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PULSED FLOW AND SURFACE COATINGS TO MITIGATE FOULING

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ABSTRACT

Crystallization fouling on heat transfer surfaces is a major problem in many fields of the process industry. Adverse effects, such as decreasing thermal efficiency, increasing pressure drop and production loss in consequence of cost intensive and persistent cleaning procedures, are to be mentioned. Crystallization fouling in heat exchangers is a superimposed process of deposition and removal. The objective of this work is to mitigate fouling by enhancing removal applying two complementary approaches:

(1) Increased fluid forces using pulsed flow

(2) Decreased adhesion forces between wall and crystals using different surface coatings with varied free surface energies.

Experimental examinations were carried out with a calcium sulfate solution in a rectangular flow channel (Re ≈ 3000 - 3600). Two exchangeable stainless steel plates were mounted on a heating element, so that different surface coatings could be examined. The coatings applied in this work were SICAN and SICON®. A pulsed flow was imposed in intervals during the heat transfer process. Different modes of pulsation were tested.

The examinations showed that applying pulsed flow to an adhering process leads to a decreased final fouling resistance. The decrease of the final fouling resistance results from increasing fluid forces. For certain parameter combinations a balance between deposition and removal occurs and leads to an asymptotic fouling resistance. The experiments using surface coatings revealed to lengthen the induction period.

INTRODUCTION

Crystallization fouling on heat transfer surfaces is a major problem in the process industry. There are several disadvantages linked to the appearance of fouling. The growth of crystals leads to an increase of the heat transfer resistance, which can be quantified by the thermal fouling resistance $R_{\rm f.}$

$$R_{f} = \frac{1}{U_{f}} - \frac{1}{U_{0}} \tag{1}$$

 U_f describes the overall thermal resistance of the fouled plate and U_0 the overall resistance of the clean plate. The fouling resistance may also be quantified using a mass based approach (Mayer et al., 2010), m_f describes the fouling

mass, λ_f thermal conductivity of the fouling layer and ρ_f the density of the fouling layer:

$$R_f \propto \frac{m_f}{\lambda_f \rho_f} \tag{2}$$

Additionally an increase of the pressure drop occurs due to roughness effects (Albert et al., 2011) and the reduction in flow area of the duct.

Both mechanisms reduce the efficiency of the whole process. Oversized heat exchangers and peripheral devices or periodic cleaning procedures are common approaches to handle this issue. This results in a reduced economic and ecological efficiency of a process.

The fouling process can be divided into two sections, the induction period and crystal growth period (Geddert, 2009). During the induction period no significant decrease of the heat transfer can be detected. During the crystal growth period an increase of the thermal resistance occurs. Both periods contain deposition and removal processes. Depending on process parameters different fouling progressions can appear:

(a) Linear progression: The ratio of deposition and removal is constant or the removal can be neglected

(b) Decreased progression: The rate of growth decreased over time, no final value will be achieved

(c) Asymptotic progression: Deposition and removal are in balance, a final value will be achieved

(d) Sawtooth progression: Removal rate is not constant, pieces of the deposit are being removed on the microscale.

FOULING MITIGATION



Fig. 1 Deposition and removal on a heat transfer surface [Geddert, 2011]

Numerous factors affect fouling and cleaning [Geddert, 2011], see Fig. 1. As the crystallization fouling process is a superposition of deposition and removal, two general mitigation approaches are conceivable:

(1) Reduction of deposition

(2) Enhancement of removal

In the following examination, the enhancement of removal shall be observed.

The fouling mass rate \mathring{m}_{f} is defined as the difference between the deposition mass rate \mathring{m}_{d} and the removal mass rate \mathring{m}_{r} . Förster (2001) specified the removal rate using the removal probability Γ which is a function of the ratio of shear force F_{τ} and adhesion force F_{ad} (Chen et al., 1995).

$$\dot{m}_f = \dot{m}_d - \dot{m}_r = \dot{m}_d \left(1 - \Gamma \left(\frac{F_\tau}{F_{ad}} \right) \right) \tag{3}$$

Referring to this equation the removal rate can be increased by increasing the acting shear force on the crystals and decreasing the adhesion force between crystal and surface.

Besides the possibility of increasing the main flow velocity to enhance the removal rate, Augustin (2003) introduced the approach of a pulsed flow to reduce the fouling tendency of calcium sulfate. The superposition of a steady base flow and an oscillating flow movement results in a pulsed flow. The experiments with pulsation showed a continuous removal of crystals, which led to a quasi-stationary state of the fouling resistance. In further investigations a non-continuous pulsation was applied where the period of pulsation was interrupted by a period of steady flow conditions. It can be seen that for short enough intervals the results are similar to the continuous pulsation (see Fig. 2).

The application of pulsed flow was also used for the enhancement of cleaning processes. Gillham et al. (2000) revealed the cleaning time of whey protein fouling to be significantly decreased by pulsed flow in comparison to a steady cleaning procedure. Blel et al. (2009a, 2009b) examined bacterial removal under pulsed flow conditions and noticed an increase of the constant removal rate in comparison with steady flow conditions.



Fig. 2 Fouling curves for different delay times, $w_{stat} = 0.25$ m/s, W = 1 [Augustin, 2003]

The possibilities of surface modification are described by several authors. Förster (1999) and Geddert (2009) showed increased induction periods by the use of energetic surface modifications (DLC, SICAN, SICON®). The main characteristic of these surfaces is the reduction of adhesion forces between crystals and surface. Combined with shear forces driven by the fluid flow the removal rate can be enhanced (Geddert, 2009). Geddert (2009) also examined the free surface energies of the used surfaces (see Fig. 3), but did not found a clear correlation between free surface energy and induction period. Zhao et al. (2005) examined the fouling behavior of a Ni-Cu-P-PTFE composite coating and the influence of free surface energy on the adhesion force. They identified a free surface energy range in which the adhesion force is minimal and concluded the potential of such a coating to reduce mineral and biofouling [Zhao et al. 2005]. Zettler et al. (2005) examined several coatings based on different coating technologies. They did not find a correlation between surface energy and fouling behavior, but it tends to reduced fouling at reduced surface energies. Gao et al. (2006) described an extension of the induction period and enhanced anti-fouling properties by the use of a Ni-based implanted tube at boiling conditions. Al-Janabi et al. (2011) examined solvent based, water based and electroless Ni-P-BN coatings whereby the solvent based and Ni-P-BN coatings showed a significantly decreased final fouling resistance.



Fig. 3 Surface energy of the different coatings on untreated stainless steel [Geddert, 2009]

Based on these observations the two fouling mitigation strategies were examined. In the presented work both approaches are investigated separately to attain a deeper understanding of the main parameters influencing the fouling process. In future work both approaches will be combined applying the most effective parameters of the present work.

EXPERIMENTAL SETUP

The experiments were carried out with a plate heat exchanger, see Fig. 4. The different surfaces were electrically heated and the temperature was measured by two thermocouples to calculate the thermal fouling resistance. To eliminate the influence of smaller fluctuations of the electrical power on the fouling resistance, the electrical power at each time step was used for the calculation of $R_{\rm f}$. For parallel testing, two test surfaces could be mounted on

the heating element, one on each side. This led to the possibility of examining different surface modifications and their influence on the fouling process. The outer body of the flow channel contained two transparent sections made of PMMA to observe the fouling process visually.

Coming from a storage tank, 150 L of aqueous calcium sulfate solution was pumped in circle. After the centrifugal pump, the solution passed through a heat exchanger, which enabled a constant inlet temperature. Finally, it passed through the investigated test section. A detailed description of the test rig can be found in (Förster, 1999).



Fig. 4 Test section [Förster, 1999]

For pulsating experiments, the plant was equipped with a pulsator. The pulsator consisted of a piston pump with the suction side shut.

A pulsed flow consists of a steady flow (generated by the centrifugal pump) which is superimposed by an oscillating flow movement (generated by the pulsator). The resulting ideal velocity function is shown in Fig. 5. If the maximum oscillating flow velocity is greater than the steady flow, a temporary flow reversal occurs. The characteristic parameter is the dimensionless waviness W:



Fig. 5 Stationary and oscillating component of pulsating flow [Augustin, 2003]

In order to leave the energy input in range of steady flow conditions, the experiments shown here were carried out with temporary pulsation instead of permanent pulsation. This means that a period of pulsation is interrupted by a period of steady flow conditions. Different parameters were varied (see Fig. 6):

(i) The waviness, which describes the ratio of the maximum oscillating velocity $w_{os,max}$ and the steady velocity w_{stat} , was adjusted between 0.5 and 1.5, (ii) the pulsation interval Δt_p , which is defined as the sum of one pulsation time plus one steady interval, was varied between 10 s and 1000 s and (iii) the number of strokes were varied between 2 and 8.



Fig. 6 Schematic diagram of the main pulsation parameters

The experiments were carried out with a standard stainless steel plate. The concentration of calcium sulfate was 24 mmol/L and the fluid velocity was set to 0.12 m/s. The experiments with surface coatings were carried out at steady flow conditions ($w_{stat} = 0.1 \text{ m/s}$) and a calcium sulfate concentration of 27 mmol/L. The surface coatings applied in this work were SICAN and SICON®. SICAN is a diamond like carbon (DLC) surface where silicon is built into the a-C:H matrix, SICON® includes the combination of silicon and oxygen inside the DLC layer. The thickness of the coatings is 3 µm and did not change the original roughness of the stainless steel plate. The coatings were developed at the Fraunhofer Institute for Surface Engineering and Thin Films in Braunschweig, Germany.

Tab.	1:	Overview	of	the	test	parameters
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Parameter	Range
Steady flow velocity w _{stat}	0.1 - 0.12 [m/s]
Max. oscillating flow velocity w _{os,max}	0 - 0.15 [m/s]
Waviness W	0 - 1.5 [-]
Pulsation interval Δt_p	10 - 1000 [s]
Number of strokes t _p	2 - 8 [-]
Pulsation frequency	1.7 [Hz]
Reynolds number	3000 - 3600 [-]
Concentration CaSO ₄	24 - 27 [mmol/L]

RESULTS AND DISCUSSION

The following results show a parameter study with a variation of the waviness, the number of strokes and the pulsation intervals.

Influence of the pulsation interval

Experiments were carried out with $\Delta t_p = 10$ s, 100 s, and 1000 s with a number of strokes of $t_p = 2$. As a reference, the fouling curve of a constant flow velocity at w = 0.12 m/s is

presented. The comparison of the pulsation intervals shows no significant differences during the first 12 h. After that, an increase of the fouling resistance in each case occurs. All fouling curves at pulsed flow conditions show a more or less pronounced sawtooth progression where crystal growth is followed by a sudden removal. Generally it can be seen that with a decreasing pulsation interval the final fouling resistance is also decreased (Fig. 7). If the interval is long $(\Delta t_p = 1000 \text{ s})$ the pulsation has no influence on the fouling behavior and the final fouling resistance is approximately equal to the steady experiment. The fouling curve at $\Delta t_p =$ 10 s reached a constant value and is a 65 % reduction of the steady case. Considering the induction period, it can be seen that pulsed flow does not lead to an increase of the induction period, even a more frequent pulsation reduces the induction time.



Fig. 7 Fouling curve at constant number of strokes $t_p = 2$ and a constant waviness of W = 1.5, variation of pulsation interval Δt_p

Influence of the number of strokes

Experiments were carried out with $t_p = 2$, 4, 8 at constant $\Delta t_p = 100$ s respectively 1000 s. The fouling curves presented in Fig. 8 are examinations at $\Delta t_p = 100$ s. In general the tendency is similar to Fig. 7. Increased pulsation leads to a decrease of the final fouling resistance, which is significant in each case, and to shorter induction periods. Furthermore the sawtooth developing can be seen.



Fig. 8 Fouling curve at constant pulsation interval Δt_p =100 s and a constant waviness of W = 1.5, variation of number of strokes t_p

To further reduce the energy input, the pulsation interval was increased by a factor of ten. The fouling curves in Fig. 9 show experiments at $\Delta t_p = 1000$ s. It can be seen that the effect of pulsation is much smaller. The difference in the final fouling resistance is insignificant between the steady case and $t_p = 2$ and $t_p = 4$, respectively. Only at $t_p = 8$ a decline occurs.

In conclusion, the pulsation interval for present parameters appears to be too long to achieve a positive effect on the fouling behavior. This is in good agreement with results described by Augustin et al. (2003).



Fig. 9 Fouling curves at constant pulsation interval Δt_p =1000 s and a constant waviness of W = 1.5, variation of number of strokes t_p

Influence of waviness

The final pulsation parameter that was examined was the waviness W. The comparison includes the waviness W = 1.5, which leads to a temporary flow reversal based on equation (4) and the waviness W = 0.5 where no back flow is occurring. The resulting fouling curves, see Fig. 10, reveal that both wavinesses lead to a decrease of the final fouling resistance in comparison to the steady case. Furthermore, it is apparent that the final fouling resistance between both pulsation parameters does not differ much, but the sawtooth developing is more pronounced for W = 1.5.



Fig. 10 Fouling curve at constant number of strokes $t_p = 2$ and a constant pulsation interval $\Delta t_p = 100$ s, variation of waviness W

Fouling layer properties

As a result of the unsteady flow movement it could be determined that bigger parts of the forming crystal deposit were removed, which can be seen by the sawtooth progression of the fouling curves. Fig. 11 shows a picture of the fouling layer after the test run at steady flow conditions. The layer is uniform and adheres completely onto the heat transfer surface. In contrast, Fig. 12 shows the final fouling layer grown under pulsed flow conditions.



Fig. 11 Final fouling layer at steady flow of w = 0.12 m/s, test duration 40.6 h



Fig. 12 Final fouling layer at waviness W = 1.5, $\Delta t_p = 100$ s, $t_p = 2$, test duration 42.3 h

During the pulsation period, parts of the forming crystal deposit were removed completely or peeled off partly. In the gap between the wall and the partly peeled off layer a new fouling layer grew, which results in a layer composition. It can be hypothesized that an optimized flow configuration can lead to a complete removal of these layers.



Fig. 13 Correlation of the final fouling resistance and final mass of the fouling layer

For a further examination of these phenomena, the mass of the fouling layer was measured at the end of each experiment and compared to the final fouling resistance. Fig. 13 illustrates the correlation between both values. A linear fit seems to be a reasonable correlation for the results under pulsed flow conditions, which is in good agreement with equation (2).

The required deposited mass to reach a specific fouling resistance is less than under pulsed flow condition, due to the denser layer structure.

Influence of surface coatings

The following section illustrates the influence of different surface coatings on the fouling behavior under steady flow conditions. Fouling experiments were carried out at accelerated conditions to obtain results in relatively short time exposures due to laboratory restrictions. The concentration of calcium sulfate was increased to 27 mmol/L and the fluid velocity was decreased to 0.1 m/s. Fig. 14 illustrates the fouling curves of SICAN and SICON®. Both runs are still in the induction period after 50 h. The experiment with SICAN was continued until 350 h. After that time the measured fouling resistance was still close to zero and no visible crystals were present. The extended induction period can be explained by the low adhesive characteristic of the coatings combined with acting fluid force (Geddert, 2009). The slight increase of the fouling resistance can be explained by the deposition of crystal layers starting from a small gap at the beginning and end of the test plate. These layers covered a small part of the plate and also influenced the fluid flow.

In current investigations, the experimental conditions are being modified in order to achieve fouling in a reasonable time frame. Based on this, the combination of both fouling mitigation approaches can be examined.



Fig. 14 Fouling curve of SICAN and SICON® at steady flow conditions

CONCLUSIONS

In this study the influence of pulsed flow and surface modifications on the fouling process was examined. If the pulsation time in comparison to the steady flow velocity period is long enough, the application of a temporary pulsation leads to a significant reduction of the final fouling resistance in comparison to the steady case. The induction period could not be increased by the application of pulsed flow, even a more frequent pulsation reduces the induction period. This phenomenon is not fully understood and is currently being investigated in more detailed. The experiments using surface coatings remain in the induction period during the whole experimental time.

Currently, investigations are underway to examine the combination of both approaches. Especially if the process parameters are in a critical range so that fouling occurs on surface coatings, the combination of both seems to be a promising combination to a further reduction of fouling in heat exchangers.

NOMENCLATURE

- F_{ad} adhesion force, N
- F_{τ} shear force, N
- R_f thermal fouling resistance, m² K W⁻¹
- Re Reynolds number, dimensionless
- U_0 overall thermal resistance of the clean plate, W m⁻² K⁻¹
- U_f overall thermal resistance of the fouled plate, W m⁻² K⁻¹
- W waviness, dimensionless
- m_d deposited mass, kg m⁻²
- \mathring{m}_d deposition mass rate, kg m⁻² s⁻¹
- m_f fouling mass, kg m⁻²
- \mathring{m}_{f} fouling mass rate, kg m⁻² s⁻¹
- m_r removed mass, kg m⁻²
- \mathring{m}_{r} removal mass rate, kg m⁻² s⁻¹
- t_p number of strokes, dimensionless
- $w_{os,max}$ max. oscillating velocity, m s⁻¹
- w mean/ steady velocity, m s^{-1}
- Γ removal probability, dimensionless
- Δt_p pulsation interval, s
- λ_f thermal conductivity of the fouling layer, W m⁻¹K⁻²
- $\rho_{\rm f}$ density of the fouling layer, kg m⁻³

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