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QUANTITATIVE EVALUATION OF FOULING UNDER A WELL-DEFINED SHEAR FLOW

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ABSTRACT

We analyzed the fouling phenomena under a welldefined shear flow using a concentric cylinder viscometer with a temperature controlled bath. In the study, stearic acid was crystallized in toluene. The range of shear rates studied was set from 0 to 186 s⁻¹ so that the flow is in laminar region in our system. When crystallization under cooling occurred at a supersaturation point, some of the crystals attached on the wall, resulting in an increase in thermal resistance. A simple mathematical model based on heat transfer equations was used to analyze the temperature profile of this toluene solution. From this analysis, we obtained values of thermal resistance at various conditions. It showed that fouling rate was suppressed at a high shear field. In this talk, quantitative understanding of shear effects on fouling is reported.

INTRODUCTION

Fouling is one of the general phenomena that can be found at heat exchangers, crystallizers and many other unit operations. Fouling is the precipitation of solids on a tube wall that results in increasing thermal resistance and hence the efficiency in heat transfer becomes lower. In most processes, we have to remove these attached precipitates at regular intervals so that the efficiency of the equipment can be restored. However, this cleaning process is very expensive. Thus it is more important to know how to prevent fouling in daily operations than cleaning the fouls.

Although fouling is one of the famous and familiar phenomena in our life, little is known so far. In general, we often use an averaged flow rate of fluid to analyze our fouling data (Karabelas 2002, Kazi et al. 2002). However, this interpretation is not necessarily correct because fouling occurs at the wall and the flow rate adjacent to the wall surface is usually zero in the laminar flow regime. However, the velocity gradient at the surface of the wall does depend on the overall flow rate of the fluid. This velocity gradient is called shear. Since fouling occurs at a wall, we propose that shear at the wall is one of the important factors to be considered in studying fouling phenomena.

To study the above phenomena, we used stearic acid in toluene solution in our experiments. After preparing a toluene solution saturated with stearic acid, the solution was cooled in a concentric cylinder viscometer. In this system, shear rate is easily determined. During cooling process, we measured the temperature profile of the solution and the coolant water. The values of thermal resistance were determined by the temperature profile and a set of equations which describe heat transfer between the solution and the coolant. The effect of the shear rate on thermal resistance is reported.

EXPERIMENTAL METHOD

A concentric cylinder viscometer (BROOKFIELD, RVDV-II+Pro) was used for fouling measurement. An aliquot of toluene solution saturated with stearic acid was introduced into a sample chamber (BROOKFILED, 13R). The solubility data of stearic acid in toluene solvent is shown in Figure 1. This sample chamber fits into a flow jacket and we can control the temperature of the solution. We inserted a spindle (BROOKFILED, SC4-21) which connected to the viscometer to obtain a well-defined shear field in the sample chamber. The gap between the chamber and the spindle was about 1.1 mm. We changed the shear rate from 0 to 186 s⁻¹ in this system. Coolant water was introduced into a jacket during cooling process and the temperature of the coolant water was controlled by a water-



Figure 1. Solubility data of stearic acid in toluene. The solid line represents fitting result for obtained data.

bath (Lauda, RE104). During cooling process, we measured the temperature of both the solution and the coolant, using a set of platinum resistance temperature detectors. Figure 2 shows a schematic illustration of our system.



Figure 2. A schematic illustration of the experimental system. PRDT stands for platinum resistance temperature detector.

During cooling, crystallization of stearic acid occurs at a supersaturation point and some amount of crystallized solid can attach to a cooling wall inside the chamber. It results in thermal resistance. Typical example of our data is shown in Figure 3. Crystallization occurred at 2040 s. Before that, temperature of the solution decreased at a constant rate.



Figure 3. Time evolution of temperature under shear flow. The dotted lines are the measured temperature of solution and coolant. The solid line is the calculation result using our mathematical model. The shear rate was 186 s^{-1} .

MATHEMATICAL MODEL FOR HEAT TRANSFER

We analyzed our temperature data using a mathematical model to evaluate thermal resistance. In our model as seen in Figure 4, we assumed that

- (1) no temperature distribution in solution, spindle and coolant water, respectively
- (2) cooling occurs only at the cooling wall

(3) crystallization rate is proportional to the degree of supersaturation of stearic acid, with a crystallization rate constant of k_{crys} .



Figure 4. Temperature profile assumed in our model, before the onset of crystallization (left) and right after the occurrence of crystallization (right).

Under these assumptions, we divided the experimental data into two stages, one is cooling process without crystallization and the other is with crystallization. Before crystallization, heat transfer equations are described as,

$$\frac{dT_{sl}}{dt} = \frac{h_{sl-cool}A}{V_{sl}\rho_{sl}C_{p_sl}}(T_{cool} - T_{sl}) + \frac{h_{sl-sp}A'}{V_{sl}\rho_{sl}C_{p_sl}}(T_{sp} - T_{sl})$$
(1)
$$\frac{dT_{sp}}{dt} = \frac{h_{sl-sp}A'}{V_{sp}\rho_{sp}C_{p_sp}}(T_{sl} - T_{sp})$$
(2)

After the crystallization, heating term due to the enthalpy of crystallization should be added in the right side of equation (1). Then we get,

$$\frac{dT_{sl}}{dt} = \frac{h_{sl-cool}A}{V_{sl}\rho_{sl}C_{p_sl}}(T_{cool} - T_{sl}) + \frac{h_{sl-sp}A'}{V_{sl}\rho_{sl}C_{p_sl}}(T_{sp} - T_{sl}) + \frac{k_{prec}\Delta SC_s m\Delta H_c}{V_{sl}\rho_{sl}C_{p_sl}M}$$
(3)

We obtained calculated results using a set of equations (1)-(3) as seen in Figure 3. We assumed $h_{\rm sl-cool} \sim h_{\rm sl-sp}$ in our calculation because they represent heat transfer processes between the solution and the stainless wall (the spindle or the cooling wall) under the same shear field. The value of heat transfer coefficient was determined by the slope of the temperature decrease before the onset of crystallization whereas $k_{\rm crys}$ was obtained from the sharp increase at the onset of crystallization as indicated by the arrow in Figure 3. Our result agrees well with the observed data, indicating that our model describes heat transfer process in the system quite well. On the other hand, the difference between experimental data and simulation becomes larger after 2250 s in Figure 3. Because thermal resistance caused by fouling is not considered in the above equations, one of the possible reasons for the difference is that some amount of stearic

acid crystallized at the cooling wall and it retards heat transfer between the wall and the solution. To evaluate the retardation effect in heat transfer, we modified equation (3) with a fouling coefficient β as,

$$\frac{dT_{sl}}{dt} = (1 - \beta) \frac{h_{sl-cool}A}{V_{sl}\rho_{sl}C_{p-sl}} (T_{cool} - T_{sl}) + \frac{h_{sl-cool}A'}{V_{sl}\rho_{sl}C_{p-sl}} (T_{sp} - T_{sl}) + \frac{k_{prec}\Delta SC_s m\Delta H_c}{V_{sl}\rho_{sl}C_{p-sl}M}$$
(4)

In equation (4), we assumed that fouling occurs only at the cooling wall. The value of β is unity when the heat transfer is completely retarded. The thermal resistance $R_{\rm f}$ is related to the fouling coefficient β as

$$R_{f} = \frac{1}{h} \left(\frac{1}{1 - \beta} - 1 \right)$$
(5)

In our analysis, we evaluated the fouling constant of β using equation (4) and obtained the thermal resistance of $R_{\rm f}$ from equation (5). During crystallization, the sum of dissolved and precipitated stearic acid should be constant. Then, we obtain the concentration of dissolved stearic acid during crystallization as,

$$C = C_0 - \int k_{crvs} \Delta S C_s dt \tag{6}$$

The value of $C_{\rm s}$ was determined by the solubility data shown in Figure 1.

RESULTS AND DISCUSSION

Obtained values of $R_{\rm f}$ at various shear rates are summarized in Figure 5. The retardation effect on the heat transfer during crystallization is clearly reduced when the shear rate becomes larger.



Figure 5. The effect of the shear rate on the thermal resistance $R_{\rm f}$. The solid line is a visual guide.

Because fouling occurs during crystallization process, both crystallization rate and the degree of supersaturation are also important factors to be considered. Here we defined the degree of supersaturation, ΔS , as

$$\Delta S = (C - C_s) / C_s \tag{7}$$

According to the assumption (3) in our model, the crystallization rate is proportional to the product of k_{crys} and ΔS . Results are summarized in Figure 6.



Figure 6. The effect of shear rate on the degree of supersaturation at the onset of crystallization (a) and crystallization rates (b).

Both crystallization rates and ΔS depend on the applied shear rates. At higher shear rates, crystallization rate becomes larger and ΔS becomes small. Suppose we have an energetic barrier for crystallization, like activation energy for chemical reactions, our results indicate that the barrier height for crystallization should be lowered by the applied shear rates and crystallization occurs faster. This interpretation is reasonable because it is well-known that crystallization is initiated by an "external stimulation" such as ultrasonic (Li *et al.* 2003).

Since we used a concentric cylinder viscometer for fouling measurement, viscosity of solution during crystallization was measured as seen in Figure 7. A high shear rate drastically reduces viscosity of solution.



Figure 7. The effect of shear rate on viscosity of solution.

In crystallization process, we have many parameters which would affect fouling formation. Among them, we found that two parameters, ΔS and viscosity, were main factors which clearly affect the fouling formation. In Figure 8, we summarized the effect of ΔS and viscosity on the thermal resistance R_f . Lager values of ΔS and viscosity induce high fouling resistance, suggesting that control of the degree of supersaturation and solution viscosity would be important to avoid fouling formation.



Figure 8. The effect of ΔS (a) and viscosity (b) on the thermal resistance $R_{\rm f}$. The solid lines in the figure are guides for the eye.

CONCLUSIONS

Fouling phenomenon under a simple shear field was studied using a concentric cylinder viscometer. A mathematical model was built to evaluate the thermal resistance on the cooling wall as a function of shear rate and a strong correlation was found. Shear rates surely affect the fouling formation. In addition, two parameters, ΔS and viscosity of solution during crystallization, were found to be main factors which affected the fouling formation. Further investigation should be done to clarify physical aspects of thermal resistance, such as uniformity and thickness of deposited crystal layer, and the effect of ΔS and the viscosity on $R_{\rm f}$. In this paper, we emphasize that a mathematical model, which is developed on the basis of the physical process such as heat transfer and crystallization, is quite practical to evaluate fouling.

NOMENCLATURE

- A Heat transfer area between the cooling wall and the solution , m^2
- A' Heat transfer area between the solution and the spindle, m^2
- C concentration of dissolved solute, kg/kg-solvent
- $C_{\rm s}$ saturated concentration of solute, kg/kg-solvent
- $C_{\rm p}$ heat capacity, J/kg K
- *h* heat transfer coefficient, $W/m^2 K$
- $k_{\rm crys}$ crystalization rate constant of solute, s⁻¹
- *m* mass of solvent, kg
- *M* molecular weight of solute, kg/mol
- $R_{\rm f}$ thermal resistance, m² K/W
- t time, s
- T temperature, K
- V volume, m³
- β fouling constant,
- ρ density, kg/m³
- ΔH the enthalpy of crystallization, J/mol
- ΔS the degree of supersaturation,

Subscript

- sl solution
- sp spindle
- cool coolant water

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