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BIOFILM FORMATION ON POLYMERIC HEAT TRANSFER SURFACES

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ABSTRACT

The biofouling affinity on different polymeric surfaces (PP, PSU, PET, PEEK) in comparison with SS 1.4301 was studied using the model bacterium *E.coli* K12 DSM 498. A test rig with seven identical chambers allowed investigations at different system conditions (throughputs, inclination of the heat transfer cell, etc.). The biofilm mass deposited on the polymer surfaces was several magnitudes smaller compared to stainless steel. The cell counts on the polymer surfaces had an opposing trend compared to the deposited biomass. The promising low biofilm formation on the polymers was attributed to the combination of their surface properties (roughness and surface free energy) when compared to SS.

INTRODUCTION

River water is frequently used for cooling in industrial processes. The resulting moderate temperatures in parts of tubing, heat exchangers and cooling towers enhance biofouling. Especially in developing and third-world countries insufficient waste water treatment and warm climate conditions lead to an increased microbiological load in streaming water. The formed biofilms affect technical surfaces through inherent metabolic processes, reduce heat transfer and increase the required pump capacity by increasing friction and decreasing the tube diameter. Heat exchanger oversizing, heat losses and increased maintenance costs are common fouling related costs (Steinhagen et al., 1993).

Several studies related to biofouling can be found in literature (Dreszer et al., 2014, Teodósio et al., 2011 and Teughels et al., 2006) but data in respect to polymeric based heat exchangers are scarce in respect to their biofouling affinity.

The choice of polymeric films is strongly connected to bacterial adhesion properties and the ability to withstand corrosion induced by inherent metabolic processes in the biofilm matrix. An investigation of biofilm formation is given for different surfaces varying in composition, roughness and free surface energy.

EXPERIMENTAL

Surface Characterization

In order to understand the processes taking place at the bacterial-substrate interface, two essential parameters of the used substrates were analyzed as are topology and free surface energy. The roughness profiles and respective roughness parameters were carried out according to DIN EN ISO 4287 via tactile scanning method. As decisive roughness parameters, the mean arithmetic roughness *Ra* and the root mean square roughness *Rq* were used.

$$Ra = \frac{1}{l}\int_0^l |z(x)|\,dx \tag{1}$$

$$Rq = \sqrt{\frac{1}{l}\int_0^l z^2(x)dx} \tag{2}$$

In eq. 1 and eq. 2 l is the single measuring length and $z(x)$ the ordinate. From a statistical point of view *Rq* is more representative as *Ra* is insensitive against extreme maxima and minima.

The characterization of the free surface energy was performed via measurement of the static contact angle of a single sessile drop on the respective surfaces. The resulting static contact angle θ at the interface depends on the thermodynamic equilibrium of the participating phases of solid, liquid and surrounding gas phase (Young, 1805; Zisman, 1963) according to:

$$\cos\theta = \frac{\gamma_{SG} - \gamma_{SL}}{\gamma_{LG}} \tag{3}$$

The free surface energy of the liquid γ_{LG} was measured via pendant drop method. The free surface energy between the solid and gas γ_{SG} was then calculated with a substitution of the interfacial energy between the solid and liquid phase γ_{SL}. There are two potential models for this substitution namely the OWRK-model (eq. 4) developed by Owens and Wendt (1969), Rabel (1971) and Kaelble (1970), which was used in the present work, as well as the method developed by Wu (1971). A comparison of both models can be found in literature (Dreiser and Bart, 2012a).

$$(1+\cos\theta)\cdot\gamma_{LG} = 2\cdot(\sqrt{\gamma_{SG}^{p}\cdot\gamma_{LG}^{p}} + \sqrt{\gamma_{SG}^{d}\cdot\gamma_{LG}^{d}}) \qquad (4)$$

In order to solve eq. 4, contact angle measurements of at least two different liquids have to be conducted.

Screening Apparatus

A screening apparatus (see Fig. 1) was developed to analyze biofouling and record the deposited biomass over time. The test rig consists of 7 parallel channels in order to have a broad reliable database in a minimum of time. Due to its flexible angle adjustment (from horizontal to vertical) it allows to simulate sedimentation effects. The channel has an inner length of 12 cm, a width of 3 cm and a channel height of 1 cm, which resembles typical distances between plates in plate heat exchangers.

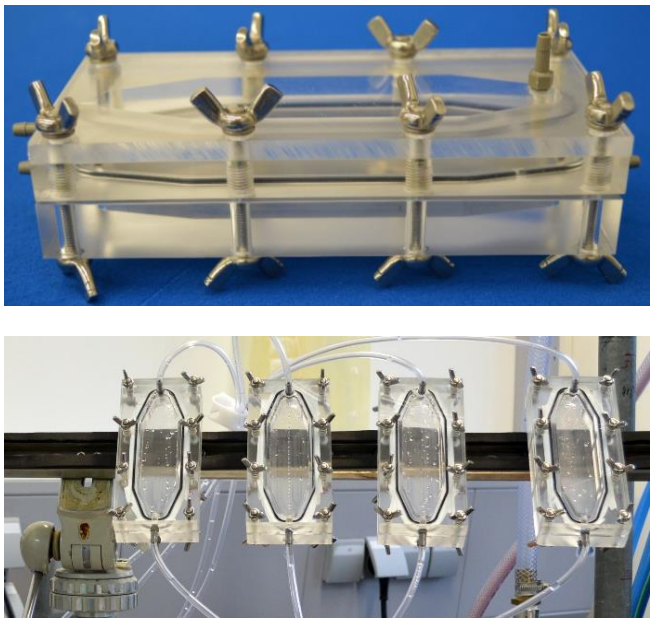

Fig. 1: Side view of a single flow channel (top) and test rig for flexible angle adjustment (bottom)

The different heat transfer surfaces act as separating layer between biological feed and heat transfer fluid (see Fig. 2). A weir guarantees an even liquid distribution and its position and size has been optimized by CFD calculations.

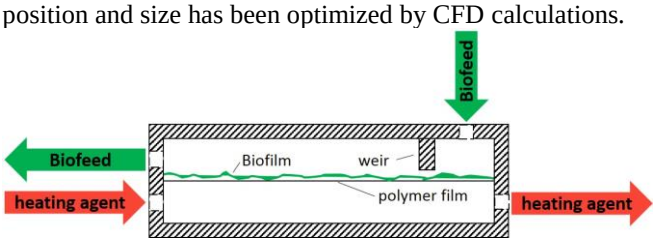

Fig. 2: Schematic composition of the flow channel

The developed flow channels allow fast *in situ* analyses of the developing biofilms on different surfaces including stainless steel (SS). The developing biofilms can be examined with epifluorescence microscopy (Manz et al., 1999) (see Fig. 3) and 3D confocal Raman microscopy by removing the flow channel cover or by utilizing the transparent cover as window for microscopy.

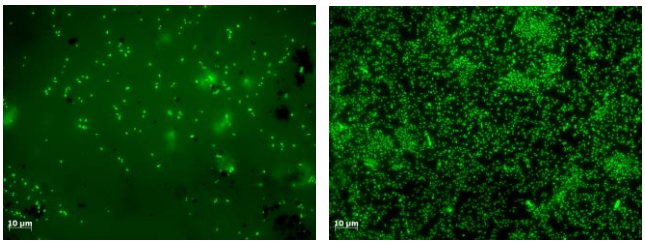

Fig. 3: Epifluorescence images of *E.coli* K12 DSM 498 on PEEK (l.) and PSU (r.) after three days exposure with 100 mL/min nutrient solution OD = 0.6 (with 600 nm), dye: SYBR Green I

These methods allow not only results for the biofilm composition at a specific time but also for the biofilm thickness at any time. Therefore the investigation of the same biofilm over a longer period of time is possible as the experiments neither have to be interrupted nor stopped.

Materials and Preparation

The biofouling affinity of several polymers was compared to SS 1.4301, which is frequently used in technical systems. Prior to testing all films were cleaned with distilled water. Two different polyether ether ketones (PEEK) from Victrex Europa GmbH, one of them containing an additional mineral filler, two different polysulfones (PSU), untreated (ut) and corona-treated (ct), polypropylene (PP) and polyethylene terephthalate (PET) from Dr. D. Müller GmbH were used. These polymers were found to be durable for desalination conditions (Christmann et al., 2012; Dreiser and Bart, 2012b) and were therefore further analyzed in concern to their biofouling characteristics.

E.coli K12 DSM 498 was used as model bacterium due to its safe and comparatively easy handling. The usage of a defined test bacterium allows therefore the reproducibility of the experiments, which is not the case when using native river water to monitor biofouling.

The bacteria were cultivated with a LB-Miller nutrient solution. The composition of 1 L LB-Miller nutrient solution is herein 10 g/L tryptone, 10 g/L NaCl and 5 g/L yeast extract. The preparatory bacterial culture was grown in an undiluted nutrient solution. This preparatory culture was incubated overnight in a heated shaking bath at 30 °C and 120 rpm in order to increase bacterial growth. The next morning 50 mL of this overnight culture was filled in 250 mL of 1:5 diluted nutrient solution (Miller). The optical density, OD, was then constantly monitored with a photometer (Lambda Bio+, PerkinElmer) and an excitation wavelength of 600 nm, which is the recommended wavelength when analyzing the OD of bacterial suspensions.

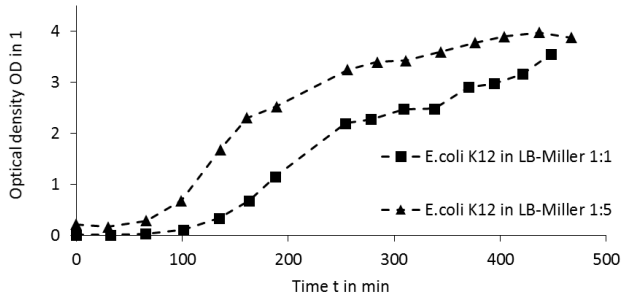

Fig. 4: Growth curve of *E.coli* K12 DSM 498 in LB-Miller nutrient solution 1:1 and 1:5

Fig. 4 shows that the dilution of the nutrient solution results in delayed bacterial growth. After the OD reached 0.6, the bacterial culture was filled into 1.5 L of 1:5 diluted nutrient solution. This batch was subsequently used as feed for the eight flow channels and operated over three days at 20°C. The flow-rate was set to 100 mL/min for each channel, which correlates to about *Re* = 105 and is therefore located in a laminar flow regime with approximately 0.005 m/s.

RESULTS

Quantity

In order to correlate the different surface properties to their general biofouling affinity, the biofilms were weighed with a microbalance before and after the experiments. The water absorption of the different polymers was assessed in preliminary experiments over the testing duration and was found to be negligible. Fig. 5 shows the accumulated biomass, including the respective standard deviation, over three days on the different surfaces.

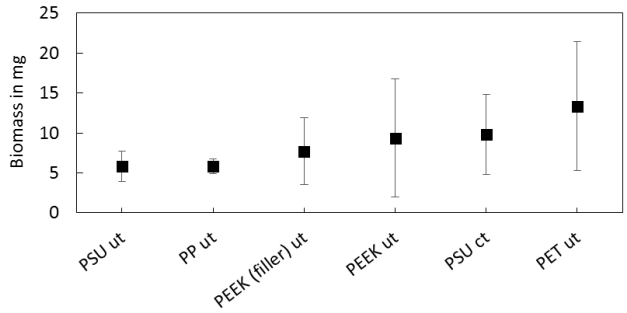

Fig. 5: Accumulated biomass over three days for different polymeric substrates

Fig. 5 shows that the accumulated biomass on the polymeric substrates is in comparatively scale. There are nevertheless differences among the polymers to be seen. PET shows the highest biomass whilst PP and PSU the lowest accumulated biomass after three days. The biomass on SS was 153±97 mg and therefore exceeded the amount of biomass on the polymeric substrates by far and is not depicted here for improved clarity.

Bacterial cell count

The bacterial cell counts were enumerated by epifluorescence microscopy after being stained with SYBR Green I as depicted in Fig. 3. The investigated biofilm area was in the middle of the flow channels in order to reduce the effects of the flow at the inlet, outlet and wall region. Fig. 7 shows the mean cell counts of the investigated area together with the accumulated biomass.

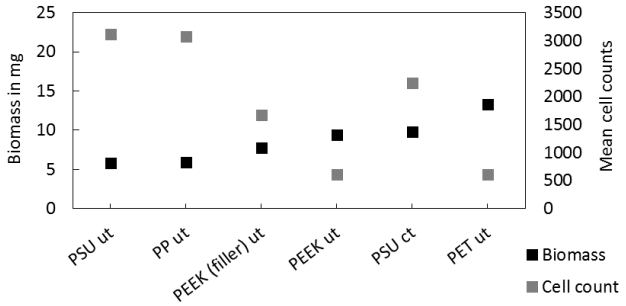

Fig. 6: Mean cell count and accumulated biomass for different polymeric substrates

The cell counts in Fig. 6 are inversely proportional to the biomass deposition, implying that a high amount of cells results in minimal production of extracellular polymeric substances (EPS). EPS are produced by the bacteria as soon as they adhere on a substrate which forms then the matrix they are imbedded in. The EPS protects the bacteria from temperature, pH and shear force variations. In contrast, a high production of EPS is based on minimal cell counts. The exception is the corona-treated PSU, where this clear opposing trend was not observed.

The effect of different EPS formation concerning the biofilm structure during the induction period was observed (see Fig. 7). The surface was not completely covered with biofilm, so the system is assumed to be dominated by roughness as the additional roughness on the surface by the bacteria increases local turbulence in the wall region but is not yet limiting heat transfer due to a relevant fouling layer. Depending on the bacteria distribution on the substrates an increased or decreased production of EPS could be optically observed.

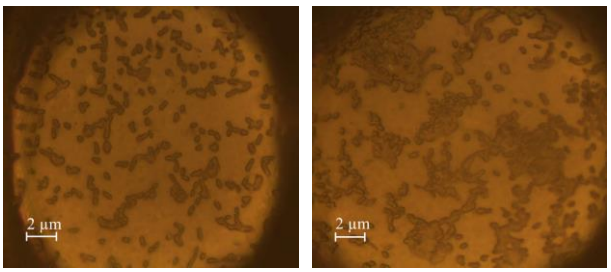

Fig. 7: Comparison of transmitted light images of PET ut (left) and PSU ut (right) after three days

As can be seen in Fig. 7, the cell growth on PET ut was punctual whilst the cells on PSU ut were primarily located in

microcolonies. How this effect can be explained due to the different surface characteristic, will be investigated in future research. A possible explanation could be the so called Quorum Sensing effect. The basic idea is that due to communication processes between the single bacteria the behavior of the biofilm itself changes as a function of the cell density. The images were taken from samples of the same batch feed.

Surface properties

Table 1 shows the measured roughness parameters of the investigated surfaces via tactile scanning method and surface free energies via contact angle measurements. Water, diiodomethane and ethylene glycol were chosen as liquids in order to calculate surface free energies according to eq. 4.

Table 1: Roughness parameters *Ra* and *Rq* and surface free energy γ_{SG} for the investigated polymers and SS (in streaming direction).

	***Ra*, [µm]**	***Rq*, [µm]**	**γ_{SG} [mN m^{-1}]**
PSU ut	0.027	0.043	45.3
PSU ct	0.031	0.039	44.4
PP ut	0.04	0.05	28.4
PEEK (filler) ut	0.852	1.073	38.5
PEEK (amorphous) ut	0.016	0.021	45.1
PET ut	0.147	0.157	45.4
SS 1.4301	0.157	0.2	33.0

PEEK (filler) includes about 30% mineral filler in order to enhance wettability. This mineral filler is reflected in the biggest roughness parameters of all investigated surfaces whilst the surface free energy is reduced compared to the amorphous PEEK. The surface of SS 1.4301 has about the roughness of the investigated PET although the biomass on SS exceeds the biomass accumulation on the polymer films by far. Water, diiodomethane and ethylene glycol were chosen as liquids in order to calculate surface free energies according to eq. 4.

The acquired surface parameters, roughness and surface free energy, seem to be linked to the deposited biofilm mass. The combination of high roughness and high surface free energy must be present, as only one high value does not seem to have a big impact on the fouling on polymers. PET for example has a comparatively high roughness and high surface free energy resulting in exceeded biofilm mass. PEEK ut on the other hand has comparable surface free energy but is much smoother, resulting in less biofouling. The surface of PEEK (filler) ut is rougher but in combination with its lower surface free energy, the deposited biomass is minimal. The reason for the exceeding biomass formation on SS 1.4301 could currently not be explained by the findings of the surface properties. Overall, the effect of total surface free energy on the accumulated biomass on polymers during the induction period seems to be more dominant compared to the roughness.

The correlations obtained for the model bacterium *E.coli* K12 DSM 498 claim further validation with more complex native bacterial biofilms (such as present in river water), which are more relevant for industrial heat exchanger applications.

CONCLUSIONS

The investigations of biological fouling on polymeric heat transfer surfaces lead to the following conclusions:

1. The studied polymers show significant advantages in quantity of the deposited biomass compared to SS 1.4301.
2. The impacts of roughness and surface free energy can be used for the development of novel surfaces for polymer film heat exchangers.
3. Accumulated biomass and cell counts are inversely proportional, resulting in e.g. increased biomass with less cells due to an increased EPS production.
4. For the validation of biofouling affinity on the investigated surfaces, further studies with more complex bacterial biofilms (e.g. from river water) are necessary.
5. Long-term studies with *in situ* biofilm thickness monitoring are requested in order to validate the findings with industry-oriented results over longer periods.
6. The developed standardized flow channel/test rig guarantees comparable results in future investigations on biofouling affinity on technical surfaces.

ACKNOWLEDGMENTS

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NOMENCLATURE

CFD	Computational fluid dynamics
ct	corona treated
EPS	extracellular polymeric substances
l	single measuring length, m
OD	optical density, -
PEEK	polyether ether ketone
PET	polyethylene terephthalate
PP	polypropylene
PSU	polysulfone
Re	Reynolds number, -
rpm	rotations per minute, min^{-1}
SS	stainless steel
ut	untreated
z(x)	ordinate, -

Greek Symbols

γ_{ij}	interfacial energy between phases I and j, mN m^{-1}
θ	static contact angle, °

Subscripts and Indices

d	disperse part
G	gas phase
L	liquid phase
p	polar part
S	solid phase

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