Fine particle removal from flue gas using emulsion liquid membrane technique

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Abstract. A novel method for removing fine particle from flue gas by using a water in oil (W/O/W) type emulsion liquid membrane (ELM) is presented. The ELM is composed of L-113A or Span 80 serve as surfactant, kerosene as diluent, and water serve as external phase and internal phase, respectively. An L9 (3^4) orthogonal test is utilized to obtain optimum experimental conditions. The result shown that the optimum operating condition for the ELM system are as follows: L-113A serve as the surfactant with concentration of 4% (v/v), stirring speed for emulsion preparing (R) is 4500rpm, internal phase to organic phase (Rio) is 5:5 (v/v) and volume ratio of W/O emulsion phase to external phase (Reo) is 1:4 (v/v), at which the removal efficiency of fine particle from flue gas by ELM system can reach 98%.

1. INTRODUCTION
Coal is important fossil fuels, which is widely used for coal combustion power plant. In the last decade, coal consumption rate has been fast increasing around the world [1]. Only in the building sector, China's energy consumption of standard coal consumed rising from 301 to 814 million ton from 2000 to 2014 are estimated increased 1.7 times [2]. Fine particle in coal flue gas will course seriously air pollutants if directly discharged, which is the mainly course of particulate matter (PM) [3]. It not only lowers atmospheric visibility, but also seriously damages the health of human being [4].

Emulsion liquid membrane (ELM) invented by Li [5] is a useful technique for the separation and concentration target ions or compounds from dilute solution. ELM consists of an internal phase, a membrane phase and an external phase, which also called surfactant liquid membrane or double emulsion membrane. This technique is widely applied in the field of heavy metal, [6, 7]and organic compounds removal[8, 9]. ELM has many advantages of high selectivity, simple operation, high efficiency and single-stage extraction and stripping processes[10].

In this study, ELM is used for removal of fine particle from flue gas. The orthogonal test is utilized to optimize operating parameters, such as surfactant concentration, volume ratio of W/O emulsion phase to external phase (Reo), volume ratio of internal phase to organic phase (Rio) and emulsification speed.

2. EXPERIMENTAL

2.1 Materials
The ELM is composed of a surfactant, a diluent, an internal phase, an external phase. Among which surfactant are adopted span80 and L-113A, respectively purchased from Shanghai Volkswagen...
Pharmaceutical Factory and Lanzhou Refinery Factory. Commercial kerosene is purchased from Tianjin Damao Chemical Reagent Plant as diluent. Deionized water prepared by Eke Pure Water Equipment (ak-ro-up-250) is used as phase external and internal phase.

2.2 Experimental methods
Fig. 1 shows the schematic map for removal fine particle from flue gas by ELM.

FIGURE 1. The Schematic map of removal fine particle from flue gas by ELM. (1) air generator; (2) flowmeter; (3) container of coal-fired fine particle; (4) tank of fine particle removal by ELM; (5), (6) absorber of exhaust fine particle filling degreasing cotton; (7) exhaust flue gas.

The simulated flue gas is prepared by air coming from air generator (Air generator QPT-500G, Shanghai Linchy analysis instrument Co. LTD.) and fine particle coming from burning coal. The simulated flue gas with fine particle is introduced through the flowmeter controlled by quality flow controller (CS200A (5SCCM), Beijing Sevenstar Electronics Co. Ltd.) to the absorption tank of ELM. When ELM system is continuously agitated by a motor driven agitator in absorption tank, most of the fine particles blown into the ELM system through air generator are trapped in the ELM system by adherence, inertial impaction, electrostatic interaction. The small part of fine particles escaped from absorption tank is accumulated in absorber of exhaust fine particle filling degreasing cotton. The experimental conditions as follows: gas flue rate is 6L/min; the concentration of flue gas is 8000mg/m3, 50 ml of emulsion volume; 250 ml of external phase; emulsification time is 15 min.

2.3 Calculate fine particle removal efficiency
The removal efficiency (AE) for fine particle from flue gas is calculated by the formula bellow:

\[ AE = \frac{y_i - y_0}{y_i} \]  

Where, \( y_0 \) is the weight of exhaust fine particle (increased weight of cotton in absorber 5 and 6); \( y_i \) is the decreased weight of coal-fired fine particle in container 3.

3. RESULTS AND DISCUSSION

3.1 Effect of surfactant concentration
Surfactants play important role on the formation and stability of ELM. The effect of surfactant concentration in the removal of fine particle from flue gas by ELM is shown in Fig. 2.
FIGURE 2. Effect of surfactant concentration on the fine particle removal of flue gas by ELM (experimental conditions: Reo =1:5 (v/v); Rio = 5:5 (v/v); R=3000 rpm).

As seen in Fig. 2, absorption efficiency of fine particles increases as surfactant L-113A concentration increases from 1% to 4% (v/v). When surfactant concentration increases from 4 - 5% (v/v), the absorption efficiency remains a relatively stable value of 96%. The ELM does not form very well and so unstable at low concentration of surfactant. As surfactant concentration increasing in an appropriate scope, the CMC of L-113A is well-distributed on the emulsion surface. The surface tension of oil and water phase will reduce and result in more stable smaller emulsion globules, which increase the contact area of emulsion with fine particle and enhance absorption efficiency of removal of fine particles from flue gas. Therefore, excess surfactant will increase processing cost, so the surfactant concentration of L-113A is selected as 4% (v/v).

3.2 Effect of volume ratio of W/O emulsion to external phase

The volume ratio of W/O emulsion to external phase is defined as Reo. The result of the influence of the Reo on the fine particles removal from flue gas by ELM is shown Fig. 3.

FIGURE 3. Effect of volume ratio of external phase to emulsion phase on the fine particle removal of flue gas by ELM (experimental conditions: cL-113A = 4% (v/v); Rio = 5:5 (v/v); R=3000 rpm).

As shown in Fig. 3, the absorption efficiency of fine particles by ELM increases with Reo increases from 1:8 to 1:4 and reaches the highest of 95.88%. The number of plenty globules and interfacial surface area increased as Reo increased, which result in the surface area of contact with fine particles increased significantly. When Reo continuously increase above 1:4, influence isn’t obvious and the absorption efficiency of fine particle remains stable value at 95.9%. Therefore, an optimum Reo for ELM system is 1:4.

3.3 Effect of volume ratio of internal phase to organic phase

Fig. 4 exhibits the effect of volume ratio of internal phase to organic phase (Rio) on the removal efficiency of fine particle.
FIGURE 4. Effect of volume ratio of internal phase to organic phase on the fine particle removal of flue gas by ELM (experimental conditions: cL-113A = 4% (v/v); Reo =1:4 (v/v); R=3000 rpm).

As seen from the figure 4, when Rio increases from 3:7 to 5:5(v/v), the absorption efficiency of fine particles is constantly raising from 82.14 to 94.20%. While Rio continuously increases from 5:5 to 7:3(v/v), the absorption efficiency of fine particles conversely decreases from 94.20 to 85.73%. It is well known that emulsion stability is a significant factor to successful ELM system. The volume of organic phase increased as Rio increasing from 3:7 to 5:5(v/v), which results in the thick of organic membrane increasing. The strength of emulsion wall increased with the thick of membrane increase, so made it’s endure swelling of breakage and made the increase of stability of emulsion. For this reason, the removal of fine particles from flue gas by ELM increases with Rio increase from 3:7 to 5:5(v/v). Whereas, the organic phase increase and the internal phase decrease as Rio continuously increases from 3:7 to 7:3. The large Rio makes membrane become thick and make emulsions become unstable for emulsion globule swelling. Therefore, the absorption efficiency of removal fine particles decreased. And so the optimal Rio in the following experiment is determined as 5:5.

3.4 Effect of stirring speed
The effect of stirring speed for preparing emulsion on removal of fine particle from flue gas is shown in Fig. 5.

FIGURE 5. Effect of stirring speed on the fine particle removal of flue gas by ELM (experimental conditions: cL-113A = 4% (v/v); Reo =1:4 (v/v); Rio = 5:5 (v/v)).

As seen from figure 5, fine particles absorption efficiency increase with the stirring speed for emulsions preparing increase from 1000 to 4500 rpm. However, when the stirring speed increases from 4500 to 6000 rpm, the absorption efficiency of fine particles obviously decrease from 96.88% to 89.81%. This phenomenon can be explained by increased higher speed results smaller emulsion globules and so results absorption efficiency for the increasing interfacial area. But, excessive stirring speed will cause the emulsion globule rapidly swell and breakdown. Consequently, it is important to choose an optimal stirring speed at which the swelling diminish and the absorption efficiency for fine particles.
particle at a higher level. The appropriate stirring speed is selected 4500rpm in the experimental range.

4. Orthogonal test
The orthogonal experiment design is a method utilizing an orthogonal experiment table to organize experiments and analysis experimental results scientifically [11]. Orthogonal test is also an effective measurement for assaying the comprehensive effect of more environmental factors for the results, which can reduce the number of experiments associated with the analysis. Through it provides a reasonable amount of information for testing the optimal combination from a fewer number of assays [12]. Each orthogonal table has its own mark denoted as $L_n(t)^c$, where $L$ represents the orthogonal table, $n$ is the total number of experiment, $t$ is the number of levels of each factor, and $c$ is the maximum allowed number of factors [13]. An orthogonal $L_9(3^4)$ test (four factors and three levels) is used to investigate the optimal experiment condition of removal fine particle by ELM. In this experiment, A, B, C and D representing for four main factors (surfactant concentration, volume ratio of internal phase to organic phase (Rio), volume ratio of W/O emulsion to external phase (Reo) and stirring speed) respectively and their three levels are listed in Table 1.

| Level | A, Surfactant concentration (%) | B, Rio(v/v) | C, Reo(v/v) | D, Stirring speed(rpm) |
|-------|---------------------------------|-------------|-------------|------------------------|
| 1     | 4                               | 4:6         | 1:2         | 4000                   |
| 2     | 5                               | 5:5         | 1:3         | 5000                   |
| 3     | 6                               | 6:4         | 1:4         | 6000                   |

According to the Table 1, four factors and three levels of $L_9(3^4)$ orthogonal is constructed. Experiment is carried out in accordance with four factors and three levels. The number 9 of $L_9(3^4)$ stands for the number of experiments. As all possible parameter grouping are carried out, the number of experiments would be 81. Therefore, orthogonal test can significantly decrease the number of experiments. The results of the orthogonal test and difference analysis are presented in Table 2.

| Level | A     | B     | C     | D     | $Y_{jk}(%)$ |
|-------|-------|-------|-------|-------|-------------|
| 1     | 1     | 1     | 1     | 1     | 92.46       |
| 2     | 1     | 2     | 2     | 2     | 90.62       |
| 3     | 1     | 3     | 3     | 3     | 91.72       |
| 4     | 2     | 1     | 2     | 3     | 92.25       |
| 5     | 2     | 2     | 3     | 1     | 92.53       |
| 6     | 2     | 3     | 1     | 2     | 94.59       |
| 7     | 3     | 1     | 3     | 2     | 92.05       |
| 8     | 3     | 2     | 1     | 3     | 92.24       |
On one hand, according to the range of \( R_j \) from Table 2, the orders of the orthogonal test of four influential factors from large to small is D > A > B > C. Among which four influential factors are stirring speed (D), surfactant concentration (A), volume ratio of internal phase to organic phase (B), volume of ratio of emulsion phase to external phase (C), respectively. That is, one of the largest of four influential factors is stirring speed (D) of emulsion prepared.

On the other hand, it can obtain optimum value of each factor depended on the maximum and minimum value of \( R_j \). Take factor A as an example, because the orders of value from high to lower is \( \bar{y}_{11} > \bar{y}_{12} > \bar{y}_{13} \). As a result the optimum A \(_1\) value can obtain highest absorption efficiency. Meanwhile, the optimal of B, C, and D value can be obtained by the similar way, which are listed in Table 3. So, the optimal levels of each factor are \( A_1 \), \( B_2 \), \( C_3 \), \( D_1 \), respectively.

In other words, the optimal experimental conditions are \( A=4\% \), \( B=5:5\text{ (v/v)} \), \( C=1:4\text{ (v/v)} \), \( D=4000\text{ rpm} \). But in consideration of single-factor experiment, high stirring speed can reduce thickness of emulsion globules to increase the absorption efficiency, and higher volume ratio of emulsion phase to external phase can decrease emulsion swelling to increase absorption efficiency. Therefore, the optimum experiment conditions are \( A=4\% \), \( B=5:5\text{ (v/v)} \), \( C=1:4\text{ (v/v)} \), \( D=4500\text{ rpm} \).

5. CONCLUSIONS
The new removal of fine particle from flue gas by ELM technique is studied. L-113A is used as the surfactant for stabilizing emulsion. The optimum operating conditions are as follows: the surfactant concentration as \( 4\% \), volume of emulsion phase to external phase \( \text{(Reo)} \) as \( C=1:4\text{ (v/v)} \), volume of internal phase to organic phase \( \text{(Rio)} \) as \( 5:5 \text{ (v/v)} \) and stirring speed for emulsion preparing as \( 4500\text{ rpm} \). The removal efficiency can reach 98\%. This method for removal fine particle from flue gas has advantages of high removal efficiency, the reused membrane phase.

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