

Fouling of Tube Surfaces: Modeling of Removal Kinetics

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Fouling of tube surfaces from flowing solutions or suspensions is examined. A new approach to the removal aspects of this process is presented linking removal to the morphological and mechanical characteristics of deposit microstructure. A simplified description of deposit microstructure is employed, involving a collection of geometrically similar roughness elements. A dynamic situation is envisioned whereby these elements grow due to deposition and suffer breakage under the action of hydrodynamic forces. A mathematical model for the macroscopic fouling behavior based on the techniques of population balances is obtained and numerically analyzed. The behavior found is in agreement with experimental evidence.

Introduction

The development of deposits on surfaces exposed to flowing solutions or suspensions is usually a serious problem in industry and an important problem in general. From a macroscopic point of view, two processes can be identified which compete to determine the temporal evolution of the deposit characteristics. The first process is the *deposition*, which combines the transport of fouling species from the bulk of the fluid to the surface and their incorporation to the growing deposit. The second process is the *removal* of the deposited material in the form of particles or particle clusters or deposit fragments by the action of hydrodynamic forces or thermal stresses. The competition between these two processes can result in a variety of behaviors as far as the macroscopic characteristics of the deposit are concerned. Further elaboration on the steps involved in fouling and the various regimes can be found in the reviews by Epstein (1983, 1988). In this article special attention is paid to fouling of tube surfaces, and the related resistance to heat transfer.

Of the two processes mentioned above, deposition has received relatively more attention. Thus, although the subject is far from closed, deposition models exist for particulate (cf. Papavergos and Hedley, 1984) and crystallization fouling (Hasson et al., 1978). In certain cases these models are in reasonable accord with the experimental data obtained during the early stages of deposit development where removal is still

unimportant (Watkinson, 1983). On the other hand, little has been said about the removal process. It is accepted that the main factors causing deposit removal are the hydrodynamic forces exerted by the flowing fluid and the thermal stresses developed when temperature gradients exist. However, it is not yet clear when the above factors are sufficient to cause removal to a significant extent (Sutor et al., 1977), or what are the mechanisms by which they act. Needless to say, the empirical relations that are used to describe their effect have little predictive value.

The only theoretical approach that has been proposed to explain the action of hydrodynamic forces is the turbulent burst model by Cleaver and Yates (1973). The deposit here is viewed as a collection of particles adhering on the substrate, and the lift forces exerted on them by the turbulent bursts are considered responsible for their dislodging and subsequent removal. Turbulent bursts are idealized as viscous stagnation-point flows distributed periodically over the substrate, and information about their frequency and spatial distribution is used to obtain expressions for the removal rates. However, it may be observed that there exist lift forces due to inertial effects, which act continuously as opposed to the intermittent turbulent bursts. The former are considered unimportant by Cleaver and Yates, while in fact, they are comparable in magnitude for a wide range of particle sizes. They are usually confused with the Saffman lift force (Saffman, 1965) but the correct form is given by Leighton and Acrivos (1985). Moreover, the drag forces on attached particles (O'Neill, 1968), which also act

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continuously, are much larger in magnitude than the lift forces, and they are effective in detaching particles. This is verified by hydrodynamic and detachment experiments (Visser, 1976; Van den Tempel, 1972; Sharma et al., 1992) where drag and centrifugal forces are found to be of comparable magnitude. These observations, together with the experimental evidence supplied by Yung et al. (1989), where particle detachment was found to be initiated by rolling rather than lift-up, cast doubt on the significance of the Cleaver and Yates model.

Given an expression for the deposition rate, J_D , a balance for the deposit mass per unit area of substrate can be written in the form:

$$\frac{dm_i}{dt} = J_D - J_R \quad (1)$$

The most widely used expression for the removal rate, J_R , is that proposed by Kern and Seaton (1959); see also Taborek et al. (1972), where it is taken to be proportional to the deposited mass and the mean shear stress exerted by the flowing fluid. This is expressed as:

$$J_R = k\tau_w m_i / \psi \quad (2)$$

where m_i is the deposit mass, ψ is the so-called deposit strength, and k is a proportionality constant. When this expression is introduced into the deposit mass balance it predicts that the deposit mass approaches an asymptotic value exponentially, provided that the deposition rate is constant. In many instances experimental data have been fitted reasonably well with that expression (Taborek et al., 1972; Morse and Knudsen, 1977).

However, many objections can be raised against such an approach. First and most important, no obvious method presents itself for predicting what the deposit strength is, other than fitting to experimental data. Furthermore, it is not clear what is the precise physical meaning of "deposit strength" and how it can be measured. The same comments can also be made about the proportionality constant k . Secondly, there is no evidence that under conditions of no deposition the deposit mass will decline exponentially as suggested by Eqs. 1 and 2. In fact, it is more reasonable to assume that a deposit suddenly exposed to a high velocity surge (of a nonfouling fluid to which it is insoluble) will exhibit very fast removal of some fraction that is amenable to removal and little change thereafter. The accompanying change conforms better to a step change than to a smooth kinetic event. Finally, it is emphasized that the above approach is purely empirical, and a closer look at the removal process warrants consideration. Such a closer look must necessarily focus on the structure and properties of the deposit and their complex interaction with the growth process and the hydrodynamic field. In this way a physically sound description of the removal process will emerge and will provide the basis for successful modeling and prediction.

The following discussion aspires to be a first step in this direction by introducing a simple view of the deposit as a collection of roughness elements. The formations that are proposed to be identified with the roughness elements may be colloidal particle aggregates or polycrystalline agglomerates. In the following section the proposed model and the key assumptions are exposed, followed by the introduction of the main parameters at the single roughness element level and the

mathematical description of the collective behavior by the techniques of population balances. Subsequently, the predicted evolution and the long-term characteristics of the deposit are discussed for simple external conditions (i.e., constant hydrodynamic conditions and deposition rate). Finally, the predictions of the model are tested against experimental evidence. It is noted that experimental studies carried out in this laboratory on scaling due to supersaturated solutions of heavy metal sulfides (Andritsos and Karabelas, 1991a,b) and of calcium carbonate (Andritsos and Karabelas, 1992) have provided information helpful in shaping some aspects of the proposed model.

Development of Population Balance-Type of Model

General description and key assumptions

In these introductory considerations a simple view of the deposit is adopted by idealizing it as a collection of noninteracting roughness elements distributed on the substrate. Each one of them is growing in size as a result of the deposition process, while at the same time, it is amenable to breakage and partial or complete removal under the action of the forces exerted by the flowing fluid. Thus, it is proposed that removal does not necessarily proceed particle by particle exclusively, as assumed in the model by Cleaver and Yates (1973), but also by breakage of larger formations, such as particle clusters, branches, dendrites, and so on, which are subject to an increasing hydrodynamic load as they grow. The roughness elements are assumed to behave in a random manner, as if isolated from each other, but it is also assumed that their population is large enough so that statistical rules for their behavior exist. A measure of their size, such as mass, volume, or a linear dimension is taken to be the characteristic statistical parameter. The total mass of the deposit is considered to be equal to the sum of roughness element masses. Finally, it is assumed that the number of roughness elements per unit area is fixed.

In order to obtain a complete picture that can be put in mathematical form, it is first necessary to concentrate on the roughness element level and the problem of growth and breakage. Regarding the growth rate various assumptions can be made. For example, an isolated roughness element in contact with the flowing solution or suspension can be considered and the growth rate expressed in terms of the mass-transfer characteristics of such a system, provided that mass transfer is controlling the growth. Otherwise the surface incorporation problem can be considered from the same point of view. Statistical rules can be also prescribed for the growth, since it appears reasonable to argue that no two roughness elements, equal in size at some instance in time, will have the same fate even in the absence of removal. However, since the focus here is on the removal aspects of the fouling problem, a simpler view of the growth phase will be adopted. The roughness elements will be assumed to have the same mass growth rate, independently of their size, so that this growth rate multiplied by their number per unit area equals the macroscopic deposition rate.

The other key issue is roughness element breakage and removal. As a first step it is considered pertinent to assume that breakage occurs when the intrinsic stress induced by the flowing fluid exceeds a critical value. The loading history as well as the dynamic nature of the loading will be ignored at this point.

Two questions arise immediately. First, how to find the stress field in a roughness element, as well as the critical value that it can withstand, and secondly how to obtain a kinetic expression for the macroscopic removal process out of this apparently static picture which is proposed as the basic ingredient of deposit removal.

The problem of the intrinsic stress field is certainly complicated. At this point, however, some reasonable order-of-magnitude estimates based on assumptions about the shape of the roughness elements and a simple external loading condition would suffice. Regarding the macroscopic kinetic behavior, the following view is proposed. A population of roughness elements of a given size exhibits a distribution of resistances to breakage which depend on their size and therefore evolve as the size changes due to growth. If that change favors breakage it will be possible for removal to occur and the rate of this process will be directly related to the rate of growth or the deposition rate. In other words changes are not possible under conditions of no deposition unless the hydrodynamic conditions change. Of course, one may argue that removal could be possible under prolonged exposure to a force field due to deposit fatigue or similar phenomena but such considerations are beyond the scope of this article.

Some comments are now in order about the spatial distribution of the roughness elements. As already stated, they are considered to be isolated from each other and their number to remain fixed. It would perhaps be more realistic to assume that the sum of the masses of the roughness elements is not equal to the total deposit mass but just a fraction of it, since they might develop on a more or less inactive part of a tenacious deposit. However, such a view is not implemented here. The assumption of a constant roughness element number density is also not well founded. Yet, it may be argued that during the later stages of fouling there is no particular reason for a systematic variation in the number of the roughness elements, and these stages are probably described more closely by the proposed model. The same argument may also be used to justify the proposition that the mean distance between roughness elements is of the same order of magnitude as some characteristic linear dimension. In this way an estimate of the number density can be obtained.

Population balance

The view proposed here can be described mathematically by the techniques of population balances which have been applied to various similar problems such as particle agglomeration, crystal growth, droplet breakage, and so on. For a comprehensive review see Ramkrishna (1985). Let therefore $P(y, t)$ represent the probability density of the size distribution of a sufficiently large sample of roughness elements. Here y may represent any characteristic quantity related to the size of the roughness elements such as mass, volume, or a linear dimension. The dependence on t reflects the fact that the size distribution evolves in time. Let also the rate at which roughness elements of size y break into smaller pieces be represented by F . Let $P_x(x)$ represent the probability density of the fragment size distribution where x is the fraction of a roughness element that remains intact after breakage. Then, the equation governing the evolution of the size distribution is given by:

$$\frac{\partial P}{\partial t} + \frac{\partial PG}{\partial y} = -PF + \int_y^\infty P(y')FP_x(y/y')dy'/y' \quad (3)$$

Here G is the growth rate of a roughness element which may be a function of its size only. If y represents mass then G is constant as discussed before. The LHS of Eq. 3 expresses the change of the population due to growth. The first term on the RHS is the rate at which roughness elements of size y break into smaller pieces. The last term represents the rate at which roughness elements, larger than y , break into pieces such that the remaining fragment is of size y . With the help of Eq. 3 one can find the evolution of the mean mass of the roughness elements which is directly related to the total deposited mass. If the mass of a single roughness element is denoted by m , one has:

$$\begin{aligned} \frac{d\langle m \rangle}{dt} &= \frac{d}{dt} \int_0^\infty Pm dy = \int_0^\infty \left(\frac{\partial Pm}{\partial t} + \frac{\partial PGm}{\partial y} \right) dy \\ &= \int_0^\infty P \frac{dm}{dt} dy - \int_0^\infty PFm dy \\ &\quad + \int_0^\infty m dy \int_y^\infty PFP_x dm'/m' \end{aligned}$$

and after some manipulation:

$$\frac{d\langle m \rangle}{dt} = \langle G_m \rangle - \int_0^\infty PmF(1-x_m)dm. \quad (4)$$

Here $\langle G_m \rangle$ is the mean growth rate and

$$\langle 1-x_m \rangle \equiv \int_0^1 P_x(1-x)dx$$

is the mean detached mass fraction of the roughness elements suffering breakage. Multiplying by the number of roughness elements per unit area, N_0 , we obtain an expression for the total deposited mass:

$$\frac{dm_t}{dt} = N_0 \langle G_m \rangle - N_0 \int_0^\infty PmF(1-x_m)dm. \quad (5)$$

The term $N_0 \langle G_m \rangle$ represents the macroscopic deposition rate, J_D , as has been stated already, while the second term in Eq. 5 corresponds to deposit removal. It can be observed that the assumption, made in other models (that is, Kern and Seaton, 1959), of a removal rate that is proportional to the deposit mass is not strictly supported by the above formulation unless the mean removed fraction, $\langle 1-x_m \rangle$, and the breakage rate, F , are independent of the size of the roughness elements.

Intrinsic stress field and breakage rate

As has been stated already, a roughness element suffers breakage when the intrinsic stress imposed by the flow field exceeds a critical value which is taken to be time-independent and spatially random inside each roughness element. Some geometrical considerations are now in order. It is assumed that the roughness elements grow in such a manner that they maintain a self-similar shape which can be characterized by a single

parameter, that is, their height, h . The shape is taken to be a pyramid with square cross-section and side that decreases with distance from the base. Then the volume is given by:

$$V = \int_0^h S(z') dz' = a_B^2 h \int_0^1 (1-z')^2 dz' = \frac{1}{3} a_B^2 h, \quad (6)$$

where, for simplicity, we have taken $S(z) = a_B^2(1-z)^2$ so that the side of the cross-section decreases linearly. Here z is a dimensionless distance from the roughness element base. The side of the base, a_B , may also depend on the height in some prescribed manner.

As has already been stated, the rate of change of the mass of each roughness element due to deposition is assumed independent of size. Then, the same will be true for the volume. It is further assumed that the base side is related to height by:

$$a_B = \gamma h^{n/2} \quad (7)$$

where γ is a constant. Then since dV/dt is constant and equal to the mass growth rate divided by deposit density, using Eqs. 6 and 7 one obtains:

$$\frac{dh^{n+1}}{dt} = 3J_D/\rho N_0 \gamma^2 \quad (8)$$

This can be used to obtain an expression for the growth rate, $G(h)$, in the population balance, if the height, h , is chosen as the size variable. It is noted here that for $n < 2$ the height increases faster than the base side, that is, if $n=1$ then $h \propto t^{1/2}$, $a_B \propto t^{1/4}$, and in general $h \propto t^{1/(1+n)}$, $a_B \propto t^{n/(2n+2)}$.

We now turn to the problem of estimating the nominal intrinsic stress of a roughness element by considering a segment cut by a plane parallel to the base. The force exerted on such a segment by the flowing fluid is estimated as the product of the hydrodynamic wall shear stress multiplied by the area of that segment that is exposed to the flow. This approximation is considered appropriate for roughness elements immersed in the viscous sublayer of the turbulent flow. It is noted parenthetically here that such an estimate gives reasonable agreement when applied to the case of a sphere resting on a plane and exposed to a slow linear shear flow, where an analytical result is available (O'Neill, 1968). The force, calculated in this way, is now equated to the product of the cross-sectional area, which is the base of the segment, times the nominal intrinsic stress, τ_{IN} , at the same position. The cross-sectional area is $S(z) = a_B^2(1-z)^2$ and the area exposed to the flow is $S_e(z) = 2(h^2 + a_B^2/4)^{1/2} a_B(1-z)^2$. Therefore:

$$\tau_{IN} = \tau_w 2(h^2 + a_B^2/4)^{1/2} / a_B = \tau_w \frac{2}{\gamma} (h^{2-n} + \gamma^2/4)^{1/2} \quad (9)$$

and it can be observed that the nominal intrinsic stress, found according to the above arguments, is independent of position, which simplifies the calculations. It must be also noted that for $0 \leq n < 2$ the intrinsic stress increases as the roughness element height increases. Then removal is possible, while for $n=2$, where all linear dimensions grow at the same rate, this is not possible since the intrinsic stress does not change.

Finally, the kinetic aspects of the removal process can be

considered. It is assumed that for each roughness element there exists a strength limiting plane whose position is random. Then, the fraction of a population of roughness elements of a certain size that suffers breakage at a certain level of external loading is:

$$f = \int_0^1 dz \int_0^{\tau_{IN}} P(\tau) d\tau = \int_0^{\tau_{IN}} P(\tau) d\tau \quad (10)$$

Here $P(\tau)$ is the critical stress probability density which is assumed independent of position, roughness element size, and previous history. Now, as the size of the roughness elements increases so do the intrinsic stress and the fraction that suffers breakage. Then, if at a certain instance in time, t , $(1-f)$ roughness elements exist out of an initial population, at time $t + \Delta t$ their size will increase by Δy and the broken fraction by Δf . Therefore the fraction of those existing at time t that is broken within Δt is $\Delta f/(1-f)$, and the rate of removal is $[(df/dy)/(1-f)] (dy/dt)$ or

$$F = \frac{df/dy}{1-f} \frac{dy}{dt} = g(y)G(y) \quad (11)$$

where

$$g(y) \equiv \frac{df/dy}{1-f} \quad (12)$$

Before concluding this section an important point is emphasized. As has been mentioned already, according to the model proposed here it is not possible to have any changes in the absence of deposition. This can be seen in Eq. 11 where the breakage rate, F , is proportional to the growth rate.

Predicted Behavior

Steady-state distribution

The fragment size probability density, P_s , is uniform when expressed in terms of the roughness element height because the nominal intrinsic stress is spatially uniform. Therefore, it is convenient to use the roughness element height as the key size variable. Then the population balance becomes:

$$\frac{\partial P}{\partial t} + \frac{\partial PG}{\partial h} = -PgG + \int_h^\infty PgGdh'/h' \quad (13)$$

We now turn to the behavior that can be predicted by the picture set in the previous section, by first exploring the existence of a steady state that may be established after an infinite time. Setting the time derivative equal to zero in Eq. 13 and differentiating once with respect to h one can eliminate the integral term to obtain:

$$\frac{d^2 P_\infty G}{dh^2} + \frac{dP_\infty gG}{dh} + \frac{P_\infty gG}{h} = 0 \quad (14)$$

This can be readily integrated to give:

$$P_{\infty} = \frac{ch}{G} \exp \left[- \int_0^h g(h') dh' \right] \quad (15)$$

Furthermore, since $g = -d[\log(1-f)]/dh$, as can be seen from Eq. 12, the above expression becomes:

$$P_{\infty} = \frac{ch}{G} (1-f). \quad (16)$$

Here c is a normalizing constant given by:

$$c = \left[\int_0^{\infty} \frac{h}{G} (1-f) dh \right]^{-1}. \quad (17)$$

The integral above must be finite and this provides a condition on f for the existence of steady state.

In order to proceed further the critical stress probability distribution must be known, although some of the conclusions drawn are independent of the particular form of the distribution. Obviously, such a matter is not trivial. However, significant information on the possible forms of the critical stress distribution can be found in the related fracture mechanics literature (cf. Herrmann, 1990). A log-normal distribution is tentatively used here. This is characterized by two parameters, namely the mean stress, τ_M , and a standard deviation, $\ln \sigma$,

$$P(\tau) = \frac{1}{\sqrt{2\pi} \ln \sigma} \exp \left[- \frac{\ln^2(\tau/\tau_M)}{2 \ln^2 \sigma} \right] \frac{1}{\tau} \quad (18)$$

Then, from Eq. 10 the fraction suffering breakage at a certain external stress is:

$$f = \frac{1}{2} \left\{ 1 + \operatorname{erf} \left[\frac{\ln(\tau/\tau_M)}{\sqrt{2} \ln \sigma} \right] \right\}. \quad (19)$$

This can be recast in terms of the roughness element height, h , by use of Eq. 9. Assuming, now, that the mean intrinsic strength of the roughness elements is much larger than the hydrodynamic wall shear stress one can simplify Eq. 9 to:

$$\tau_{IN} = \tau_w \frac{2}{\gamma} h^{(2-n)/2} \quad (20)$$

This suggests a way to nondimensionalize the height, h , by defining:

$$h_0 = (\gamma \tau_M / 2 \tau_w)^{2/(2-n)} \quad \text{and} \quad \bar{h} = h/h_0. \quad (21)$$

Then, Eq. 19 can be written as:

$$f = \frac{1}{2} [1 + \operatorname{erf}(\beta \ln \bar{h})] \quad (22)$$

where

$$\beta = \frac{1 - (n/2)}{\sqrt{2} \ln \sigma} \quad (23)$$

Finally, since the growth rate, G , is proportional to h^{-n} , the steady-state distribution can be rewritten:

$$P_{\infty} = \frac{\bar{h}^{n+1} \operatorname{erfc}(\beta \ln \bar{h})}{\int_0^{\infty} \bar{h}^{n+1} \operatorname{erfc}(\beta \ln \bar{h}) d\bar{h}}. \quad (24)$$

The mean height can now be found from:

$$\langle \bar{h} \rangle = \int_0^{\infty} \bar{h} P_{\infty} d\bar{h},$$

and performing the integration one finds:

$$\langle \bar{h} \rangle = \frac{n+2}{n+3} \exp[(2n+5)/4\beta^2]. \quad (25)$$

As has been mentioned before, this holds provided $0 \leq n < 2$.

Similarly, the mean base side of the roughness elements can be found as:

$$\langle a_B \rangle = 2\gamma h_0^{n/2} \frac{n+2}{3n+4} \exp[(8n^2+5n)/16\beta^2] \quad (26)$$

and the mean mass of the roughness elements:

$$\langle m \rangle = \frac{1}{3} \rho \gamma^2 h_0^{(1+n)} \frac{n+2}{2n+3} \exp[(3n^2+8n+5)/4\beta^2] \quad (27)$$

Finally, as discussed before, we introduce the assumption that the mean distance between roughness elements exhibits the same behavior as their base side. In this way we are able to obtain an expression for the total deposited mass per unit area. We have $N_0 = 1/\langle a_B \rangle^2$, and hence:

$$m_t = \frac{1}{12} \rho h_0 \frac{(3n+4)^2}{(2n+3)(n+2)} \exp[(7n^2+24n+20)/16\beta^2] \quad (28)$$

Two observations can be made on the basis of the above result. The first is that the asymptotic value of the deposited mass exhibits a dependence on the hydrodynamic wall shear stress of the form:

$$m_t \propto \tau_w^{-2/(2-n)} \quad (29)$$

as can be found by substituting the definition (Eq. 21) for h_0 into Eq. 28. Thus, in qualitative agreement with experimental data the deposit mass decreases as the hydrodynamic wall shear stress increases. On the other hand the existing models postulate an exponent between $-1/2$ and -1 (Kern and Seaton, 1959; Taborek et al., 1972; Muller-Steinhagen et al., 1988). The second observation is that the deposition rate does not affect the asymptotic value of the deposited mass. The deposition rate affects only the time-scale by accelerating or decelerating the same sequence of events. Of course, one cannot preclude effects of the deposition rate on the structural characteristics and the strength of the roughness elements. However, such considerations are beyond the scope of this article.

Temporal evolution

The time-dependent problem can be put in dimensionless form by writing:

$$\bar{g} = \frac{2\beta}{\sqrt{\pi}} \frac{\exp[-\ln \bar{h} - \beta^2 (\ln \bar{h})^2]}{\operatorname{erfc}(\beta \ln \bar{h})}, \quad (30)$$

(cf. Eqs. 12 and 19), replacing G by \bar{h}^{-n} , and defining the time-scale as:

$$t_c = \frac{\rho \gamma^2 N_0 h_0^{(1+n)}}{3J_D} \quad (31)$$

When mass transfer controls the deposition process then $J_D \propto \tau_w^{1/2}$, as can be deduced from the well-known expressions such as the Linton-Sherwood correlation (cf. Hasson et al., 1978). It is pointed out that in typical cases of crystallization fouling (Andritsos and Karabelas, 1991a,b, 1992) convective diffusion to the deposit surface is found to be the controlling step and this correlation is appropriate. It is also appropriate for colloidal particle deposition (cf. Papavergos and Hedley, 1984). Using this result and $N_0 \propto \langle a_B \rangle^{-2}$, one obtains:

$$t_c \propto \tau_w^{-[3/2 + n/(2-n)]}. \quad (32)$$

On the other hand, if the deposition rate is not controlled by mass transfer and J_D is independent of τ_w then:

$$t_c \propto \tau_w^{-[1 + n/(2-n)]} \quad (33)$$

Again, in qualitative agreement with experimental evidence the proposed model predicts that the time-scale decreases as the hydrodynamic wall shear stress increases although the dependence is stronger than assumed by existing models (cf. Eq. 2). We note here, however, that if one adopts a phenomenological approach of the form of Eqs. 1 and 2, given that $m_i \propto \tau_w^{-1}$ (as is often the case) and $J_D \propto \tau_w^{1/2}$, the time constant has to exhibit a $\tau_w^{3/2}$ dependence for consistency. This is what Eqs. 29 and 32 predict for $n=0$.

In view of the above observation the parameter n is set equal to zero in the subsequent discussion. Then, the population balance takes the form:

$$\frac{\partial P}{\partial t} + \frac{\partial P}{\partial h} = -Pg + \int_h^\infty Pg dh' / h' \quad (34)$$

where bars have been dropped for convenience. The above equation was solved numerically using finite-difference approximations for the spatial and temporal derivatives and the trapezoidal rule for the calculation of the integral term in the RHS. Numerical diffusion errors were eliminated by use of equal space and time increments. A delta function was used as an initial condition, which implies that all the roughness elements start with zero size.

In Figure 1 the height distribution is shown for several characteristic times. As expected, the peak which represents intact roughness elements decreases in size to become zero eventually, leaving behind a tail of smaller fragments which have the same fate until a steady state is reached. In Figure 2 the distribution

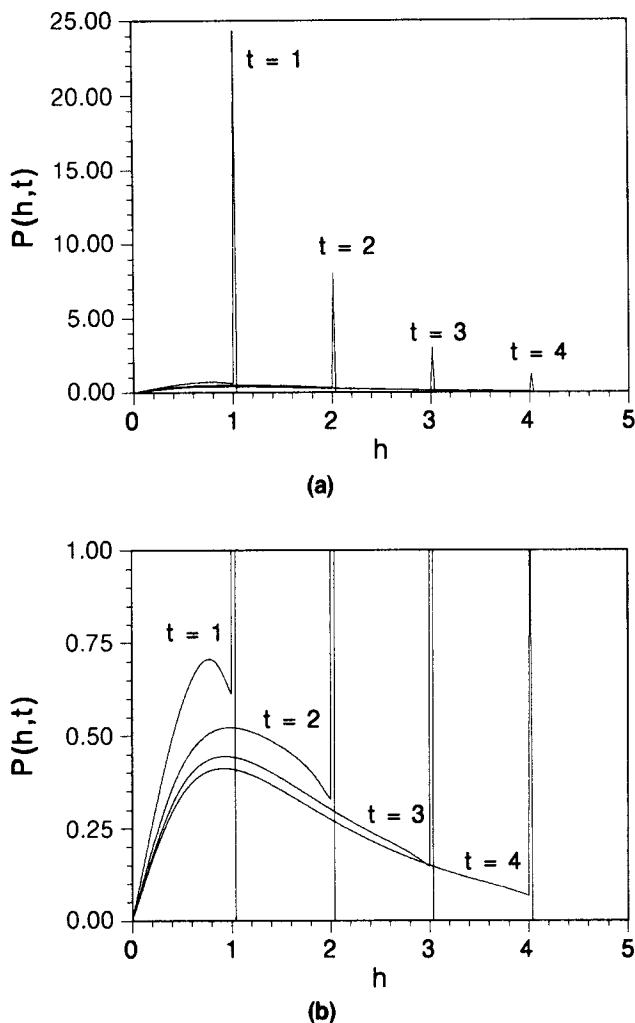


Figure 1. Evolution of the roughness element size distribution.

(a) The peak representing intact roughness elements shown for characteristic times; (b) detail of the tail left behind which represents roughness elements that suffered breakage; ($n=0$, $\beta=1$).

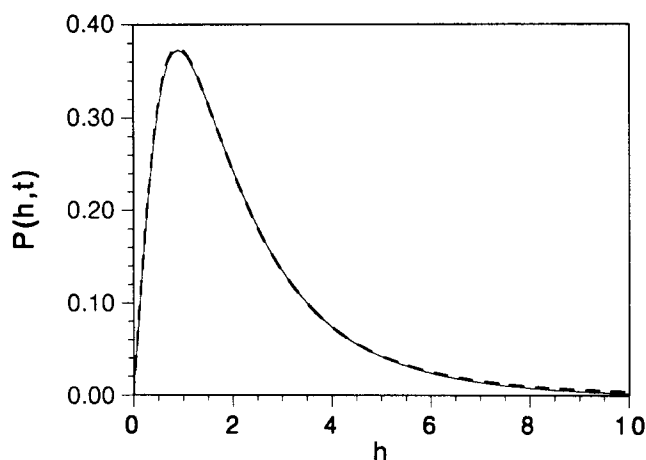


Figure 2. The steady-state roughness element size distribution.

—: numerical result at $t=20$ for $n=0$, $\beta=1$; - - -: analytical result (cf. Eq. 24).

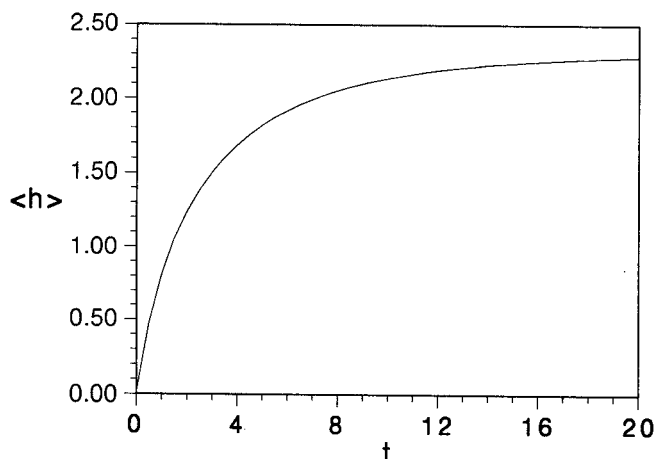


Figure 3. The mean height of the roughness elements vs. time for $n=0, \beta=1$.

The asymptotic value is 2.32; note that $\langle h \rangle$ is also proportional to the deposit mass.

at relatively large times is shown and it compares favorably with the steady-state distribution given analytically by Eq. 24. Of more interest from a practical standpoint is the first moment which is the mean height and is also proportional to the mean mass of the roughness elements and to the total deposited mass per unit area. This is shown in Figure 3 where after an initial period of almost linear growth, which corresponds to negligible removal, the growth rate slows down and eventually the steady state is reached. The shape of the curve is qualitatively similar to experimental data exhibiting asymptotic fouling. This is also shown in Figure 4 for various values of β which is the only parameter that appears explicitly in the formulation and represents the spread in breakage strengths. Apart from the strong influence on the steady-state value this parameter has also a strong effect on the time-scale of the problem. Now, if the curves shown in Figure 4 could be described by the simple model of Eqs. 1 and 2 then a plot of $(\langle h \rangle_f / t) \log(1 - \langle h \rangle / \langle h \rangle_f)$ vs. t would be a constant. As can be seen from Figure 5, the model proposed here exhibits a different trend, especially during the initial stages of the evolution process. However, all the curves approach the same value, from which it is concluded

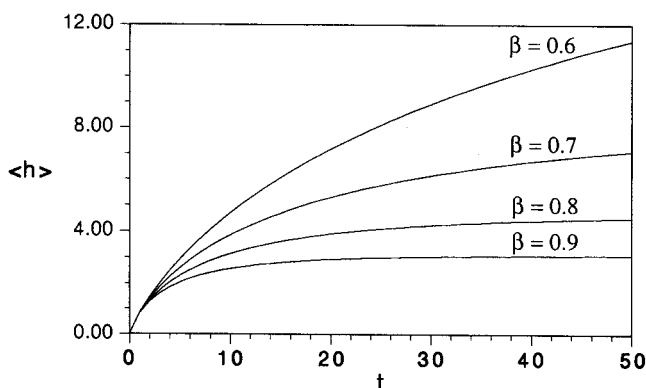


Figure 4. The mean height of the roughness elements for $n=0$ and characteristic values of β which represent the spread in breakage strengths.

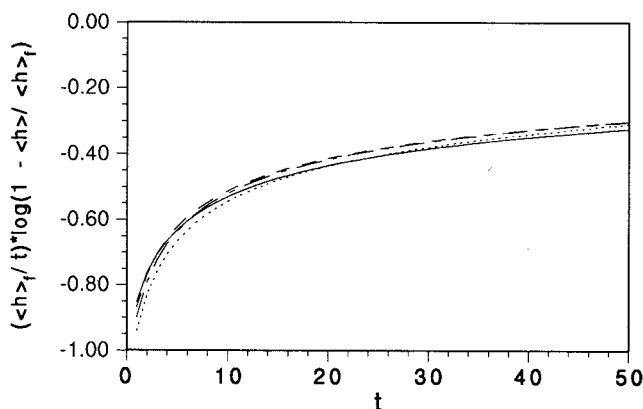


Figure 5. The data of Figure 4 plotted in a different form.

If the evolution of $\langle h \rangle$ could be described by a simple exponential these data would fall on straight horizontal lines. —, $\beta=0.6$; ---, $\beta=0.7$; — — —, $\beta=0.8$; ····, $\beta=0.9$.

that the time-scale exhibits the same dependence on β as the asymptotic mass or:

$$t_c \propto \exp(5/4\beta^2) = \exp(5 \ln^2 \sigma / 2). \quad (35)$$

Comparison with Experimental Evidence

At this point it is considered premature to perform detailed comparisons of the proposed model with experimental data. The main scope of the preceding discussion is to propose a rational mechanism that, at least partially, accounts for the removal aspects of fouling. However, some key features of the model need to be emphasized in order to demonstrate that predictions are not unreasonable.

The main emphasis of the analysis has been on the effect of the hydrodynamic parameters on the asymptotic value of the deposit mass and on the time-scale of the fouling process. As has been stated already, the predicted trends are in agreement with experimental evidence suggesting that an increase in velocity results in a smaller asymptotic mass or fouling resistance which is reached more rapidly (cf. Epstein, 1983).

Unfortunately, appropriate experimental data sets where velocity is systematically varied while other parameters, conditions and deposit characteristics remain constant have not been identified in the literature. Knudsen and coworkers have performed extensive experimental studies of heat exchanger fouling but most data sets have been obtained in a limited velocity range. Lee and Knudsen (1979) report a fourfold increase in fouling resistance by a 50% velocity reduction, which is in accord with Eq. 29. More detailed studies of the effect of velocity in heat exchanger crystallization fouling with CaCO_3 can be found in Watkinson and Martinez (1975) and also in Hasson (1962). However, their data have been obtained under constant wall temperature conditions, which implies that deposit buildup resulted in reduction of the temperature at the deposit/fluid interface and a consequent reduction of the deposition rate. Yet, there are several reasons suggesting that this reduction cannot fully account for the falling character of the fouling curves and that removal is present and important. In Hasson's run B, where sufficient details about the deposit surface temperature are available, calculations of the deposition rate based on the model by Hasson et al. (1978) show

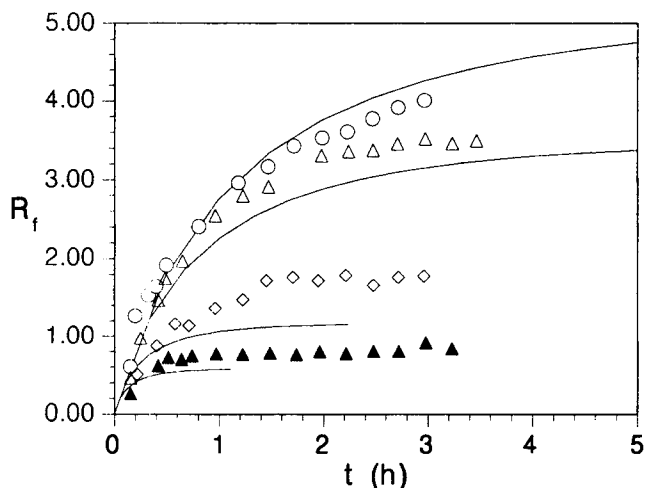


Figure 6. Comparison of experimental data on CaCO_3 fouling by Watkinson and Martinez (1975) with the present model.

The fouling resistance R_f is in $10^{-4} \text{ h ft}^2 \text{ }^\circ\text{R/Btu}$; \circ , $v=0.8 \text{ m/s}$; \triangle , $v=1 \text{ m/s}$; \diamond , $v=1.87 \text{ m/s}$; \blacktriangle , $v=2.79 \text{ m/s}$. The solid lines are numerical results from the current model. The asymptotic fouling resistance was adjusted for the lowest velocity and calculated for the other cases.

that it can be reduced by a factor of about four during the entire course of the run. This is not sufficient to account for the observed total reduction factor which is more than 20. Furthermore, if removal were totally absent runs at higher velocities would correspond to higher deposition rates and hence higher fouling resistance. For the same reason, no velocity effect should be observed in Watkinson and Martinez since surface reaction is claimed to control the deposition. Yet,

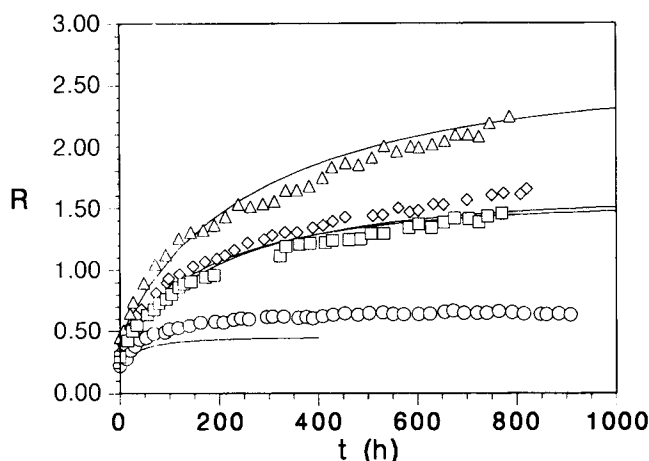


Figure 7. Comparison of experimental data on CaCO_3 fouling by Hasson (1962) with the present model.

The overall heat-transfer resistance R is in $10^{-3} \text{ h m}^2 \text{ }^\circ\text{K/kcal}$; \triangle , run A at final velocity $v=39 \text{ cm/s}$ and tube diameter $d=16 \text{ mm}$; \diamond , run B at $v=50 \text{ cm/s}$ and $d=16 \text{ mm}$; \square , run C at $v=42 \text{ cm/s}$ and $d=16 \text{ mm}$; \circ , run D at $v=125 \text{ cm/s}$ and $d=8 \text{ mm}$. The solid lines are numerical results from the present model. The asymptotic fouling resistance was adjusted for the lowest velocity and calculated for the other cases.

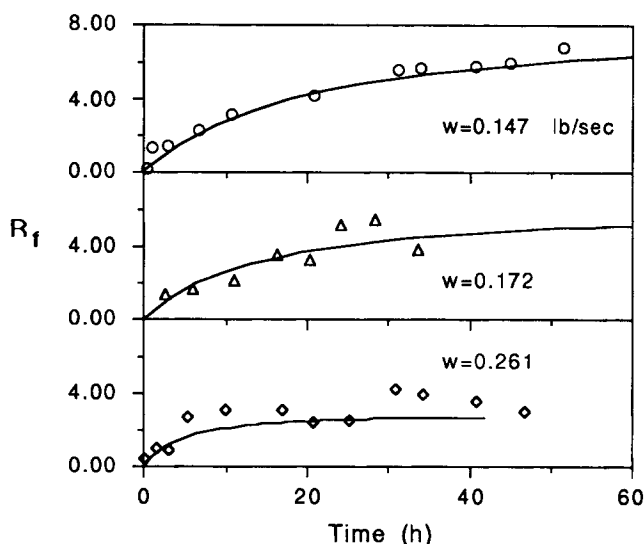


Figure 8. Comparison of experimental data on particulate fouling from a sand-water slurry of Watkinson and Epstein (1970) with the present model.

The fouling resistance R_f is in $10^{-5} \text{ h ft}^2 \text{ }^\circ\text{R/Btu}$; the asymptotic fouling resistance was adjusted for the lowest velocity and calculated for the other cases.

in both their experiments higher velocity corresponds to lower fouling resistance.

Despite the above obvious shortcomings, a comparison of data from those studies with the present model is considered instructive and is presented in Figures 6 and 7. The asymptotic fouling resistance, assumed proportional to deposit mass, was adjusted for the lowest velocity and calculated for the other cases by use of Eq. 29. A mean value for the deposition rate was estimated from the available data and used to find the time-scale by combining Eqs. 28 and 31. Thus, for $n=0$, one finds:

$$t_c = \frac{3}{2} \exp(-5/4\beta^2) m_i / J_D \quad (36)$$

This obviates the need to specify values for the geometric and material parameters of the model except for β , which was taken to be unity. The same value for the deposition rate was employed for all cases in Figure 6 since surface reaction was claimed to be the deposition rate controlling step in the experiments of Watkinson and Martinez.

The same procedure was also applied with the experimental data on particulate fouling by Watkinson and Epstein (1970). Again, the asymptotic fouling resistance was adjusted for the lowest velocity and used to calculate the value for the other cases. The values for the deposition rates supplied in that paper were used to find the time-scales. As can be seen from Figures 6, 7 and 8, the behavior of the present model can be considered quite reasonable and encouraging. However, a more detailed evaluation of the predictive capabilities of the model in quantitative terms would require comparisons with complete data sets including accurate measurements of deposit properties.

Another important point that deserves to be discussed is that, according to the present model, the deposition rate does

not affect the sequence of fouling events and in particular the final deposit mass, but merely controls the time-scale. In contrast, previous models predict a direct effect of the deposition rate on the asymptotic mass, since deposition and removal are presumed independent. Apart from the hydrodynamic conditions, on which both deposition and removal depend, this effect can be manifested through the concentration difference, which is the driving force for deposition, and through temperature which controls the coefficients multiplying this driving force (cf. Taborek et al., 1972; Epstein, 1983). The prediction of the current model may appear to be an oversimplification and probably will not hold true if some kind of interaction between roughness elements is allowed or a link between deposition rate and deposit strength is proven to exist or if a different mode of removal (independent of growth) is operative. On the other hand, it must be observed that such a direct and dramatic effect of concentration has not been unambiguously and universally proven to exist either. Gudmundsson (1981) reports an effect of concentration in some cases of particulate fouling and none in other. A clear effect is evident in the experiments of Blochl and Muller-Steinhagen (1990). Muller-Steinhagen et al. (1986) also observe a linearity between fouling particulate concentration and asymptotic fouling resistance although there is considerable scatter in their data. In a later study (Muller-Steinhagen et al., 1988), covering different velocity ranges, no significant effect is observed and this is attributed to the velocity ranges being irregularly extreme. The same uncertainty regarding concentration effects applies also for crystallization fouling. In support of the present model the following observation may be pointed out from the experimental data of Morse and Knudsen (1977), (run 22), Hasson (1962), (run D) and Watkinson and Martinez (1975) (run at $v = 1$ m/s); in all these cases the deposit is dominated by calcium carbonate, the hydrodynamic conditions are similar, and the time-scales and hence the deposition rates are vastly different, yet the asymptotic fouling resistances are remarkably close to the same value of approximately 20×10^{-3} h ft² °R/Btu.

Similarly, temperature affects the rate of deposition and an Arrhenius type of behavior for the asymptotic fouling resistance is proposed in the model by Taborek et al. (1972). This is apparently verified by the experimental data presented by Dunqi and Knudsen (1986), as discussed in Epstein (1987). It may be observed, however, that the chemical composition of the deposits was not the same in all cases considered, being dominated by magnesium silicate in some and by calcium carbonate in other. What we argue here is not that a temperature effect does not exist, but rather that the temperature effect on the deposit strength and structural characteristics must be first unambiguously elucidated and separated.

Concluding Remarks

A new approach to the problem of fouling has been presented which is viewed as a preliminary step to understand the removal process by focusing on the structure and properties of the deposit layer. The deposit is modeled as a collection of roughness elements, which may represent polycrystalline agglomerates or colloidal particle aggregates. The basic ingredient of the removal process is taken to be failure of the roughness elements when they reach a size that cannot withstand the

hydrodynamic load. The macroscopic kinetic behavior is directly linked to the deposition and growth process and the polydispersity of strengths. Mathematical complexity is kept to a minimum by the introduction of simple assumptions about the roughness element geometry, growth process, the flow induced intrinsic stress field, and the breakage process. The behavior predicted agrees qualitatively with experimental trends in the evolution of macroscopic characteristics and the effects of the flow field on the removal rate and the asymptotic value of the deposit mass.

An interesting suggestion brought forth in this modeling effort is that the intrinsic deposition rate affects only the time-scale of the combined deposition/detachment process, but not the asymptotic value of the deposited mass. To what extent this statement is true is difficult to ascertain at present.

The present model may be refined in several ways. Roughness element consolidation may be taken into account in order to approach the physical picture of coherent deposits more closely. Changes in the geometrical characteristics in coordination with deposit morphology studies can also be made. The breakage behavior is another aspect of the model that can be improved. Thus, different critical stress distributions, breakage modes (that is, gross fragmentation vs. attrition) or criteria can be used, and finally time effects due to the unsteady nature of loading can be included within the presented formalism.

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Notation

- a_B = side of the base of a roughness element, m
- $\langle a_B \rangle$ = mean side of the base of the roughness elements, m
- c = proportionality constant
- f = fraction of roughness elements suffering breakage
- F = roughness element breakage rate, s⁻¹
- g = function related to roughness element growth rate, kg⁻¹, m⁻¹ or m⁻³
- G = roughness element growth rate, kg/s, m/s or m³/s
- $\langle G_m \rangle$ = mean mass growth rate of the roughness elements, kg/s
- h = height of the roughness elements, m
- $\langle h \rangle$ = mean height of the roughness elements, m
- h' = dummy variable of integration
- h_0 = length scale for nondimensionalization of height, m
- \bar{h} = dimensionless height of roughness elements
- $\langle h \rangle_f$ = mean roughness element height at steady state
- J_D = fouling species mass deposition rate, kg/m²·s
- J_R = fouling species mass removal rate, kg/m²·s
- k = proportionality constant, s⁻¹
- m = mass of a roughness element, kg
- $\langle m \rangle$ = mean mass of roughness elements, kg
- m' = dummy variable of integration
- m_t = total mass of deposit per unit area, kg/m²
- n = geometrical parameter related to the shape of the roughness elements
- N_0 = number of roughness elements per unit area, m⁻²
- $P(y, t)$ = roughness element size probability density, kg⁻¹, m⁻¹ or m⁻³
- P_∞ = roughness element size distribution at steady state
- P_x = fragment size probability density
- $P(\tau)$ = breakage stress probability density, m²/N
- S = cross-sectional area of a roughness element, m²

S_e = area of a roughness element exposed to flow, m^2
 t = time, s
 t_c = time-scale, s
 V = volume of a roughness element, m^3
 x = fraction of a roughness element adhering after breakage
 x_m = mass fraction of a roughness element adhering after breakage
 y' = dummy variable of integration
 y = roughness element size, kg, m or m^3
 z = dimensionless distance from base of a roughness element

Greek letters

β = dimensionless parameter related to spread of breakage strengths
 γ = geometrical parameter related to the size of the roughness elements
 ρ = roughness element density, kg/m^3
 τ_{IN} = intrinsic stress of a roughness element, N/m^2
 τ_M = mean breakage strength of a roughness element, N/m^2
 τ_w = hydrodynamic wall shear stress, N/m^2
 ψ = deposit strength, N/m^2

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