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Absorption kinetics analysis for removal of fine particle from simulated flue gas by emulsion liquid membrane

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Abstract. In order to study the mechanism of fine particle removal from flue gas by emulsion liquid membrane (ELM), adsorption model was investigated. Three adsorption kinetics equations (first order, second order and third order) were applied to simulate the uptake of fine particle from flue gas by ELM. The results show that the experimental data was fitted better with the pseudo-second-order equation model than those with other equation models. In order to further study the diffusion mechanism, the intraparticle diffusion model and the liquid film diffusion model were employed to study the adsorption mechanisms of ELM. The result suggests that the two stage adsorptions happened for fine particle from flue gas by ELM and these steps accord with intraparticle diffusion models.

1. Introduction

Nowadays, human can hardly survive without energy, which plays an important role to the economic growth and social development. Nevertheless, energy consumption of the world mainly stems from fossil fuels, especially coal, which can produce greenhouse gas emissions and cause environmental pollution. The global energy demand connectivity tendency leads to increase electrical power generation, while more than 30% of electric power is generated by coal combustion [1]. In the worldwide, coal consumption rate has been fast increasing for the last decade [2]. However, coal can't completely be consumed by combustion, and so cause some pollutants directly to let out to atmosphere, which can cause environment pollution and also seriously threaten to human health [3]. Among which, fine particle, i.e. particulate matter (PM), is one kind of biggest harmful pollutant. Fine particle in atmosphere not only cause serious environmental pollution for reduce visibility, but also severe threaten to the respiratory system of human being [4]. Research shows that when people long-term exposure to combustion-related air pollution can increase risk of cardiopulmonary and lung cancer mortality [5].

Emulsion liquid membranes (ELM) technique was invented by Li in 1968 and has been studied ever since [6]. For ELM have advantages of high interfacial area, high efficiency, high selectivity, simultaneous extraction and stripping in the ELM system [7], it is defined as the second generation of clean and efficient separation technology following extraction technology. ELM is a high potential method for separation and enrichment. It is wide used for removal arsenic (V) [8], phenol [9], gadolinium [7], uranium [10] and cadmium [11] from wastewater, and extraction of precious metals of palladium [12] and earth ions [13] and in hydrometallurgy. Researchers also use ELM technique to separate and enrich drugs, such as diclofenac [14] and acetaminophen [15].



2. Experimental

2.1. Materials

The ELM was composed of surfactant, diluent, internal phase and external phase. Among which L-113A was adopted as surfactant, purchased from Lanzhou Refinery Factory. Commercial kerosene (regent grade) was served as diluent purchased from Tianjin Damao Chemical Reagent Plant.

2.2. Preparation of W/O emulsion

First, a membrane phase was got by mixing L-113A with kerosene. Then, homogenous emulsion liquid was prepared by adding deionized water into the membrane phase (oil phase) in a homogenizer (FS-2, Beijing Huarui Science Equipment co. LTD) at a high stirring speed and kept stirring for a certain time. In ELM system, kerosene was used as diluent, L-113A was served as surfactant, and deionized water was used as internal phase and external phase.

2.3. Experimental method

The simulated flue gas was 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 was introduced through the flowmeter controlled by quality flow controller (CS200A (5SCCM), Beijing Sevenstar Electronics Co. Ltd.) to the vessel filling with fine particle, and the flue gas with fine particle lead into absorption tank of ELM. In absorption tank, while ELM was continuously agitated by a motor driven agitator, most of the fine particles blown into vessel through air generator were trapped by adherence, inertial impaction and electrostatic interaction. The small part of fine particles escaped from absorption tank in exhaust flue gas was accumulated in absorber filling degreasing cotton. The experimental conditions as follows: the flue rate of simulated flue gas is 6L/min; L-113A concentration in membrane phase is 4%(v/v); stirring speed of for emulsion preparation is 4000rpm and emulsification time is 15 min; volume ratio of internal phase to organic phase (membrane phase) (Rio) is 5:5(v/v); volume ratio of W/O emulsion phase to external phase (Reo) is 1:4(v/v).

2.4. Adsorption kinetics

The adsorption kinetics was carried on for fine particle removal of ELM system. The concentrations of fine particle were determined at ELM absorption time of 30, 60, 90, 120, 150, 180, 210, 240, 270 and 300min, respectively. At time t , the adsorbed amounts Q_t (mg/g) is obtained by equation (1).

$$Q_e = \frac{(C_t - C_0)V}{m} \quad (1)$$

Where C_t (mg/L) is the liquid-phase concentration of fine particle in ELM at any time t (min), V denotes the volume of ELM, m denotes the mass of ELM used, respectively.

The first order, second order and third order (Elovich) kinetic models were applied to simulate the uptake of fine particle by ELM with time t , which are given as follow equation (2), (3) and (4), respectively:

$$\ln(Q_e - Q_t) = \ln(Q_e) - k_1 t \quad (2)$$

$$\frac{t}{Q_t} = \frac{1}{Q_e} t + \frac{1}{k_2 Q_e^2} \quad (3)$$

$$Q_t = (1/\beta) \ln(\alpha\beta) + (1/\beta) \ln t \quad (4)$$

Where Q_e (mg/g) is the equilibrium adsorption capacity; k_1 (min^{-1}) and k_2 (g/mg/min) are the rate constants of the pseudo-first-order and pseudo-second-order adsorptions, respectively; α (mg/g/min) is the initial adsorption rate; and the parameter β (g/mg) is related to the extent of surface coverage and activation energy.

Moreover, the liquid film model and intraparticle diffusion model were further used for exploring the adsorption mechanisms of ELM system, and they are represented by equation (5) and (6), respectively.

$$\ln(1 - F) = -k_{fd}t \quad (5)$$

$$Q_t = k_{di}t^{0.5} + B \quad (6)$$

Where k_{fd} (min^{-1}) denotes the rate constant of liquid film diffusion; F equals to Q_t/Q_e ; k_{di} ($\text{mg.g}^{-1}.\text{min}^{-1}$) is the rate constant and B is the linear fitting intercept for intraparticle diffusion model.

3. Results and discussion

3.1. Adsorption kinetics of the ELM

The change of adsorption capacity versus time was shown in Figure 1. It could be seen from Figure 1, the adsorption capacity of the ELM increased quickly in the first 120min and then increased slowly until the equilibrium arrived at 200min. The adsorption kinetics data for the ELM system was analyzed using three kinetics models and the plots were shown in Figure 2-4, respectively. Their corresponding kinetic parameters were calculated and summarized in Table 1.

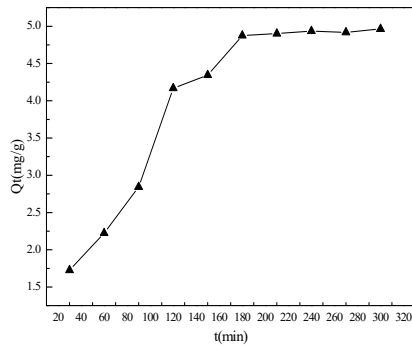


Figure 1. The variation of Q_t at different time in ELM

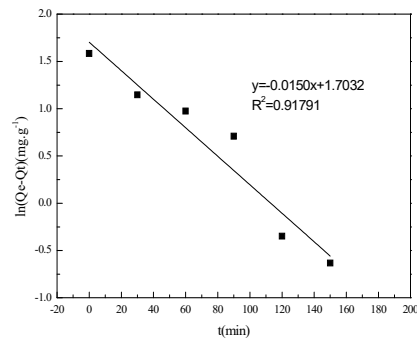


Figure 2. Linear fitting of first order kinetic equation

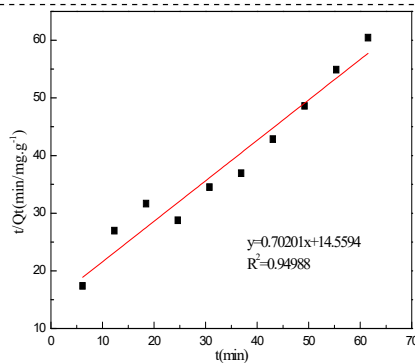


Figure 3. Linear fitting of second order kinetic equation

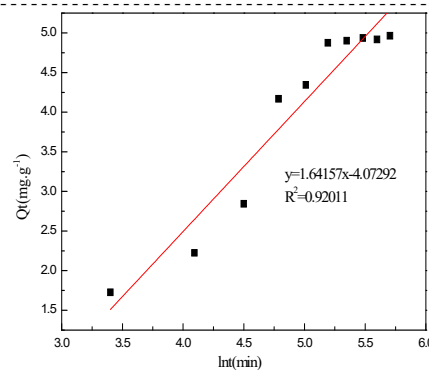


Figure 4. Linear fitting of third order kinetic equation

Table 1 Kinetic parameter

Model	Parameter		Model	Parameter	
Pseudo	$Q_e/\text{mg.g}^{-1}$	4.875	Pseudo	$Q_e, \text{fit}/\text{mg.g}^{-1}$	4.875
first order	K_1/min^{-1}	0.015	second order	$Q_e, \text{exp}/\text{mg.g}^{-1}$	1.424
	R^2	0.91791		$K_2/\text{g.}(\text{mg.min})^{-1}$	2.892×10^{-3}
Pseudo	$\alpha/\text{mg.}(\text{g.min})^{-1}$	0.1373		R^2	0.94988
third order	$\beta/\text{g.mg}^{-1}$	0.6092			

(Elovich) R^2 0.92011

As shown in Figure 2, Figure 4 and Table 1, the points in Figure 2 and Figure 4 calculated by pseudo-first-order equation and by pseudo-third-order equation are scattered and gave lower R (0.91791 and 0.94011, respectively), which suggested that pseudo-first-order and pseudo-third-order equation model are not suitable for to predict the adsorption mechanism of the absorption for fine particle by ELM. On the contrary, in Figure 3, the points near to the line and the correlation coefficient value R is 0.94988 better than in Figure 2 and Figure 4. The experimental data were fitted better with the pseudo-second-order equation than with first and third order equations. As stated above, the adsorption kinetics data for ELM system could be fitted well based on the pseudo-second-order model.

3.2. The liquid film and intraparticle diffusion model

In order to further study the diffusion mechanism, liquid film diffusion model and intraparticle diffusion model were employed to study the adsorption mechanisms of fine particle by ELM. The Fitting plots were shown in Figure 5 and Figure 6, and the statistically parameters for liquid film model and intraparticle diffusion model were shown in Table 2.

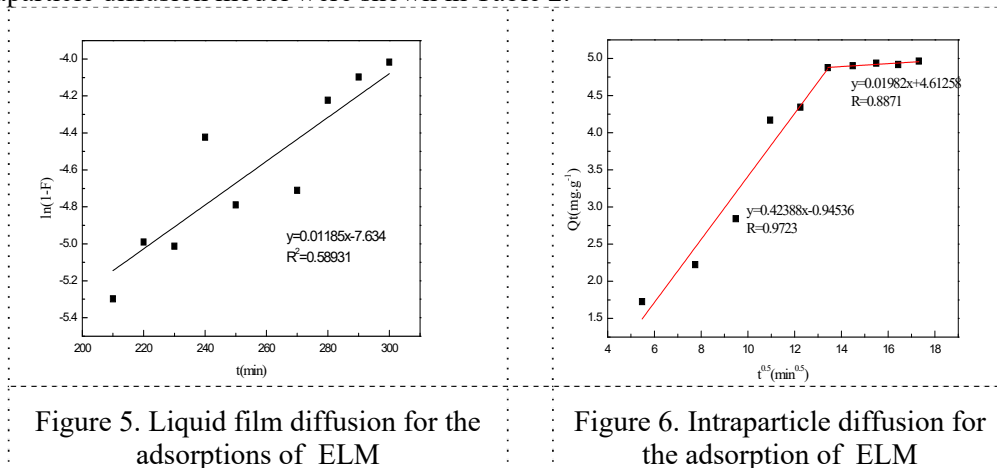


Figure 5. Liquid film diffusion for the adsorptions of ELM

Figure 6. Intraparticle diffusion for the adsorption of ELM

Table 2 model parameters of liquid film diffusion and particle diffusion model

Model	Parameter		Model	Parameter	
Liquid film diffusion model	k_{fd}/min^{-1}	0.01185	Intraparticle diffusion model	K_{d1}	0.42388
	intercept	-7.634		K_{d2}	0.01982
	R	0.7677		B_1	-0.94536
				B_2	4.61258
				R_1	0.9723
				R_2	0.8871

As shown in Figure 5, the points were much dispersed, and the correlation coefficient R was only 0.58931 as can be seen in Table 2, which indicates that the liquid film diffusion model wasn't suitable for describe the adsorption of fine particle by ELM.

As can be seen from Figure 6, the plots of Q_t versus $t^{0.5}$ was nonlinear in the whole time range, but it could be separated into two liner regions, and the correlation coefficient R were 0.9723 and 0.8871 respectively as shown in Table 2, which suggested that two stage adsorptions should be happened for ELM adsorption processes and consistent with the intraparticle diffusion model. The first liner portion of the plots denoted external mass transfer, i.e. the fine particle transported from flue gas to external phase of ELM by faster speed. The second linear portion of the plots represented that the fine particle transport from external phase of ELM to the surface of microspheres of emulsion and slowly adsorbed to approach to saturation. The second stage mass transfer is the control step.

4. Conclusion

This study explored the mechanism of removal fine particle from simulated flue gas by ELM. The adsorption of fine particle by ELM was analysed with three kinetics models. The results suggested that the pseudo-second-order model fitted well for explaining the mechanism of fine particle absorption by ELM system. In order to further study the diffusion mechanism, liquid film diffusion model and intraparticle diffusion model were employed to study the adsorption mechanisms of ELM. The results shown that two stages adsorptions happened for ELM adsorption processes. The two stages indicated that the fine particle transfer from flue gas to external phase and further transfer to the surface of microspheres of emulsion might occur simultaneously and both corresponding to intraparticle diffusion models. The second stage was the control step for fine particle absorption from flue gas by ELM.

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