



## Wettability modification and restraint of moisture re-adsorption of lignite using cationic gemini surfactant

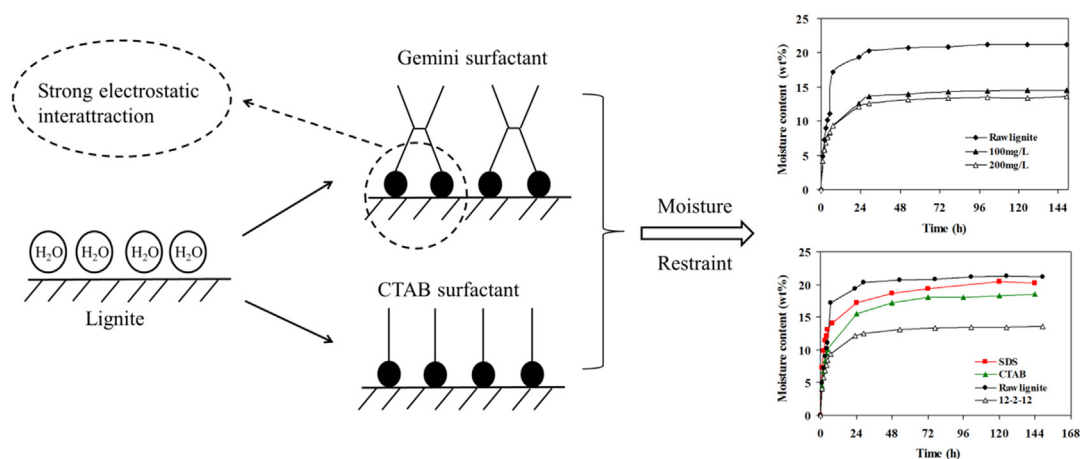


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### GRAPHICAL ABSTRACT



### HIGHLIGHTS

- Wettability modification and restraint of moisture re-adsorption of lignite using surfactant 12-2-12 were investigated.
- Electrostatic forces play a dominant role in surfactant 12-2-12 adsorption.
- The oxygen-containing functional groups of coal surface are beneficial for surfactant 12-2-12 adsorption.
- Surfactant 12-2-12 was more effective than CTAB in resisting moisture re-adsorption.

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### ABSTRACT

Adsorption of surfactants onto lignite surface may result in wettability changes and slow the re-adsorption of moisture onto dried lignite. In this work, a cationic gemini surfactant, ethanedyl-1,2-bis(dimethyl dodecyl ammonium bromide) (12-2-12), was chosen, and its adsorption characteristics and the influence on the surface wettability of lignite were investigated. The results suggest that a strong attraction takes place between the negatively charged lignite surface and a gemini surfactant with positively charged head groups to form a monolayer adsorption in low surfactant concentration. Electrostatic forces play a dominant role in the adsorption of a cationic gemini surfactant on the lignite surface while hydrogen bonding does not. The adsorption density of a gemini surfactant on coal surfaces increased with

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an enhancement in the amount of polar oxygen containing functional groups and the addition of KCl. A decrease in wetting heat indicated that the lignite treated with the gemini surfactant led to a significant decrease in the hydrophilicity of lignite. The high concentration of oxygen-containing functional groups increased the negative charge of the coal surface, which is beneficial for adsorption of the cationic gemini surfactant adsorption onto the coal surface that decreased the hydrophilicity of lignite effectively. The lignite wettability surface modification results were compared with the traditional cationic surfactant cetyltrimethyl ammonium bromide (CTAB); these results suggested that the unique chemical structure of the gemini surfactant with two hydrophilic and two hydrophobic groups had a more prominent impact on lignite surface hydrophilicity. The gemini surfactant adsorbed on lignite surface also had a significant improvement in resisting moisture re-adsorption and could create pathways toward utilizing lignite industrially.

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## 1. Introduction

Coal plays an important role in supplying primary energy, as well as the raw materials for many chemical syntheses; however, lignite sees little use in spite of its large reserves and low cost because of its higher moisture content (up to 25–40%) and low energy output relative to other coals [1]. The abundance of oxygen-containing groups is the key reason for the strong hydrophilicity of lignite. Water in lignite has a negative impact on its use due to several factors; those factors include not only drying and transportation factors but also reactions related to its combustion, gasification and liquefaction, as well as high tendency to air-slake and self-ignite [2]. Pre-drying is one possibility to enable the utilization of lignite; however, various oxygen-containing functional groups which exist on the coal surface serve as adsorption sites for moisture in the air, and the abundance of these hydrophilic groups in low-rank coal enables more water molecules to re-adsorb [3]. Moisture re-adsorption on the coal inevitably occurs and causes the loss of the drying effects [4].

Hydrothermal and thermal treatments were evaluated as possible means of upgrading and dewatering low rank coal, especially in lignite and soft bituminous coals [5,6]. Removal of the hydrophilic oxygen-containing functional groups permanently changes the surface characteristics to hydrophobic which inhibits water re-adsorption [7,8]. Nevertheless, hydrothermal and thermal treatments inevitably result in phenolic wastewaters, which are difficult to address and greatly increase costs [9]. These drawbacks mean that finding an economically attractive alternative method for low rank coal upgrading and dewatering is necessary.

One effective way of addressing the above problem is to modify the wettability of coal surfaces using surfactants. In recent years, alteration of solid surface wettability using surfactants has been intensely studied [10–13]. Various studies have been carried out on the adsorption properties of surfactants from aqueous solutions onto substrates such as quartz, silica, fabric, montmorillonite and activated carbons [14–18]. The solid surfaces are either positively or negatively charged in the aqueous medium by ionization/dissociation of surface groups or by the adsorption of ions from solution onto a previously uncharged surface. The surfactant molecules adsorbed onto solid surfaces altered the wettability of water-wet and oil-wet solid surfaces depending on the exposure of either hydrophobic groups or hydrophilic groups. The orientation and degree of wettability changes mainly depend on the types and adsorption characteristics of surfactants [19]. Crawford et al. checked the ability of three surfactants (anionic, cationic and non-ionic surfactants), to alter the surface hydrophobicity of three Australian coals, suggesting that coals of different ranks and characteristics of surfactants have significant influences on the surface wettability of the coals [20]. The existence of abundant hydrophilic oxygen-containing functional groups, such as carboxyl and phe-

nolic, in low rank coal or oxidized coal leads to the decrease of hydrophobicity of the coal surface. The types of surfactants, its adsorption and coal rank have a close relationship with coal surface wettability. It has been indicated that the mechanism of surfactants on solid surfaces occurs in two ways; the hydrophilic heads of surfactant adsorbed on the polar part of solid surface with the hydrophobic tail exposed, or the hydrophobic tails of the surfactant interacted with the non-polar part of solid surface leaving hydrophilic heads outward [21]. In general, lignite surface with strong negative charge adsorbs the positive head groups of cationic surfactants, which leads to the coal surface hydrophilicity decreasing. It is accepted that the amount adsorbed on the solid surface influences the wettability because excess adsorption may result in multilayer formation of surfactant molecules on the solid surface [19,22].

Compared with their single-tailed counterparts, the gemini surfactants that possess both two hydrophobic tails and two cationic hydrophilic heads have been demonstrated to be more efficient in their adsorption ability on solid surfaces [23–25]. Adsorption of serial cationic gemini surfactants with different length of alkyl chains on coal pitch surface had been studied, with the enhancement of hydrophilicity due to the inferior hydrophilic adsorbate [26]. Unfortunately, research in the area of wettability modification of lignite using gemini surfactants is not well-developed, therefore, this study focuses on a new, highly effective wettability alternative in the form of a cationic gemini surfactant. The unique chemical structure of gemini surfactants with two hydrophilic and two hydrophobic groups and the impact of surfactant hydrophilic heads on lignite surface carboxyl and hydroxyl polar groups probably have profound effects on lignite surface wettability changes, which could bring lignite surface wettability alteration from hydrophilicity to hydrophobicity and discourage moisture re-adsorption of dried lignite.

In this paper, a cationic gemini surfactant, ethanediyl-1,2-bis(dimethyl dodecyl ammonium bromide) (12-2-12), was chosen for lignite wettability modification. For comparison, the single-chain cationic surfactant, cetyltrimethylammonium bromide (CTAB), and anionic surfactants, sodium dodecyl sulfate (SDS) and sodium dodecyl benzene sulfonate (SDBS), were also chosen. The surfactant 12-2-12 adsorption characteristics onto lignite and its influence on coal surface wettability, as well as restraint of moisture re-adsorption of lignite were fully investigated.

## 2. Experimental

### 2.1. Materials

Yuanbaoshan lignite from China was selected for this work. The lignite was milled and sieved into <0.074 mm. The results of proximate analysis and elemental analysis are given in

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