

COMPOSITE FOULING, FUNDAMENTALS AND MECHANISMS

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Summary

Recent research is reviewed on fouling of the process equipment used in industrial water systems where composite crystallization and particulate and corrosion fouling may be present. Crystallization or precipitation fouling is the most studied type of fouling. Other types of fouling are studied in various degrees under conditions where one type of fouling occurs in isolation. In contrast, in practical industrial applications usually several types of fouling occur simultaneously. Though some recent studies address the importance of composite fouling, in general little attention has been paid to the relative significance and the interactive effects of different fouling processes when they occur simultaneously.

1. Background

Surface fouling reduces the performance of various types of process equipment such as evaporators, condensers, and reverse osmosis units, which are used in desalination, power generation, and water purification and wastewater treatment plants. Although these processes might be very different from each other, they all suffer from similar fouling problems. Combating the fouling of process equipment costs industries billions of dollars each year and can render some processes uneconomic which are otherwise technologically and environmentally viable.

Crystallization, particulate, and corrosion fouling are usually present in industrial water

systems; however, the fouling propensity of water is routinely studied with one mechanism isolated. There have been numerous separate studies on crystallization fouling, on particulate fouling, and on corrosion fouling of various types of process equipment. These are reviewed separately in this encyclopedia and are also discussed briefly below. This paper addresses composite fouling of industrial water systems when any two of these types of fouling occur simultaneously. This is the real condition encountered in most industrial applications and an area where research is very much lacking.

Epstein's (1983) classification of fouling processes and mechanisms is used to provide a frame of reference. He classified fouling mechanisms and its types in terms of a 5×5 matrix, with five major categories and five sequences of events in each category. The types of fouling are crystallization, particulate, biological, corrosion, and chemical reaction fouling. Chemical reaction fouling, even though it can involve both organic and inorganic material, mainly involves organic material and is usually encountered in petroleum refineries and petrochemical plants. Crystallization, particulate, corrosion and biological fouling are mainly encountered in industrial water systems. Crystallization fouling is subdivided into precipitation fouling and solidification fouling. Solidification does not occur in desalination equipment because of the operating temperature of the system. Therefore, crystallization fouling is the only type of precipitation fouling present in these systems and, hence, precipitation and crystallization fouling will be used interchangeably in this document. Precipitation fouling in heat exchangers is due to the presence of concentration gradients as a result of a temperature gradient at the surface. In the case of reverse osmosis membranes, precipitation fouling is due to the presence of concentration gradients, which result from concentration polarization. Particulate fouling is present in both heat exchangers and membrane systems. Corrosion fouling may be encountered only in the presence of a corroding surface and therefore is not applicable to membrane processes (*ex situ* corrosion, which can result in the deposition of particulate corrosion products, is considered under particulate fouling). Biological fouling is present in membranes but is not present in normal evaporator surfaces because of the high operating temperatures; however, it will be present in other types of heat transfer equipment such as vacuum evaporators and condensers which operate at lower temperatures which are suitable for the growth of biological organisms. Therefore, in desalination equipment a combination of several types of fouling is present irrespective of the actual separation technique used.

In all types of fouling, five stages are present in different degrees of significance (Epstein 1983). These are initiation, transport, attachment, removal, and aging. The initiation, delay, or induction period is the time lag observed before the formation of a fouled layer takes place. Transport refers to the movement of particles or ions from the bulk fluid to the surface by mass transfer processes. Attachment refers to the adherence of deposit components to the surface, which is governed by Van der Waals and attractive double layer forces or to surface integration in the case of crystallization. Removal encompasses dislodgment of deposits from the surface to the bulk fluid. The removal process may or may not occur immediately after the deposition process has started. Aging is the alteration of a chemical or crystal structure (e.g. polymerization or dehydration) that begins immediately after the deposition process has started. Aging usually strengthens the deposit.

Since fouling mechanisms are usually synergistic, experiments are commonly designed to isolate one type of fouling. For example, on-line filters may be used to eliminate the possibility of particulate deposition when studying crystallization fouling. Recent investigations (Bansal et al. 1997; Gill and Sheikholeslami 1997; McGarvey 1999; Sheikholeslami 2000) have confirmed the presence of particulate deposition during crystallization of some species and the synergistic effects of the two processes. The interactive effects of corrosion and crystallization fouling have also been observed in some studies; corroding surfaces not only result in corrosion fouling but also affect the rate of precipitation of supersaturated salts and the resulting deposit structure (Sheikholeslami 1984; Sheikholeslami and Watkinson 1986). To reduce difficulties in the interpretation of results from complex fouling processes, corrosion resistant surfaces are usually used to isolate fouling mechanisms in studies of industrial water systems.

To understand the interactive effects of different fouling mechanisms in composite fouling, it is useful to first briefly consider the significant factors in each individual fouling type.

2. Crystallization Fouling

Precipitation fouling occurs because of the existence of a supersaturated solution. In supersaturated solutions, ionic species are transferred either by diffusion and/or by bulk transport to the solid surface. This is followed by attachment of nucleated species by integration into the crystalline deposit. Detailed reviews of experimental data, theories and models of precipitation fouling are available (Hasson 1978, 1997; Bott 1988, 1997). The effect of pH, temperature, velocity, surface material and geometry on the rate of scale formation of various salts has been shown by some investigators (Watkinson 1983; Sheikholeslami and Watkinson 1986; Bohnet 1987; Andritsos and Karabelas 1995; Andritsos et al. 1996, 1997). The effect of temperature and pH on the scale structure was also studied for some salts (Andritsos et al. 1996; Mori et al. 1996). However, the key features of crystallization fouling are most usefully discussed in terms of models that have been developed to describe the process.

Since water chemistry affects precipitation significantly, a suitable model should incorporate its effects. Of the various models that have been developed for fouling, only the model by Hasson et al. (1978) and the one derived from it by Andritsos et al. (1997), take the full water chemistry into account when predicting the rate of precipitation fouling. The latter removed the simplifying assumptions for low and high pH values from the earlier ionic diffusion model. Chan et al.'s (1989a, b) numerical model was developed for CaCO_3 based on multispecies mass transport and incorporated momentum, heat, and mass transfer for both laminar falling-films and turbulent flow in the annulus; a fully developed flow velocity was assumed for determination of a mean turbulent velocity. As Chan et al.'s (1989a, b) model does not really represent the flow behavior in the turbulent region and Andritsos et al.'s (1997) model is only an extension of Hasson et al.'s (1978) model, the original ionic diffusion model of Hasson et al. (1978) can be used with sufficient accuracy to discuss the effect of significant parameters in crystallization fouling. A drawback in all these models is the neglect of the effect of removal. The argument for this exclusion is that the strength and adherent nature of pure crystallization scales is such that removal need not be considered.

Hasson et al. (1978) developed the ionic diffusion model to predict the fouling rates of CaCO_3 . The model was based on the radial diffusion of Ca^{2+} and CO_3^{2-} ions from the bulk of the fluid followed by crystallization of CaCO_3 on the wall. Two different equations were provided to predict the deposition rate depending upon the solution pH, one for $\text{pH} < 8.5$ and the second for $\text{pH} > 8.5$. This was done to take into account the diffusion of predominant species, which are pH dependent.

For pH levels below 8.5,

$$\frac{K_1}{4K_2} \left(\frac{w}{k_r[\text{Ca}^{2+}]^2} + \frac{K_{sp}}{[\text{Ca}^{2+}]^2} \right) \left(\frac{w}{k_D[\text{Ca}^{2+}]} + \frac{[\text{CO}_2]}{[\text{Ca}^{2+}]} \right) = \left(1 - \frac{w}{k_D[\text{Ca}^{2+}]} \right) \left(\frac{[\text{HCO}_3^-]/2}{[\text{Ca}^{2+}]} - \frac{w}{k_D[\text{Ca}^{2+}]} \right)^2 \quad (1)$$

For pH levels above 8.5,

$$w = \frac{k_D[\text{Ca}^{2+}]}{2} \left\{ 1 = \frac{[\text{CO}_3^{2-}]}{k_r[\text{Ca}^{2+}]} + \frac{k_D}{k_r[\text{Ca}^{2+}]} \right\} \left\{ 1 - \sqrt{1 - \frac{4[\text{CO}_3^{2-}]}{[\text{Ca}^{2+}]} \left(1 - \frac{K_{sp}}{[\text{Ca}^{2+}][\text{CO}_3^{2-}]} \right)} \right\} \left(1 + \frac{[\text{CO}_3^{2-}]}{[\text{Ca}^{2+}]} + \frac{k_D}{k_r[\text{Ca}^{2+}]} \right)^2 \quad (2)$$

Equation (1) can be rearranged to give an explicit function for w , as is Eq. (2). This model can be modified to predict the crystallization rate of other inverse solubility salts. The modification for CaSO_4 has already been developed (Hasson 1981; Gill and Sheikholeslami 1997; Sheikholeslami 1998).

In general, the model predicts that increasing the bulk and surface temperatures will increase the rate of precipitation fouling; the surface temperature is a much more significant factor than the bulk temperature. The effect of solution pH is incorporated through the use of solubility and dissociation constants; increasing the pH decreases the solubility and enhances crystallization fouling. Where diffusion is the controlling step, the rate of fouling increases with velocity. Velocity does not affect the gross rate of deposition at higher velocities where the reaction is the controlling step. As mentioned above, the effect of shear stress and removal cannot be predicted by any of the available crystallization models. To incorporate the removal by shear stress, a generalized removal term that was originally proposed by Kern and Seaton (1959) and then modified by Taborek et al. (1972) can be used, as shown by the following equation:

$$W_{\text{removal}} \propto \frac{\tau x_{\text{deposit}}}{S_d} \quad (3)$$

3. Particulate Fouling

Particulate fouling has also been the subject of many investigations and various models have been proposed to predict the extent of particulate fouling in process equipment. Detailed reviews of the experimental results and theoretical models are available (Gudmundsson 1981; Epstein 1988, 1999; Bott 1995).

Particulate fouling may originate in two ways. It may occur due to pre-existing particulates in the process stream or by generation of particulate matter under the operating conditions of the system. Particulate matter may be generated in a supersaturated solution if crystallization takes place in the bulk fluid or if already deposited material on the surface is released to the bulk fluid by erosion or by sloughing off. Therefore, supersaturation can contribute to particulate fouling as well as crystallization fouling. In particulate fouling, particles may be of colloidal size where diffusion is the mechanism of transport and attachment is controlled by Van der Waals forces, electric double layer, and Born energies. The resistances to diffusion and surface attachment processes are in series. Transport is one of the better-understood stages of fouling and the deposition flux for particulate fouling is represented by the following equation:

$$w_{part} = k_{transport} (c_{p_{bulk}} - c_{p_{surface}}) \quad (4)$$

For submicron sized particles, the transport coefficient, $k_{transport}$, is the well-known mass transfer coefficient which can be calculated from well-established equations in the literature (Treybal 1980).

For larger particles, the inertial and impaction forces become dominant; therefore, in this case, $k_{transport}$ is not equal to the mass transfer coefficient. Figure 1 is a representation of change in the transport coefficient with particle size. The same order of magnitude for particle size (the abscissa in Figure 1) is applicable to both liquids and gases. The frictional velocity U^* is obtained by Eq. 5.

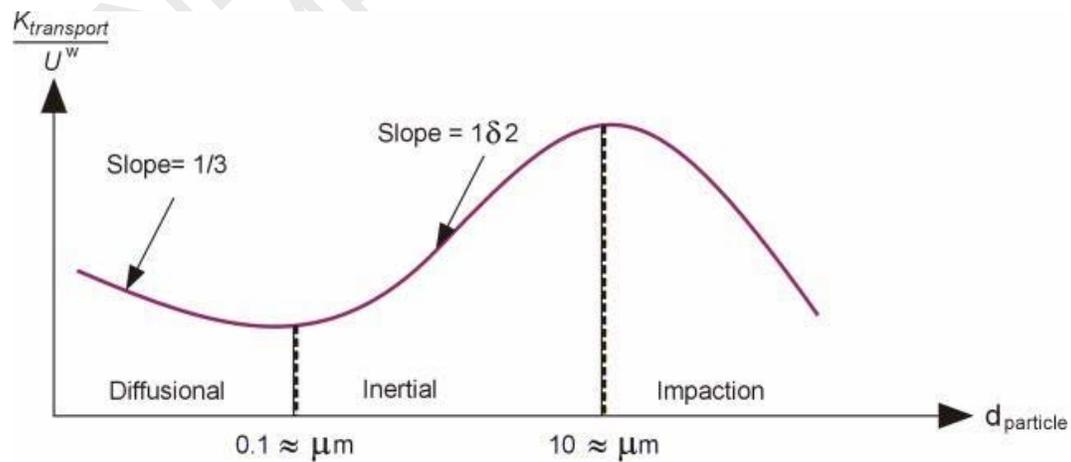


Figure 1. Three regimes of particle transport.

$$U^* = U_b \sqrt{f/2} \quad (5)$$

The rate of attachment may be approximated (Epstein 1988a) using a factor called the sticking probability S_p , which represents the fraction of all the particles transported to the surface, which stick to the surface or via an attachment coefficient. Therefore, the following equation will define the rate of attachment for particulate fouling:

$$w_{part} \approx S_p k_{transport} c_{p,bulk} = k_{r_a} c_{p,bulk} \quad (6)$$

The attachment coefficient,

$$k_{r_a},$$

should be determined experimentally. The sticking probability can be considered as unity when particles and surface have zeta potential of opposite sign. However, even in this case, when the surface is covered by particles then the zeta potential of the surface will become that of the particles and the sticking probability will decrease. The coefficient for the rate of attachment is temperature dependent and follows an Arrhenius relationship. For colloidal particles,

$$k_{r_a}$$

is determined using a more theoretical approach using attractive and repulsive forces, which results in a relationship of the following form:

$$k_{r_a} \propto e^{-E/RT}, \quad (7)$$

Since the value of

$$k_{r_a}$$

is not currently available in the literature, for composite fouling, an approximation can be obtained by comparing the filtered and unfiltered runs under otherwise identical operating conditions and calculating

$$k_{r_a}$$

from the difference in the rate of deposition for the filtered and unfiltered runs. If particulate fouling exists in isolation

$$k_{r_a}$$

can be approximated from the deposition rate for conditions where the process is attachment controlled. Where transport is the controlling mechanism, the value of

k_{r_a}

is of little significance.

The total rate of particulate deposition can be obtained by assuming resistances in series. Therefore, the particle deposition rate is given by the following expression, which was cited by Epstein (1988):

$$w_{part} = \frac{c_p}{\frac{1}{k_{D_p}} + \frac{1}{k_{r_a}}} \quad (8)$$

This equation shows that under some circumstances particle deposition is greatly enhanced by increasing the surface temperature. Particulate concentration and diffusivity are of significance as well. Increasing the velocity would increase the deposition at low velocities where transport is the controlling factor. At higher velocities, the rate of attachment controls deposition. Particulate deposition usually results in a deposit that is not very adherent and rather prone to removal. Therefore, use of a removal term is necessary and Eq. (3) can be applied to this case as well. The effect of removal becomes significant at high shear stresses and for thick deposits. This would usually result in an asymptotic fouling behavior for particulate fouling.

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