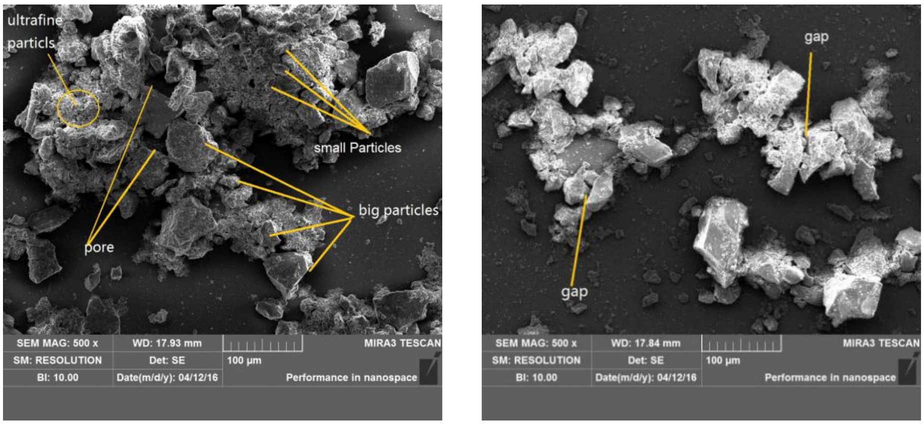


Fig. 8. Relationship between the chaos index and stirring speed

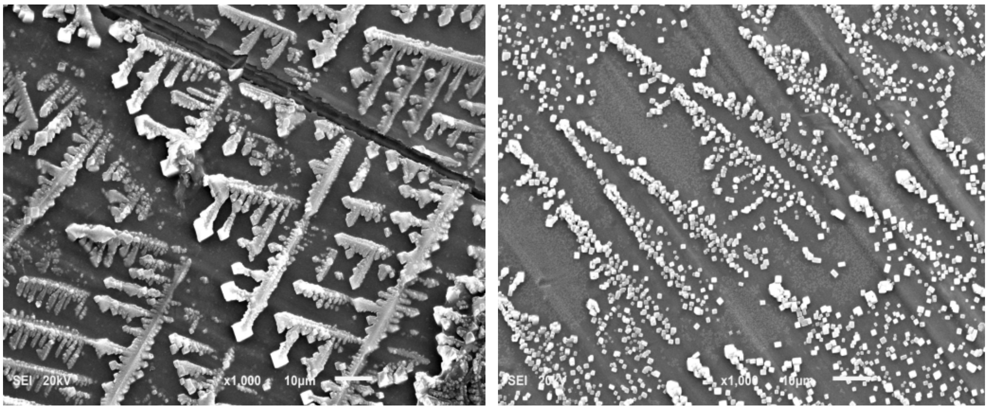
### 3.5. SEM analysis

Fig. 9 presents the SEM images of flocs before and after shearing. The flocculation effect of the flocculant on small particles is stronger than that on large particles. Fine particles tend to aggregate together easily and are close to each other with quite small gaps, which makes the flocs strong and gives them recovery ability. However, large particles can hardly bind closely and make the floc have larger pores and gaps. This gap was named by as a fragile connection (Wang et al., 2018b). Flocs containing large particles are very fragile and tend to break from the gaps. It was generally difficult to obtain large and compact flocs at the same time (Zhang et al., 2019, Ofori et al., 2011). Therefore, the flocculation and breaking characteristics of flocs are very important for the precise control of flocs.



(a) Before shear (b) After shear

Fig. 9. SEM of flocs before and after shearing



(a) Before shear (b) After shear

Fig. 10. SEM of flocculant before and after shearing

There are two main reasons for the irreversibility of floc breakage. First, polymer chains and the net structures in water are vulnerable to mechanical shear. The bonds between particles and molecular chains, as well as C-C bonds that act as the backbones of the polymer chains, are not able to bear the shearing force and then would fracture. In particular, it preferentially breaks in the middle of the molecular chain, as mentioned by (Basedow et al., 1979). At the reflocculation stage, fragmentary molecular chains will meet, collide and adsorb mutually to reform a new net by the intermolecular force. However, C-C bonds can hardly spontaneously recover. Therefore, the length of the molecular chain decreases overall. Even after a long time, it is impossible to return to the previous state. As shown in Fig. 10, shearing has obvious effect on the structure of flocculant. So, the bridging flocculation function becomes worse, and the breakage of flocs is irreversible. If the dosage of the polymer added is low, not intermolecular interaction but intramolecular association may be the main pathway between molecular chains; thus, it would lead to a worse reflocculation effect. Fig. 11 is an illustration of flocculation, breakage and reflocculation.

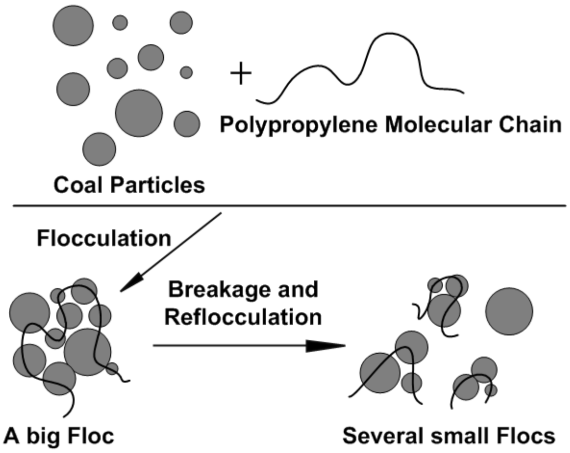


Fig. 11. Illustration of flocculation, breakage and reflocculation

Another crucial reason is the reduction of the reabsorption efficiency of the flocculant on particles. The number of effective adsorption sites on the surface of particles is limited(Blanco et al., 2005). When a polymer is mixed with coal tailings, it will adsorb onto the particles and then bring them close to each other to form flocs. When shear force is applied, the polymer chains and flocs will fracture. Meanwhile, residual adsorption functional groups with long or short molecular chains are left on the surface of the particles and occupy the effective adsorption sites. This would hinder other polymer chains from adsorbing onto the surface of particles. Therefore, the possibility of particles reforming flocs after shear decreases.

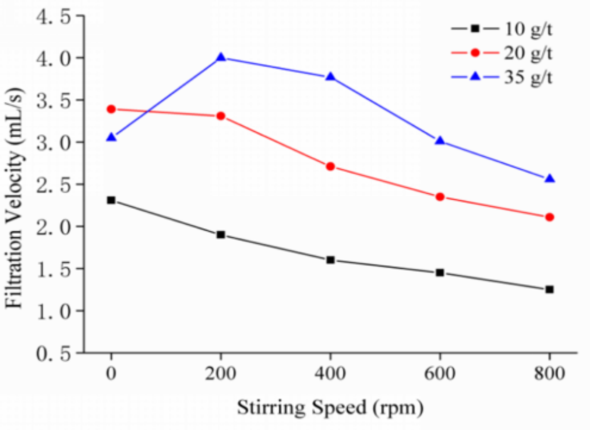
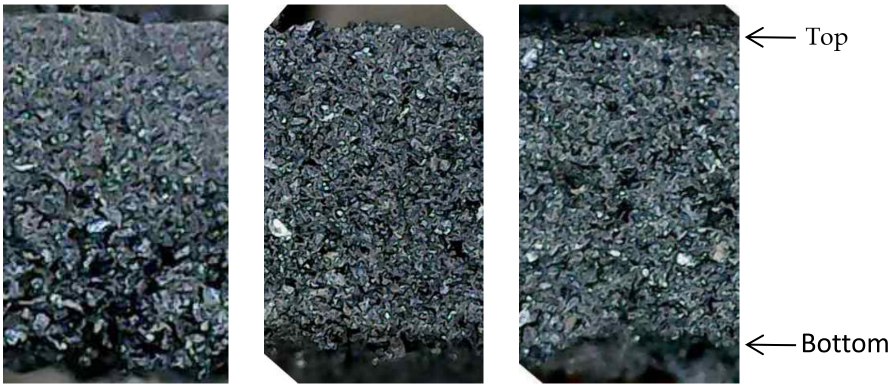


Fig. 12. Filtration velocity as a function of stirring speed and dosage

Fig. 12 presents the results of dewatering. As seen in this figure, for the case of dosages of 10 g/t and 20 g/t, increasing the stirring speed results in the decrease in filtration velocity. However, the case of the dosage of 35 g/t shows something different. Before shear, the filtration velocity at the dosage of 35 g/t is lower than that at 20 g/t, which is caused by the over dosage of flocculant. In the case of 35 g/t, with the increase in stirring speed from 200 rpm to 800 rpm, the filtration velocity first increases and then decreases. When the stirring speed is 200 rpm, the filtration velocity reaches its maximum. This suggests that shearing could enhance the dewatering of flocs formed at a high dosage. Similar conclusions have also been drawn by. further illustrate the reason, we studied the structure of the filter cake formed at a dosage of 35 g/t.

Fig. 13 shows the longitudinal section of filter cake formed by raw coal tailings without any flocculant. We found that the size of particles increases from the top to the bottom of the cake. Large particles are distributed at the bottom of cake. Fine particles are deposited on the top surface and form a compact/dense layer. This compact layer could greatly hinder the permeation of water and thus result in a poor filtering efficiency. Fine or large particles are evenly distributed in the filter cake, which is attributed to the flocs formed by the flocculant. Then, a relatively favorable distribution of pores is formed. This type of distribution of pores benefits the passage of water. After shearing, the floc is restructured. Therefore, the particles and pores in the cake are rearranged. The shear could also promote the removal of inter and intra-floc liquor to improve dewatering efficiency.



(a) Raw coal (b) Dosage 35 g/t (c) Shear with 200 rpm

Fig. 13. Filter cake

## 4. Conclusions

Coal tailings from a coal separation plant were flocculated by a polymer and then exposed to different shear strengths. The effects of shear-induced breakage and reflocculation on the floc structure, settling and dewatering of coal tailings were examined. The following are the main conclusions of this study:

- Increasing shear strength results in a decrease in settling velocity of flocculated tailings. Shear of 200 rpm (170.6 $s^{-1}$) leads to the loss of half of the settling velocity. However, in high dosage cases, 200 rpm-300 rpm could improve the clarity of the supernatant.
- Small particles are flocculated preferentially, especially for particles below 10 µm. With the increase in dosage, the critical particle size for the occurrence of flocculation increases. Large flocs break preferentially, and the model of floc rupture is mainly large-scale fragmentation caused by shear stresses.
- The breakage of flocs is irreversible. However, flocs can reflocculate to some degree, and their structures can be reconstructed. The chaos index can quantitatively reflect the degree of flocculation or reflocculation of coal tailings.
- At high dosage conditions, shear could enhance the dewatering performance of flocs by reconstructing the filter cake.
- Controlling the structure of flocs by dosage and shear can help obtain the proper settling, clarifying and dewatering performance of coal tailings.

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