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Interaction between quaternary ammonium surfactants with coal pitch and analysis surfactants effects on preparing coal pitch water slurry



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- The wetting of the OTAC, CTAC and HDBAC aqueous solution the surface of coal pitch obeys Zisman theory.
- The value of adhesion work of the OTAC, CTAC and HDBAC aqueous solution on the surface of coal pitch is basically the same.
- The mass concentration of coal pitch water slurry preparing by OTAC can reaches 62–67% and shows a pseudoplastic behavior.



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1. Introduction

Coal pitch is one of most important products in our daily life and industry [1], with 50–60% of the total yield [2]. The utiliza-

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ABSTRACT

The wetting behavior, adsorption amount and Zeta potential of aqueous solutions of the octadearyl dimethyl ammonium chloride (OTAC), cetyl trimethyl ammonium chloride (CTAC) and benzyl hexadecyl dimethyl ammonium chloride (HDBAC) on coal pitch surface were investigated. When the content of surfactant was constant, along with the growing of slurry concentration, the apparent viscosity of coal pitch water slurry (CPWS) increased. On the other side, it declined when shear rate grew. There was a liner relationship between contact angle and surface tension, so Zisman theory was well agreed with this wetting system. The adsorption amount of dispersants on coal pitch surface increased with the growing of solution concentration. The Zeta potential grew at first, and then stayed the constant.

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tion of it occurs during, among others, binder [3], impregnant [4], needle coke [5], carbon fiber [6], nanometer spherical carbon [7], paving and building materials [8]. However, great quantities of coal pitch are utilized insufficiently and the situation is expected to get worse as the development of coking industry. Therefore, it is urging research on new applications for coal pitch.

CPWS was first put forward by our research team [9,10] which came up with the concept of coal water slurry (CWS) [11,12] and

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petroleum asphalt slurry technology. A large number of experiments have revealed that dispersant plays a very important role in preparing the CPWS. Quaternary ammonium surfactant (JL-CO1) is found to be suitable dispersant to prepare CPWS. However, it is not clear what the JL-CO1 specific structure is, which is unbeneficial to investigate the CPWS. On the basis of the fact, CPWS was prepared with OTAC, CTAC and HDBAC of which all have exact structure and similarity to JL-CO1. It turned out that OTAC was an appropriate dispersant to prepare CPWS while the others are not suitable. Therefore, the rheological behavior of CPWS preparing by OTAC was investigated. In addition, the wetting behavior, adsorption amount and Zeta potential of OTAC, CTAC and HDBAC on coal pitch surface were discussed to reveal the mechanism of preparing CPWS.

2. Experimental

2.1. Materials

OTAC, CTAC, HDBAC and BTB are all produced by Aladdin Chemistry Co., Ltd., Fengxian District, Shanghai. Coal pitch was purchased from Cooking Group Company Limitied, Shanxi.

2.2. Surface tension measurements

The surface tension of surfactant aqueous solutions was measured by the Wilhelmy ring method using a surface tension meter under 298 K [13,14].

2.3. Coal pitch surface properties measurement

The composition and surface properties were investigated by determining of softening point, ultimate analysis [15], proximate analysis [16], SEM-EDS [17], XPS [18], et al., in order to investigate elementary composition, surface morphology and surface functional groups.

2.4. Contact angle measurements

The contact angles measurements of aqueous solutions on coal pitch surface were carried out with the sessile drop method by HARKE-SPCA contact angle meter [19–22]. Coal pitch plate was washed several times with first acetone and then double distilled water to remove the impurity. Within 60 s, a certain volume of droplets was dripped on the coal pitch slice until contact angle values remain constant. The trials were repeated four times by placing a droplet on the new parts of the slice. Then, a new slice was placed in the platform, and the above experimental operation was repeated four times. The standard deviation of the contact angle values did not exceed 2°. On the basis of values of surface tension, contact angle, spreading coefficient and adhesion work were figured out in order to evaluate the wettability. And the curve of $\gamma_{1g} - \cos \theta$ was carried out to find out whether this wetting property fit to Zisman theory, obtaining the critical surface tension.

2.5. Adsorption of dispersants on coal pitch surface

Dispersants used in this experiment were all quaternary ammonium surfactants. The pH being 7.5–8.5, surfactants adsorb Bromothymol blue (BTB) to form the association complex in the phosphate buffered solution. The variation in color of BTB is proportional to dispersant concentration, which can be determined using the UV spectrophotometry [23]. The experiments showed that the maximum absorption wavelength of BTB was 616 nm and absorbance declined with the increasing of surfactant dosage. Therefore, wavelength of 616 nm was determined and utilized in the follow-up experiments. A series of surfactant aqueous solutions was preparing in the laboratory, respectively. Suspension, the aqueous solution of surfactants mixed with the powdered coal pitch with a certain proportion (0.02 g/mL), stirring and centrifuging for 30 min at 298 K. Taking above 1 mL testing of upper clear liquid add into the 25 mL volumetric flask, then adding 1 mL OP-10 solution 5 mL BTB solution whose concentration is 1.6×10^{-4} mol/L and 2.5 mL buffer solution whose pH is 7.7, mixture of 0.2 mol/L monopotassium phosphate solution and 0.2 mol/L disodium phosphate solution, in sequence. Adsorption amount of dispersants on the coal pitch surface was calculated using the equation as follow:

 $\Gamma = (C_0 - C)V/G$

where Γ is adsorption amount (mg), *C* is concentration of dispersant after adsorption (mg/L), *C*₀ is original concentration of dispersing agent (mg/L), *V* is bulk of dispersant solutions (L), *G* is weight of coal pitch powder (g).

2.6. Zeta potential measurements

The measurements of Zeta Potential for the aqueous solution of surfactants on the powdered coal pitch were performed by the electrophoresis technology with the JS94H. The aqueous solution of surfactants mixed with the powdered coal pitch with a certain proportion (0.02 g/mL), oscillating under 298 K for half an hour. The standard deviation Zeta potential values did not exceed 0.5 mV [24–26].

2.7. Viscosity measurements and rheological properties determination of CPWS

Frozen coal pitches were pulverized for 3 s with high-speed disintegrating machine and sieved with 60 mesh standard sieves, which mono-disperse particles of size ranged from 0.2 to 0.3 μ m that conforms to normal distribution.

Slurries, uniform mixture of coal pitch powders and distilled water, were formed through strong agitation. To guaranteed the quality of slurries that stirred at 1000 rpm for 20 min. The viscosity measurement is performed by a rheometer (NXS-4C, China). Before measurement the slurries are allowed to stand for 5 min to release entrapped air. The shear rate range is $0-100 \text{ s}^{-1}$. The times of up run and down run are both 2 min, and the temperature is kept at 298 K.

3. Results and discussion

3.1. The performance of dispersants

As can be seen from Fig. 1, the change tendency of surface tension of OTAC was similar to that of CTAC and HDBAC, which the tendency decreased firstly and remained unchanged beyond its CMC.

3.2. The properties of coal pitch

3.2.1. Elemental analysis and industrial analysis of coal pitch

The softening point measured by Ring-Ball method was $110 \,^{\circ}$ C which belongs to high temperature coal pitch. The ultimate analysis and proximate analysis of coal pitch were taken by TQ-3 carbon hydrogen elements analysis instrument, SDTGA5000, German. The results can be seen in Table 1. The experimental results show that C, H, O, N, and S are key elements for the composition of coal pitch. Carbon is the main elements of coal pitch whose content is beyond 90%, and volatile component is higher than the others. This

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