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Simultaneously Removal of Tar and Dust by Emulsion Liquid Membrane

Simultaneously Removal of Tar and Dust by Emulsion Liquid Membrane

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Abstract. The new method for removal of tar and dust from coke oven flue gas by emulsion liquid membrane was investigated. The emulsion liquid membrane consists of kerosene as membrane solvent, L-113B as surfactant, distilled water as internal phase and external phase. Effect of different operating parameters such as L-113B concentration, volume ratio of oil phase to internal phase, volume ratio of emulsification speed were investigated on the removal efficiency of tar and dust from coke oven flue gas. Optimum operating conditions were obtained and the tar removal efficiency can reach up to 98.32% and the dust removal efficiency can reach up to 96.37%.

1. Introduction

Coke oven flue gas contains harmful substances such as polycyclic aromatic hydrocarbons, heterocyclic aromatic hydrocarbons, benzopyrene, benzene and benzene homologues. If the coke oven flue gas containing these pollutants discharge into the atmosphere, not only pollute the surrounding environment, but also threaten the health of human being [1-2].

At present, ground dust removal station technology and U-tube double suction technology are applied into removing the dust of coke oven flue gas. When using the ground dust removal station technology to remove dust, due to the high tar content in the flue gas, the surface of the bag is attached with wet mud and mixed with tar components, so that the dust on the bag is not easy to blow off and the bag is clogged. When the U-tube double suction technology is used for removal of dust, a large amount of coal gas will escape due to the opening of the furnace door.

The emulsion liquid membrane separation technology has the advantages of high mass transfer efficiency and simple operation. It is these advantages that emulsion liquid membrane separation technology is widely used in metal ion extraction [3, 4], drug extraction [5], waste treatment [6, 7].

The emulsion droplets have a large specific surface area and can be in full contact with the dust, and the oily emulsion droplets have good solubility to the tar. Therefore, the removal effect of tar and dust is much higher than that of the aqueous medium.

2. Materials and methods

2.1 Materials

Tar and dust are from a Coking Plant in Tangshan; kerosene was purchased from Tianjin Damao Chemical Reagent Co., Ltd.; L-113B was supplied by Lanzhou Refinery; xylene was purchased from Tianjin Yongda Chemical Reagent Co., Ltd.

2.2 Preparation of the W/O emulsion and operating conditions
Firstly, L-113B was dissolved with kerosene in a beaker to obtain the oil phase. Then the internal phase was added dropwise to the solution mentioned above at a homogenizer speed of 1000 rpm. After the internal phase was added dropwise, the homogenizer speed was increased to the required homogenizer speed and operating at the required homogenizer speed for 15 min to obtain a milky white color W/O (water in oil) emulsion.

Operating conditions: absorption time: 40 min; gas flow velocity of tar: 100 mL/min gas flow velocity of dust: 800 mL/min; separation stirring speed: 630 rpm.

2.3 Determination of tar content by UV spectrophotometer
0.1937g tar dissolved and set the volume to 100ml with xylene. A concentration of 1937 mg/L tar standard solution was obtained, and was diluted to concentrations of 11.62, 17.43, 23.24, 29.05, 38.70 and 46.48 mg/L, respectively.

The standard curve drawn with the tar concentration as the abscissa and the absorbance at the wavelength of 290 nm [7] as the ordinate is shown in figure1. The equation obtained by linear fitting is as below:

$$C = 0.04492A \tag{1}$$

Where C and A denote the tar concentration and the tar absorbance of the exhaust gas absorption bottle, respectively.

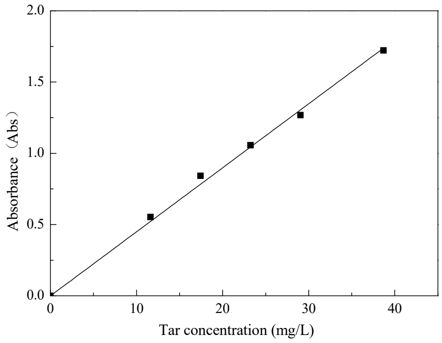

Figure 1. The standard curve of tar.

2.4 Amount determination of the tar and dust in exhaust gas.
The weighted dry degreasing cotton in the exhaust gas absorption bottle was used to measure the removal efficiency of tar and dust.

2.4.1 Amount determination of the tar in exhaust gas. After certain time absorption, the cotton was soaked with xylene to make absorbed tar dissolved. Then, xylene was filtrated in a volumetric flask. Washed cotton with xylene for several times and filtrate in the above volumetric flask each times, then volume to volumetric flask scale with xylene. The absorbance of this solution was measured at 290 nm and the concentration of the tar was determined according to the formula (1). The following formula was used to calculate the tar removal efficiency:

$$\text{Tar removal efficiency} = \frac{m_1 - C\times 4500}{m_1}\times 100\% \tag{2}$$

Where m_1, C denote the mass of tar reaction and the tar concentration of the exhaust gas absorption bottle, respectively.

2.4.2 Amount determination of the dust in exhaust gas. Dry the degreasing cotton in the exhaust gas absorption bottle and weighted. The difference weight of the degreasing cotton after and before the absorption is the content of dust in the exhaust gas. The following formula was used to calculate the dust removal efficiency:

$$\text{Dust removal efficiency} = \frac{m_2 - m_3}{m_2} \times 100\% \tag{3}$$

Where m_2, m_3 denote the mass of dust reduced in the reaction flask and the added mass of degreasing cotton, respectively.

3. Flow chart of removal of tar and dust

The experiment process for simultaneously removal of tar and dust is shown in figure 2. Air is supplied by air generator (QPA-5LP, Shanghai Lingxi Instrument Co., Ltd.). The simulated tar-containing flue gas and dust-containing flue gas were carried into the emulsion liquid membrane absorption bottle. Most of the tar and dust were removed and a small amount of tar and dust were captured by the degreasing cotton in the exhaust gas absorption bottle.

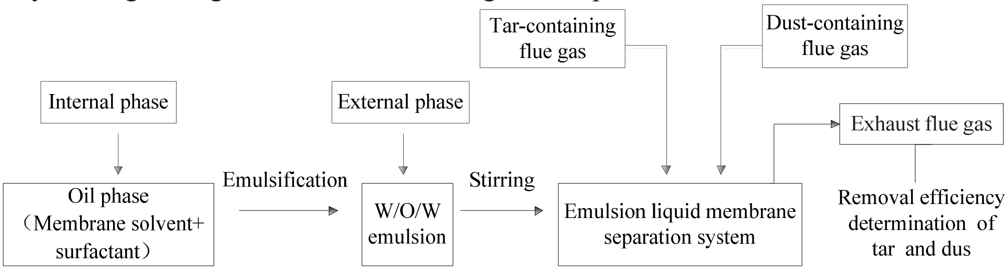

Figure 2. Flow chart of removal for tar and dust simultaneously

4. Results and discussion

4.1 Effect of L-113B concentration

The effect of L-113B concentration on the removal efficiency of tar and dust is shown in figure 3.

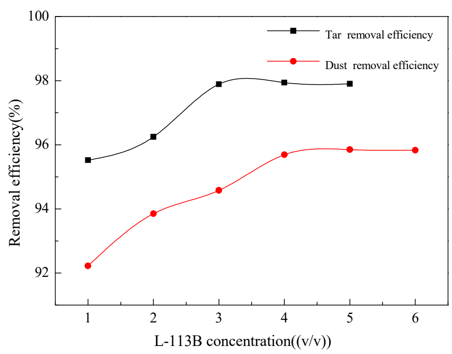

Figure 3. Effect of the concentration of L-113B on simultaneous tar and dust removal efficiency. (Experimental conditions: volume ratio of emulsion to external phase: 1:4; volume ratio of oil phase to internal phase: 3:2; emulsification speed: 4000 rpm).

As shown in figure 3, the tar and dust removal efficiency increased and then remained unchanged with the increase of l-113B concentration. At low surfactant concentration, the amount of surfactant is insufficient for surrounding all the internal aqueous phase which leads to more break-ups of emulsion droplets and instability of the membrane [8]. However, when the concentration of L-113B exceeds a certain level, excessive amount surfactant tends to increase resistance at the interface between the external phase and the oil phase, which does not favour to the transfer of the tar from external phase to oil phase. Moreover, the emulsification of the surfactant during the stirring process causes swelling phenomenon, which causes instability of the W/O system and high concentration of surfactant can worsen this phenomenon [9]. Therefore, 4% (v/v) was chosen as the optimum concentration of L-113B.

4.2 Effect of volume ratio of oil phase to internal phase

The influence of volume ratio of oil phase to internal phase on the removal efficiency of tar and dust is shown in figure 4.

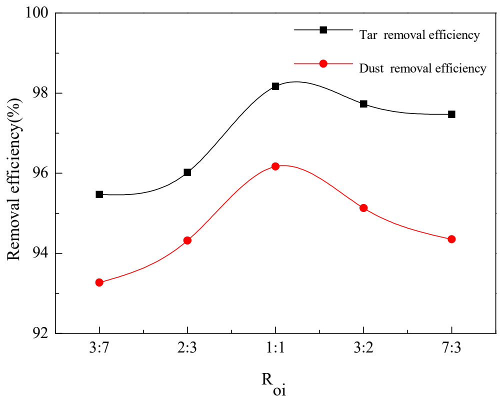

Figure 4. Effect of the volume ratio of oil phase to internal phase on simultaneous tar and dust removal efficiency. (Experimental conditions: volume ratio of emulsion to external phase: 1:4; concentration of L-113B: 4% (v/v); emulsification speed: 4000 rpm).

In figure 4, it is observed that the removal efficiency of tar and dust increases with increase of volume ratio of oil phase to internal phase from 3:7 to 1:1. The viscosity of the emulsion decreases with increase of the volume ratio of oil phase to internal phase, which leads to a decrease of the diameter of emulsion droplets [10]. This may lead an increment of the interfacial contact area between the tar and emulsion, and so lead to an increment of the chance of colliding between the dust and emulsion droplets. In addition, the amount of the membrane solvent increases with the volume ratio of oil phase to internal phase increase, which is benefit for the absorption and dissolution tar. However, the tar and dust removal efficiency decreases when the volume ratio of oil phase to internal phase is between 1:1 and 7:3. This is because the amount of L-113B increases with the increase of the volume ratio, which will aggravate the swelling of the emulsion and cause the emulsion unstable. 1:1 was selected as the best volume ratio of oil phase to internal phase.

4.3 Effect of volume ratio of emulsion to external phase

The effect of volume ratio of emulsion to external phase on the removal efficiency of tar and dust is shown in figure 5.

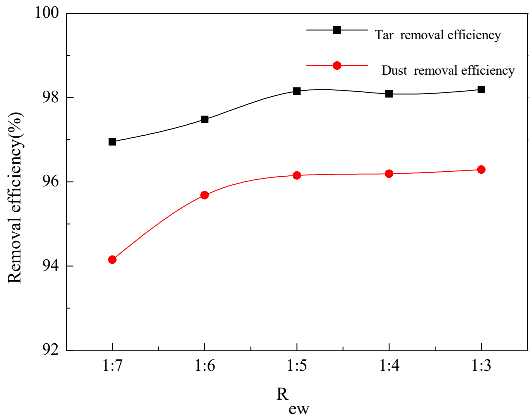

Figure 5. Effect of the volume ratio of emulsion to external phase on simultaneous tar and dust removal efficiency. (Experimental conditions: volume ratio of oil phase to internal phase: 1:1; concentration of L-113B: 4% (v/v); emulsification speed: 4000 rpm.).

It is seen that the removal efficiency of tar and dust increases with increase of the volume ratio of emulsion to external phase from 1:7 to 1:5 in the figure 5. Owing to the fact that the number of effective emulsion droplets and mass transfer area increase with the increasing amount of the emulsion [11], the diffusion of the tar and the probability of the dust colliding with the emulsion droplets enhances the removal effect of tar and dust. However, when the volume ratio of emulsion to external phase increased from 1:5 to 1:3, the tar and dust removal efficiency is almost unchanged. Continue to increase the volume ratio of emulsion to external phase might lead to the increment of the mass transfer surface area owing high number of emulsion globules forming. But the hydration surfactant formed emulsion breakage also increases for the continuous movement of water molecules into the internal phase. 1:5 was selected the optimal volume ratio of emulsion to external phase for consideration of the economy.

4.4 Effect of emulsification speed

The effect of emulsification speed on the removal efficiency of tar and dust is exhibited in figure 6.

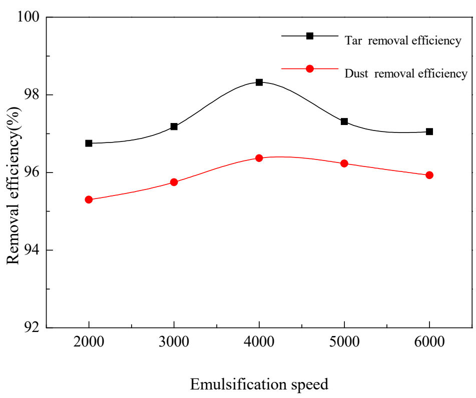

Figure 6. Effect of the emulsification speed on simultaneous tar and dust removal efficiency. (Experimental conditions: volume ratio of oil phase to internal phase: 1:1; concentration of L-113B: 4% (v/v); volume ratio of emulsion to external phase: 1:5.).

It is observed that the tar and dust removal efficiency increases as the emulsification speed increase from 2000 to 4000 rpm. The higher emulsification speed gives better mixing of surfactant, membrane solvent and internal phase and results forming smaller emulsion ball, and so enhances the stability of W/O emulsion. The smaller size of emulsion droplets with larger contact area between tar/dust and emulsion promote the removal efficiency of tar and dust. However, when the emulsification speed increases from 4000 to 6000 rpm, the tar and dust removal efficiency decreases for the too small size of the emulsion droplets will cause swelling of emulsion and lead to break-ups of the emulsion droplets, so the removal efficiency of tar and dust from flue gas reduced. Therefore, 4000 rpm was selected as the best emulsification speed.

5. Conclusion

Simultaneously removal of tar and dust by emulsion liquid membrane method is experimentally studied. The optimal operating conditions are as follows: absorption time is 40 min; gas flow velocity of tar is 100 mL/min; gas flow velocity of dust is 800 mL/min; stirring speed is 630 rpm; volume ratio of emulsion to external phase is 1:5; volume ratio of oil phase to internal phase is 1:1; L-113B concentration is 4% (v/v); emulsification speed is 4000 rpm. Under the above operating conditions, the tar removal efficiency can reach up to 98.32% and the dust removal efficiency can reach up to 96.37%. This new method has the advantages of high efficiency, simple operation and no secondary pollution.

Acknowledgement

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