ENHANCEMENT OF HEAT TRANSFER IN CRYSTALLIZATION FOULING DUE TO SURFACE ROUGHNESS

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ABSTRACT
This contribution addresses apparent negative fouling resistances in the induction phase of crystallization fouling. Therefore it focuses on the correlation between surface roughness and heat transfer enhancement. Fouling experiments with a supersaturated aqueous CaSO₄ solution in a double pipe heat exchanger were carried out at a Reynolds number of 18,000 corresponding to a flow velocity of 0.65 m/s. The measured pressure drop between inlet and outlet streams allowed the calculation of the friction factor for the present surface roughness. With the given friction factor it was possible to estimate the actual heat transfer coefficient of the inner tube. Considering the change over time in heat transfer caused by surface roughness, the fouling resistance may be recalculated. Thus, the appearance of a negative fouling resistance can be explained quantitatively.

INTRODUCTION
Fouling is generally defined as the unwanted deposition or growth of suspended, dissolved or chemically generated species from process fluids onto heat transfer surfaces. These layers can lead to a drastic increase of the heat flux resistance and therefore to a decreased efficiency of the heat exchanger. In crystallization fouling typically two phases can be distinguished: Induction and crystal growth period.

In the induction period the mechanism of crystallization fouling starts with nucleation on the surface caused by local super-saturation. Fig. 1 shows a fouling curve for an aqueous CaSO₄ solution in a double pipe heat exchanger. The duration of the induction period for the common drawn stainless steel tube is about 40 hours. The length of this period depends among other parameters on the energetic characteristics of the heat transfer surface and on the salt concentration as well as on the surface topography (Förster et al., 1999). However, for this specific time no negative effect on heat transfer can be found. The heat transfer performance even may increase. One reason why the formation of deposits on a heat transfer surface can potentially reduce the overall thermal resistance is the deposit itself. The formation of crystal clusters on the surface leads to an increase of surface roughness and therefore to an improvement of the local heat transfer coefficients due to higher turbulence (Crittenden and Alderman, 1988). Heat transfer enhancement due to fouling roughness maintained for the crystal growth period. Furthermore the formation of fouling reduces the cross-sectional area of the tube. Hence, for heat exchangers at constant flow rate conditions, the velocity will increase and therefore the film heat transfer coefficient may also be increased. Whether either or both of these effects can compensate the increase in thermal resistance of the fouling layer itself is discussed in the following.

EFFECT OF SURFACE ROUGHNESS ON HEAT TRANSFER
The effect of surface roughness on heat transfer is well established and is widely used to improve the performance of many kinds of heating equipment (Pohl, 1933; Sheriff and Gumley, 1966; Feurstein and Rampf, 1969; Taylor et al., 1992; Bohnet, 1993). The quantitative effect of roughness on heat transfer depends on the nature of the roughness, i.e., the size, the shape, orientation and distribution of the roughness elements (Mahato and Shemilt, 1968). Information about the roughness of fouling deposits in literature is rather limited (Crittenden and Alderman, 1992). Generalized correlations for the roughness structure of fouling deposits are not available. The effect of roughness on flow and consequently heat transfer was quantified using the concept of an equivalent sand grain roughness by
Nikuradse (Nikuradse, 1933). Original correlations for the heat transfer in straight pipes were either of the form:

$$Nu = a \cdot Re^b \cdot Pr^c$$

(1)

where the heat transfer coefficient is embodied in the Nusselt number, which is related to the process parameters and physical properties via Reynolds and Prandtl numbers, or semi-empirical correlations, which belong on an approach of Prandtl (Prandtl, 1933):

$$\frac{\lambda}{8} = St = \frac{Nu}{Pr \cdot Re}$$

(2)

Where $\lambda$ is the resistance due to friction and $\phi$ the correction term for Prandtl number disparate to one. With time the approaches became more and more adjusted to further measurements. Nunner (Nunner, 1956) did experiments in gas flow on the heat transfer performance of rough surfaces. For this he induced circlips, varied in shape and distance, inside tubes of different diameters. He proposed a correlation for the heat transfer and friction number for rough pipes may be estimated by:

$$K^+ = Re \left( \frac{k_s}{d_i} \right) \sqrt{\frac{\lambda}{8}}$$

(8)

Based on his experiment he proposed the correlation:

$$\eta = \frac{\varepsilon_{Nu}}{\varepsilon_\lambda} = \log \frac{Pr^{0.33}}{K^{0.247}} - 0.32 \cdot 10^{-3} K^+ \log Pr + 1.25$$

(9)

With this correlation for the heat transfer and friction characteristics of smooth and rough surfaces it is possible to calculate the increase of heat transfer for rough pipes by means of pressure drop measurements. They are necessary to determine the friction factor $\lambda$ and the equivalent sand grain roughness $k_s$. Using Eqs. (5) and (9), the Nusselt number for rough pipes may be estimated by:

$$Nu = Nu_0 \left( \frac{Pr^{0.33}}{K^{0.247}} - 0.32 \cdot 10^{-3} K^+ \log Pr + 1.25 \right)$$

(10)

$Nu_0$ is the Nusselt number for smooth pipes, calculated by an approach of Petukhov (Petukhov and Popov, 1963)

$$Nu_0 = \frac{\lambda_0}{1.07 + 12.7 \sqrt{\frac{\lambda_0}{8} \left( Pr^{2/3} - 1 \right)}}$$

(11)

Another correlation for heat and mass transfer for turbulent pipe flow is given by Hughmark (Hughmark, 1975). In his work he proposed that the wall region consists of a developing laminar layer at the wall and a transitional region. Both have different characteristics. Hence, considering the two distinct regions in the vicinity of the wall, he provides a three-resistance model for the three regions of turbulent pipe flow with molecular and eddy diffusion properties:

$$\frac{Nu}{Nu_0} = \left( \frac{\lambda}{\lambda_0} \right)^{\frac{m}{2}}; \quad m = Pr^{0.5} + 1.5; \quad Pr > 1$$

$$m = Pr + 1.1; \quad Pr < 1$$

(7)

Two effects may contribute to heat transfer enhancement due to roughness: An increase in surface area relative to a smooth wall and an increase in near wall turbulence. Reevaluating Nunner's work Burck (Burck, 1969) assessed that the circlips Nunner used were not in direct contact with the heat transfer surface. Thus, the roughness elements act as a turbulence promoter only but they do not increase the surface area necessarily. In his experiment Burck used different types of roughness, one being rectangular grooves in the tube wall, cut with different shapes and distances. Furthermore he varied the Prandtl numbers in a range from 3 to 180. He found a relation between the efficiency $\eta$, the Prandtl number $Pr$ and the roughness parameter $K^+$, where the latter can be written as a function of the equivalent sand grain roughness $k_s$ and the friction factor $\lambda$:

$$K^+ = Re \left( \frac{k_s}{d_i} \right) \sqrt{\frac{\lambda}{8}}$$

(8)
Hughmark used experimental data from Dipprey and Sabersky (Dipprey and Sabersky, 1963), Gown and Smith (Gown and Smith, 1968) and Kolár (Kolár, 1965) to verify his theoretical approach also for rough surfaces. These data include a sand grain roughness and roughness formed by cutting triangular threads in the tube wall, which all represent moderate roughness \((K<100)\). The model is in good agreement with the experimental data. For high Reynolds numbers and extreme roughness he established that the experimental coefficients are much less than calculated coefficients. Ceylan (Ceylan and Kelbaliyev, 2002) proposed a correlation to estimate the effectiveness parameter in fully developed turbulent flow through rough pipes based on experimental data from Eckert and Drake (Eckert and Drake, 1972). For \(Pr<50\) and \(10^4<Re<10^7\), Ceylan gives the following empirical equation:

\[
\eta = \frac{\varepsilon Nu}{\varepsilon \lambda} = 1.15 Pr^{1/4} \left(1 - 0.106 K^{1/4} \lambda \right) \lambda_0
\]

Using Eqs. (5) and (13), the Nusselt number for rough pipes is:

\[
Nu = 1.15 Nu_0 Pr^{1/4} \left(1 - 0.106 K^{1/4} \lambda \right) \lambda_0
\]

where \(Nu_0\) is the Nusselt number for smooth pipes according to Petukhov.

**EXPERIMENTAL TECHNIQUES**

For the generation of surface roughness due to crystallization fouling, experiments were carried out. The setup of the fouling mini-plant facility is schematically shown in Fig. 2. A supersaturated aqueous solution of calcium sulphate was used as process fluid. The concentration in the bulk phase was 0.027 mol/L. The concentration of the calcium sulphate solution was controlled by titrimetric analysis every 24 hours. The bulk temperature of the process fluid at the test section inlet was 42°C. The experiments were conducted under turbulent flow conditions with an average Reynolds number of 18,000 corresponding to a flow velocity in the test section of 0.65 m/s. Before entering the actual test section, the fluid flow passes a smoothing section, where the flow gets hydrodynamic fully developed. The smoothing section has a length of 450 mm and an inside diameter of 16 mm identical to that of the test section.

In general the experimental setup can be divided into three parts. The first one includes the product vessel B1 (50L) and the heating water vessel B2 (100L). The product vessel contains the aqueous solution of calcium sulphate. A rotating stirrer equalizes temperature and concentration gradients. Hot water is provided in vessel B2 by two immersion heaters with an overall electrical power of 14 kW. The second part consists of two centrifugal pumps, a cartridge filter and a plate heat exchanger. The filter avoids sedimentation of particles and secondary nucleation in the test sections since seed particles can influence nucleation behaviour considerably. The heat exchanger guarantees a constant inlet temperature of the process fluid at the test sections. The last part contains the two double pipe heat exchangers which are the actual test sections. Both heat

![Experimental setup of the double pipe heat exchanger facility](image)
exchangers are identical in construction, whereas one of them may be used for reference measurements. The heat exchangers are heated by the hot water on the shell side. The slightly super saturated solution is fed in counter current flow through the inner tubes, which have an internal diameter of 16mm and a length of 2000mm. Besides the common drawn stainless steel tubes, electro-polished stainless steel tubes were used for the current experiments. The flow rate of each test section, on the shell side as well as on the tube side, is measured by inductivity flow meters. All temperatures are measured by thermocouples. To eliminate heat losses the whole test rig is insulated by mineral wool. The pressure loss between the inlet and outlet of each test section is measured by two pressure transducers. In addition to that the absolute pressure is recorded at the inlet of each heat exchanger. Hence, with knowing the exact flow velocity the friction factor can be calculated over the time of the experiment. All measured data are connected to a data scan unit. Considering the two double pipe heat exchangers of the present study, the heat transfer in the double pipe arrangement is calculated from:

\[ Q = U A \Delta T_m \]  

(15)

where \( U \) is the overall heat transfer coefficient, \( A \) the heat transfer area consistent with definition of \( U \) and \( \Delta T_m \), the suitable log mean temperature difference (LMTD) across the heat exchanger. In case of no heat losses the heat flow rate of the hot fluid transfers completely to the cold fluid stream. Thus, the heat flow rate may also be written as:

\[ Q = \dot{m}_h c_{p,h} (T_{h,\text{i}} - T_{h,\text{o}}) - \dot{m}_c c_{p,c} (T_{c,\text{o}} - T_{c,\text{i}}) \]  

(16)

The subscripts \( h \) and \( c \) stand for the hot or cold process stream, whereas \( i \) and \( o \) stand for the inlet respectively the outlet. By equating Eq. (15) and (16) the overall heat transfer coefficient can be described with:

\[ U = \frac{Q}{A \Delta T_m} \]  

(17)

in which the heat flow rate is determined by the cold process stream. For constant mass flow rates and constant inlet temperatures on the shell side as well as on the tube side of the heat exchanger, the decrease of the overall heat transfer caused by fouling can be measured by the change in each outlet temperature.

RESULTS

Fig. 3 shows the fouling resistance of an electro-polished tube and the pressure drop characteristics versus time. In comparison to the common stainless steel tube the mean roughness depth \( R_z \) is about 50% lower. As mentioned before the duration of the induction period depends on the kind of surface modification. Due to that the induction period has not finished during the experimental time. Furthermore, the fouling resistance shows a negative value over the whole time of the experiment apart from the very beginning. This corresponds to an increase in heat transfer due to the disturbance of the viscous sub layer by the deposit surface roughness. Also the pressure drop increases over the experimental time, which is another indication for a first crystal growth on the surface. This can also be supported by the endoscopic photographs in Fig. 4, which are taken after every experimental run. The pictures do not show a compact layer at any parts of the tube. Therefore, any reduction in the crossectional area, which may lead to an additional increase of the heat transfer coefficient (Crittenden and Alderman, 1992), can be neglected.
Observing a straight pipe with an inside diameter $d_i$, respectively an outer diameter $d_o$, and $d_f$ as the inside diameter of the fouling layer, the convective heat flow rates at the inner and outer surfaces can be written as follows:

$$\dot{Q} = h_i A_i (T_{i,b} - T_{i})$$

$$\dot{Q} = h_o A_o (T_{o,b} - T_{o,W})$$

(18)

The heat flow rates through the wall and through the fouling layer are given by:

$$\dot{Q} = \frac{2\pi k_i L}{\ln\left(\frac{d_i}{d_f}\right)} (T_{i} - T_{i,w})$$

$$\dot{Q} = \frac{2\pi k L}{\ln\left(\frac{d_o}{d_f}\right)} (T_{o,W} - T_{i,w})$$

(19)

Solving Eqs. (18) and (19) with respect to the overall temperature difference and introducing a reference area $A$, the overall heat transfer can be written as,

$$\dot{Q} = U_f A (T_{o,b} - T_{i,b})$$

(20)

with $U_f$ as the overall heat transfer coefficient of the fouled surface. In case of a very thin fouling layer, the surface area of the deposit $A_f$ is approximately the area of the clean surface $A_c$. Multiplication with the reference surface area $A$ gives,

$$\frac{1}{U_f} = \left(\frac{A}{h_i A_i} + \frac{x A}{k A_m} + \frac{A}{h_o A_o}\right) + \left(\frac{x A}{k A_i}\right)$$

(21)

where $A_m$ is the corresponding average area,

$$A_m = \frac{A_c - A_f}{\ln\left(\frac{A_o}{A_i}\right) - \ln\left(\frac{A_o}{A_i}\right)}$$

(22)

The first bracket of Eq. (21) describes the overall heat transfer resistance of the clean surface $1/U_0$ at time zero of the fouling process. Hence, the fouling resistance $R_f$ obtained from the difference between a fouled surface and a clean surface is

$$R_f = \frac{1}{U_f} - \frac{1}{U_0} = \frac{x A}{k A_i}$$

(23)

Due to the fact, that $1/U_0$ is constant $h_i$ would not change over the time of a fouling process. An increase in heat transfer due to roughness effects may override the increase in fouling resistance $R_f$ to let $1/U_f$ to be less than $1/U_0$. To consider the effect of surface roughness on heat transfer during the initial stage of fouling, Eq. (21) and Eq. (23) may be expressed as follow:

$$\frac{1}{U_f} = \frac{A}{h_i(t) A_i} + R_f(t) + \frac{1}{U_0}$$

with

$$\frac{1}{U_0} = \frac{x A}{k A_m} + \frac{A}{h_o A_o}$$

(24)

The inner heat transfer coefficient $h_i$ is a function of the occurring friction factor $\lambda$.

$$h_i(t) = \frac{\lambda(t)}{\lambda_0}; \text{ respectively } \frac{Nu}{\lambda_0} = \frac{\lambda(t)}{\lambda_0}$$

(25)

To determine the friction factor $\lambda$ for the present surface roughness, pressure drop measurements between the inlet and outlet stream were conducted. Using the measured pressure drop, volume flow and fluid temperature, the friction factor can be calculated from the Bernoulli equation:

$$\lambda = \frac{\Delta p d_i}{2 L \rho w^2} - \frac{2}{\rho w^2}$$

(26)

where $d_i$ is the inner diameter of the tube, $L$ is the corresponding tube length and $w$ is the flow velocity of the process stream.

Fig. 6  Calculated heat transfer coefficient according to different models

Similar to the pressure drop, shown in Fig. 3, the friction factor increases with the first crystal growth. The characteristics is inversely proportional to the fouling resistance, which argues that both, the heat balance and the hydrodynamic, show reliable data and a comparable reproduction of the fouling process. With the given friction factor it is possible to estimate the actual heat transfer coefficient respectively the Nusselt number of the inner tube. Fig. 6 represents the heat transfer coefficients calculated from the actual friction factor over the experimental time. As mentioned before there are several empirical approaches to calculate the heat transfer in rough channels. In the following the different approaches will be discussed and compared to each other. Considering Fig. 6, in general all heat transfer coefficients increase with increasing friction factor due to crystallization fouling.
overlaid roughness. Integral roughness, like finned tubes, is in direct contact with the heat transfer surface. Hence, it is also an increase in surface area, which leads to an additional improve in heat transfer performance. Overlaid roughness may have a similar shape as integral roughness. Therefore the resistance due to friction could be identical, whereas the effect on heat transfer is different caused by the thermal contact resistance between heat transfer surface and roughness element.

Fig. 9  Recalculated fouling resistances in comparison to the fouling resistance obtained from heat balance

Fig. 9 illustrates the recalculated fouling resistance in comparison to the fouling resistance, calculated from Eq. (23). According the previous results, the highest amount of fouling is calculated with the approaches by Burck and Ceylan. Additionally, both models start with an offset. A low fouling resistance is calculated by the empirical equation of Nunner. Between those are the values obtained by using Hughmark’s equation. Further experiments have been conducted under the same process conditions. All of them show comparable results. A first step to verify the results is to explore the effects on heat transfer which were found for the induction period also for the crystal growth period. According to the empirical approach which will be used, different asymptotic fouling resistances were calculated. Also different deposit thicknesses \( x_f \) were calculated, considering Eq. (23) and Eq. (24). These deposit thicknesses can also be determined experimentally by volumetric measurements and may be compared to the calculated layer thickness according to each empirical approach. Fig. 10 represents the recalculated values of the fouling experiment with a common drawn stainless steel tube according to Fig. 1. After an experimental time of approximately 250 h the fouling resistance ended in an asymptotic value. During the crystal growth period a compact layer is formed on the heat transfer area. Hence, the crosssectional area will be reduced, which might cause an
additional increase in heat transfer due to higher fluid velocity. Therefore, the average crosssectional area has been determined by measuring the deposit thickness volumetrically at the end of the experiment. With the given reduced tube diameter it was possible to estimate the additional change in heat transfer by using the Gnielinski equation (Heat Exchanger Design Handbook, 1986) for a first step. Fig.10 illustrates the relation of each calculated thickness with the part due to reduced cross section on top of each bar. All calculated thicknesses are below the measured thickness. While the deviation by using Hughmark’s equation is 20% the equations by Burck or Ceylan are only 10% below the measured thickness.

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<th>Heat balance</th>
<th>Roughness effect</th>
<th>Restricted cross section</th>
<th>Calculated thickness</th>
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Fig. 10 Deposit thickness

In comparison to the previous results, the lowest level of deposit thickness is estimated by using Eq. (23). Taking the surface roughness of the fouling layer into account by using the equation of Nunner the estimated height of the deposit increases by 57%. Using the empirical model of Hughmark the calculated thickness is 133% higher, whereas either using the equation of Burck or Ceylan the calculated deposit height is 164% higher. Overall the fouling resistance and therefore the deposit thickness seems to be underestimated when not accounting for the roughness effects on heat transfer in crystallization fouling. Therefore, it is required to consider both the hydrodynamics and the heat balance to describe the fouling process.

CONCLUSIONS

Fouling experiments in a double pipe heat exchanger with electro-polished tubes as well as with common drawn stainless steel tubes were conducted under constant process conditions by measuring the pressure drop and the heat balance. Based on measurements of roughness effects due to fouling on pressure drop and on convective single phase heat transfer it is found that both characteristics are almost symmetrically, supported by a high reproducibility of the results. Considering the enhancement of heat transfer due to surface roughness the approaches of Burck and Ceylan lead to the highest value of fouling resistance and already show an offset of Rf(t) ≠ 0 at the very beginning. During the time where the crystal growth has not started the empirical models of Nunner and Hughmark show equivalent characteristics of fouling compared to the resistance calculated from the heat balance. In further course of the experiments, while first crystals are formed on the heat transfer surface, also the fouling resistance starts to grow by considering the change in heat transfer due to roughness effects. Thus, the appearance of negative fouling resistances can be explained and corrected. Furthermore a different interpretation of the induction period can be suggested, which is in general defined as the point in time whereas a positive fouling resistance is encountered. Published studies often give a further fragmentation of this period in an induction and a transitional part. Considering the roughness effects the transitional period would disappear as shown schematically in Fig. 11.

Exploring the roughness effects also for the crystal growth period for a fouling experiment in a common smoothly drawn stainless steel tube it is found that the calculated deposit thickness using the approaches of Hughmark, Burck and Ceylan show good agreements to the measured deposit thickness. Overall the fouling resistance and consequently the deposit thickness are underestimated by calculating just the heat balance of Eq. (23). Therefore, the experimental results replicate the significance to consider both, the hydrodynamics and the heat balance, to describe the fouling process.

For further conclusions concerning the influence of roughness effects during the initial stage of crystallization fouling, different flow regimes will be investigated. Furthermore, to get an idea of the temperature gradient along the tube and information about local super saturation of the salt solution the heat exchanger will be calculated in an iterative way to get a differential view of the results. This work is in progress.

NOMENCLATURE

A area, \( \pi dL \), m²
a constant, dimensionless
Cp heat capacity, J kg⁻¹ K⁻¹
d diameter, m
h heat transfer coefficient, W m⁻² K⁻¹
k heat conductivity, W m⁻¹ K⁻¹
K* dimensionless roughness, dimensionless

Fig. 11 Fouling curves according to different definition of \( R_f \)


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**REFERENCES**


