

## MITIGATION OF CRYSTALLIZATION FOULING IN MICROSTRUCTURED HEAT EXCHANGERS USING ULTRASOUND

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### ABSTRACT

The use of ultrasound is an opportunity to extend the operation time of microstructured heat exchangers or to remove unwanted deposition inside the microstructures. The aim of this work is the usage of ultrasound for mitigation and cleaning of crystalline depositions inside microstructured heat exchanger in a (typical) multi-layer built-up. Two different configurations, the direct and indirect coupling ultrasound into the microstructures were investigated. The experimental investigations showed that using ultrasound in microstructured heat exchangers for crystalline deposits might be a promising method to remove fouling deposits. Here, the best results were obtained for cleaning with the indirect ultrasound method.

### INTRODUCTION

The sensibility of microstructured devices regarding deposits and contamination in the microstructure is a major drawback for the industrial application of such equipment. Contamination and deposition during the operation lead to a significant deterioration of the efficiency of microstructured heat exchanger. The main advantages of microstructure devices in terms of heat and mass transfer are undone.

In recent years, basic studies were carried out to investigate the deposition behaviour of microstructured devices. In particular, crystallization fouling (Benzinger et al., 2007, Mayer et al., 2012, Bucko et al., 2012) and particle fouling (Perry and Kandlikar, 2006, Kockmann et al., 2005, Heinzl et al., 2007) were intensively studied. These investigations show that the deposits and sediments have a significant impact on the thermal and hydrodynamic behaviour and that the efficiency of the microstructured devices decreases dramatically. In order to improve the acceptance and application of microstructured devices in the industry, solutions must be developed to minimize the deposition behaviour or methods, which enable the cleaning of the microstructures. Especially, the cleaning of microstructured devices with hundreds of micro channels for industrial applications is of great importance. Here, standard methods can be applied which are also used for conventional devices, i.e. antifouling coatings, pulsating flows, flow reversal, cleaning agents, CIP (cleaning in place), etc. (Müller-Steinhagen, 2010). One additional

cleaning option is the use of ultrasound. Especially, in small sized devices (e.g. microstructured heat exchanger) the use of this method should be advantageous as microstructured devices suit well for the coupling of ultrasound. In the case of conventional heat exchangers this technology is still limited due to the large dimensions of the devices. However, some ultrasound applications for cleaning conventional heat exchangers and other apparatuses are published in literature (Ashley, 1974, Crawford, 1968, Kieser et al., 2011, Legay et al., 2013).

In Kieser et al. (2013) and Crawford (1968) the ultrasonic cleaning is based on cavitation effects. The generation of ultrasonic cavitation leads to contractions and expansions of cavities and bubble nuclei (Neppiras, 1968). Cleaning effects can be achieved mainly by shock waves and imploding liquid jets. Frequently, the effectiveness of ultrasonic cleaning may be increased if a cleaning fluid is added, so that mechanical and chemical cleaning can be expected. This kind of cleaning results in stresses between the fluid and the deposits, agitation and dispersion of contaminant by the cleaning fluid and improvement of the cleaning reactions kinetics (Cheeke and David, 2012, Ensminger and Bond, 2012).

In Ashley (1974) and Legay et al. (2013) the cleaning effects are based on vibrations. The heat exchangers and the ultrasonic transducer are directly connected and can be seen as a common vibration system (sonotrode). The vibration leads to stresses between the interfaces of heat transfer surface/deposits.

In this paper, the direct and indirect sonification was compared. For indirect sonification the ultrasound was coupled in the liquid. For the main cleaning effect based on stresses between deposits and fluid, cavitation effects cannot be excluded. By direct sonification the microstructured heat exchanger and the ultrasonic transducer were in direct contact. The cleaning effect can be described by the development of stresses between the heat transfer surface and unwanted deposits.

### ULTRASOUND CLEANING IN MICRO-STRUCTURED HEAT EXCHANGER

First attempts of using ultrasound cleaning in microstructured heat exchanger without cleaning fluids were carried out by Benzinger et al. (2005). In Figure 1 the experimental device for the direct sonification is shown. The ultrasonic transducer and microstructure built a sonotrode, vibrating the system. The vibrations leads to a removal of the deposits on the heat transfer surface. The cleaning effect can be described by the development of stresses between heat transfer surface and deposits. The microstructure with deposits and after the ultrasonic cleaning is shown in Figure 2. The experimental results show that the deposits lead to a decrease in outlet fluid temperature (Figure 3). The deposits are removed by ultrasonic sound and the flowing fluid discharged the deposit from the microstructure. Following this treatment, the thermal and hydrodynamic properties of the heat exchanger were restored.

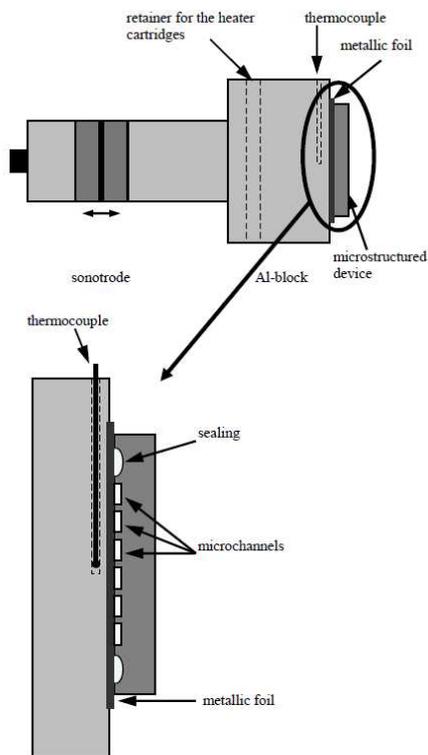


Fig. 1 Experimental microstructured heat exchangers with direct sonification (Benzinger et al., 2005)

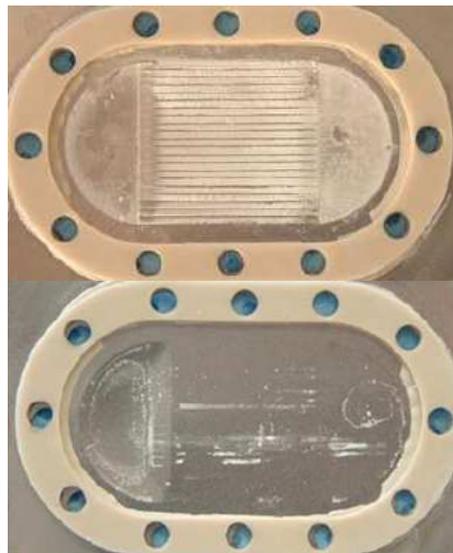


Fig. 2 Microstructure with deposits (top) and after ultrasonic cleaning (bottom).

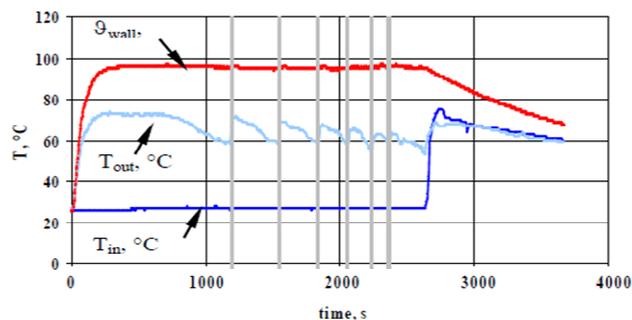


Fig. 3 Experimental results of the direct sonification.

### EXPERIMENTAL INVESTIGATIONS

#### Microstructured Heat Exchanger

For the investigations, a microstructured heat exchanger in a typical multi-layer built-up in a cross flow configuration was used. The dimensions of the microstructured heat exchanger are given in Table 1. The two investigated configurations for coupling the ultrasound waves in the microstructured heat exchanger are shown schematically in Figure 4.

For the direct method the sonotrode is directly connected to the housing of the microstructured heat exchanger. The heat exchanger with the connected sonotrode has a geometrical length which corresponds to a wavelength of approximately  $\lambda/2$ . By this, the kinetic energy of the oscillating system reached largest value. The incrustations due to fouling are exposed to stronger forces. To avoid energy dissipation of the ultrasound waves in the whole experimental setup, the entire unit (microstructured heat exchanger and sonotrode) is decoupled from the rest of the setup which is achieved using flexible pipelines. The main-cleaning effect will be obtained by the vibrations of the heat transfer surfaces, similar to Benzinger et al. (2005). The vibration should remove the crystalline incrustations.

For the indirect method the sonotrode is placed in the front of the microstructure. The ultrasonic waves are coupled into the fluid flowing into the microstructured section of the heat exchanger where the fouling occurs. Due to the propagation of the ultrasonic waves in the fluid, areas are building up in which the fluid is expanded and compressed. In these areas, stresses occur between the interfaces liquid / solid. The stresses lead to a removal of the deposits from the surface. The cleaning by the formation and collapse of cavitation bubbles in the microchannels can also not be neglected. Furthermore, the sonotrode is fixedly connected to the microstructured heat exchanger, cleaning effects due to the transfer of vibration cannot be excluded. The microstructured heat exchangers with the ultrasonic sonotrode for the direct and indirect method are shown in Figure 5.

The investigated parameters for the ultrasonic cleaning were:

- (i) The starting time for first impulse  $\tau_1$
- (ii) The time between two ultrasonic impulses  $\tau_D$
- (iii) The duration of ultrasonic impulses  $\tau_T$ .

The power of the ultrasonic generator was set to approximately 100 W measured by a power meter. An ultrasonic frequency of approximately 20 kHz was employed.

Table 1. Dimension of the multi-layer microstructured heat exchanger.

	fouling side	heating side
foil dimension, mm	16 x 16	
number of foils	10	20
number of channel / foil	34	
channel length, mm	14	14
foil thickness, mm	0.3	0.2
channel width, mm	0.2	0.2
channel depth, mm	0.2	0.1
channel wall, mm	0.1	0.1
material	1.4301 (AISI 304)	

### Experimental Setup

The flow diagram of the experimental setup is shown in Figure 6. The setup consisted of a heated water loop controlled by a LAUDA P12 thermostat. The mass flow was measured by a KROHNE magnetic-inductive flow meter. The cooling water loop was cooled by a LAUDA WK500 cryostat. For the fouling loop a KNAUER K1800 HPLC pump was used. The mass flow was measured by a SCHWING coriolis mass flow meter. In order to prevent contamination of the pump and the microstructured heat exchanger, a filter was installed in the fouling loop. The fluid temperature and pressure were measured near the inlets and outlets of the microstructured heat exchanger. The fluid temperature was measured using thermocouples (type K) by CONNATEX and pressure drop in the fouling section is measured using two pressure sensors by KOBOLD. All measured data were recorded automatically by an OMEGA data recorder and a PC.

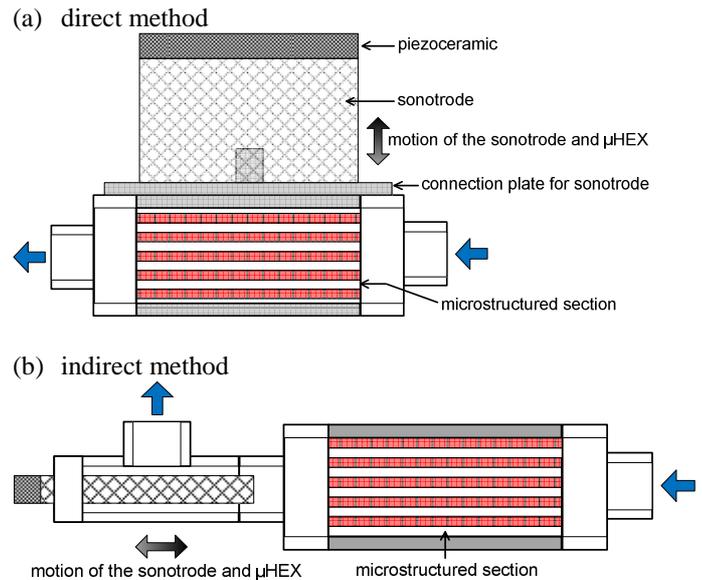


Fig. 4 Microstructured heat exchangers with the different placed ultrasonic sonotrodes

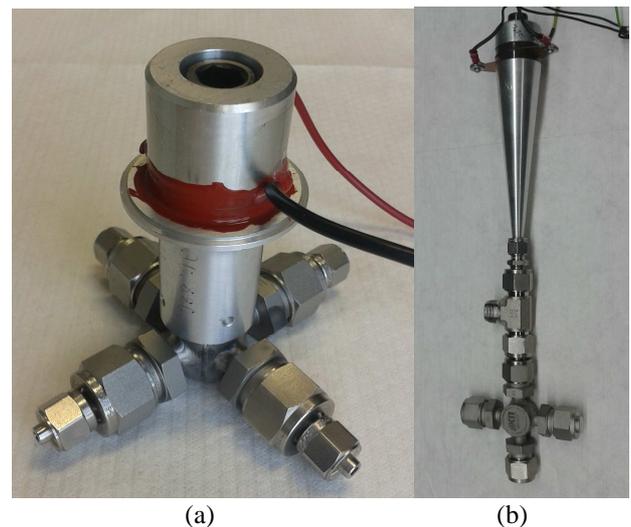


Fig. 5 Microstructured heat exchangers with the ultrasonic sonotrodes (a) directly connected and (b) indirectly connected.

### Fouling investigation

In this study, calcium carbonate ( $\text{CaCO}_3$ ) fouling was chosen because it is a common type of crystallization fouling (Bott, 1997, Hasson, 1986). As  $\text{CaCO}_3$  is poorly soluble in water, a saturated solution of  $\text{CaCO}_3$  was made from calcium nitrate ( $\text{Ca} [\text{NO}_3]_2 \cdot 4\text{H}_2\text{O}$ ) and sodium bicarbonate ( $\text{NaHCO}_3$ ). The  $\text{CaCO}_3$  is inversely soluble. The solubility decreases with increasing temperature. The parameters of the solution are shown in Table 2.

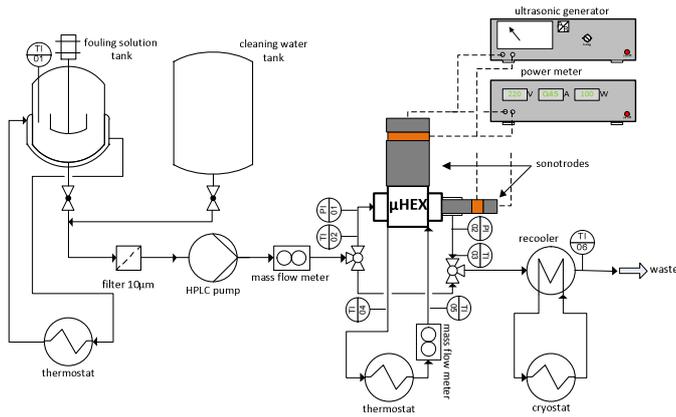


Fig. 6 Flow diagram of experimental setup for fouling investigations.

Table 2. Parameters of the fouling solution in the tank.

$c_{Ca} [NO_3]_2 \cdot 4H_2O$ [mmol/L]	$c_{NaHCO_3}$ [mmol/L]	$T_{sol}$ [°C]	pH [-]
4.99	10.99	23.2	7.1

The parameters for the fouling investigation are shown in Table 3; the values are given for clean conditions. At the beginning of the experiments deionized water was used until a steady state was reached. When constant conditions were reached, deionized water was exchanged to fouling solution and the data recording was started. Based on the measured fluid temperatures in the fouling passage, the fouling behaviour was monitored.

Table 3. Parameters and setting of all experiments for clean conditions

Experiment no.	1	2	3	4	5
<b>heating water</b>					
mass flow, kg/h	86.4	86.5	86.0	86.1	85.9
inlet temperature, °C	94.0	93.9	93.8	94.1	94.0
outlet temperature, °C	89.3	89.2	89.0	89.9	90.3
flow velocity, m/s	1.83	1.83	1.82	1.82	1.82
<b>fouling solution</b>					
mass flow, kg/h	8.96	8.94	8.91	8.87	8.92
inlet temperature, °C	25.8	25.1	26.2	25.5	25.2
outlet temperature, °C	82.5	82.1	82.1	83.3	82.5
flow velocity, m/s	0.19	0.19	0.18	0.18	0.18

### Evaluation of experimental data

During the investigations, the formations of deposits in the microstructures were observed. This resulted in a

deterioration of the heat transfer performance and can be described by the fouling resistance:

$$R_f(t) = \frac{1}{h_f} - \frac{1}{h_0} \quad (1)$$

The time-dependent variable heat transfer coefficient  $h$  can be calculated from the heat flux, the heat transfer area and the logarithmic temperature difference according as

$$h(t) = \frac{\dot{m} c_p (T_{f,in} - T_{f,out})}{A \Delta T_{log}} \quad (2)$$

The logarithmic temperature difference was calculated from the fluids inlets and outlets temperatures as

$$\Delta T_{log} = \frac{(T_{h,in} - T_{f,in}) - (T_{h,out} - T_{f,out})}{\ln \left( \frac{(T_{h,in} - T_{f,in})}{(T_{h,out} - T_{f,out})} \right)} \quad (3)$$

The heat transfer area is calculated from the perimeter, the length and the number of microchannels.

$$A = 4 b_c n_c L_c \quad (4)$$

The quantity of the cleaning is determined by the ratio of the fouling resistance before and after cleaning with ultrasound.

$$\theta = \left( 1 - \frac{R_{f,b}}{R_{f,a}} \right) \cdot 100\% \quad (5)$$

## RESULTS AND DISCUSSION

### Crystallization fouling in Microstructured Heat Exchanger

The typical time dependent behaviour of the fouling resistance in microstructured heat exchanger is shown in Figure 7. The fouling process can be split into two phases: the induction phase, where the heat flux is not changing, and a layer growth phase, where the fouling resistance increases continuously. The fouling behaviour of microstructured heat exchanger is similar to conventional heat exchangers (Bucko et al. 2012, Mayer et al. 2012).

The fouling curve in Figure 7 was determined for the multi-layer microstructured heat exchanger without sonotrodes. The induction time is approximately 20 minutes followed by a growth period. During the crystal growth period the fouling resistance first increased linearly for about approximately 40 minutes. After that, the increase of the fouling curve is declining. Due to the decreasing fluid temperature, lower levels of supersaturation result in a reduction of the fouling resistance.

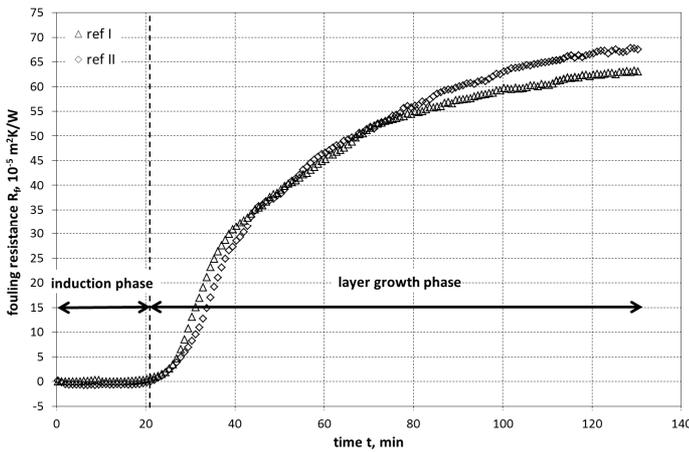


Fig. 7 Fouling resistance over time without ultrasonic cleaning (experimental conditions are the same as in the ultrasonic cleaning experiments no.1 to no.5).

**Ultrasonic Cleaning Experiments**

In Figure 4 the comparison of the indirect and direct ultrasonic cleaning method are shown. The comparison of the cleaning methods shows that the indirect cleaning may be regarded as the best choice. The quantity of cleaning for the experiments is compared in Table 4. At the beginning of the indirect cleaning, we obtained a reasonable good quantity of cleaning of about 47%. After this first good result, the quantity of the cleaning decreased with time. As crystalline layers grow inside the microstructure, the complete removal of the fouling layer with ultrasound becomes more difficult.

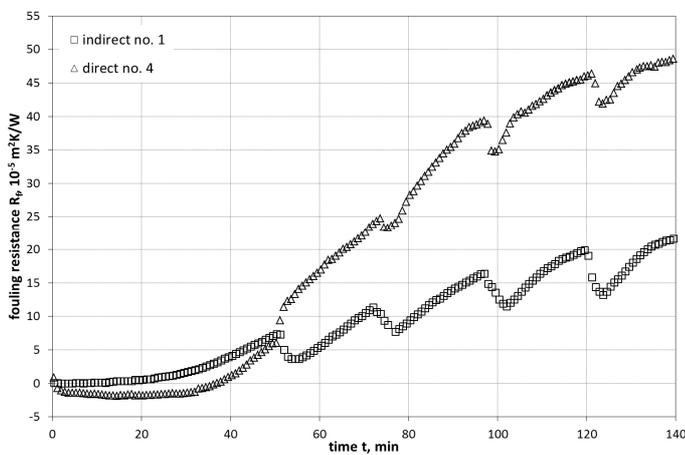


Fig. 4 Comparison of the indirect and direct ultrasonic method. Fouling Resistance over time: ( $\diamond$ ) reference test without ultrasonic cleaning, ( $\Delta$ ) direct cleaning, ( $\square$ ) indirect cleaning.

The comparison of the cleaning methods shows that the higher efficiency was observed by the indirect cleaning. In Figure 5 the results of the indirect cleaning are shown. The interval between the impulses and the duration of the impulses was varied. The interval between the impulses was reduced to 10 minutes and the duration of the impulse was 2 and 4 minutes. With these settings, the curve for experiment

no. 2 clearly shows that a complete clean condition can be achieved. In experiment no. 3, the duration of the impulse was reduced to 2 minutes. This resulted in an increase of the fouling resistance at the end of the experiment, implying that the duration of the impulses is too small.

Table 4 Parameters and results for the direct and indirect ultrasonic cleaning experiments. The value of 100% corresponds to a clean microstructured heat exchanger  $R_f = 0$ .

	indirect			direct	
	no.1	no.2	no.3	no.4	no.5
$\tau_I$ , min	50	20	10	50	50
$\tau_D$ , min	20	10	10	20	20
$\tau_T$ , min	4	4	2	4	4
$\theta$ [%]	1	46.7	100	100	0
	2	25.5	100	100	5.7
	3	28.4	100	100	12.4
	4	32.1	100	100	8.6
	5		100		0.8
	6		100		
	7		100		
	8		100		
	9		85.0		
	10		75.5		
	11		80.6		

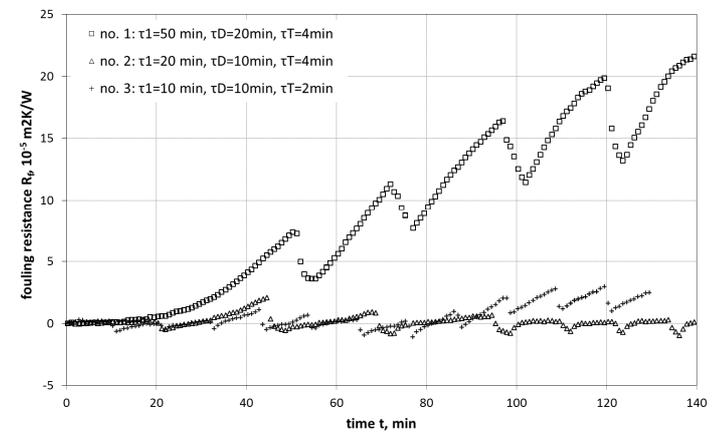


Fig. 5 Fouling resistance over time for the indirectly cleaning experiments with different parameter of the ultrasound impulses.

In Figure 6 an example for the fluid temperatures relative to experiment no.1 is shown. The effect of the ultrasound cleaning resulted in a short increase of the outlet fluid temperature, followed by a further decreasing. With increasing time, a slight increase of the heating water outlet temperature was observed. This increase resulted from the deterioration of the heat transfer due to the deposits in the fouling passage.

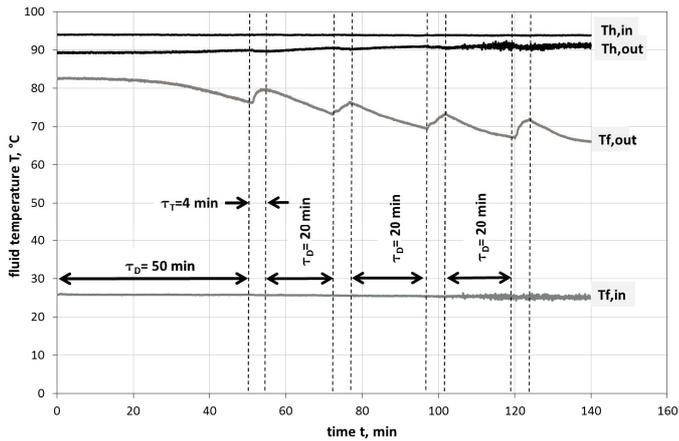


Fig. 6 Fluid temperatures over time for experiment no. 1.

During the investigations for direct cleaning under identical experimental conditions, a decrease of the cleaning efficiency was observed (see Figure 7). Possible reasons for the decreasing cleaning efficiency could be a decreasing transmission of the ultrasonic power into the microstructured heat exchanger: during the experiments, the glued connection between sonotrode and connecting plate was dissolving slowly. The building gap may have led to lower coupling of the ultrasound. For that reason, the actual power transferred to the fluid and the microstructure may have been lower in respect to the nominal power set on the ultrasound generator.

A further observation was that a complete cleaning of the microstructure was not possible, regardless of the time and duration of the ultrasonic impulse. Such a problem can be explained by the formation of local node points in the spread of the ultrasonic wave (standing wave). In these points, the cleaning performance is deteriorated. The same effects were also observed by Legay (2013).

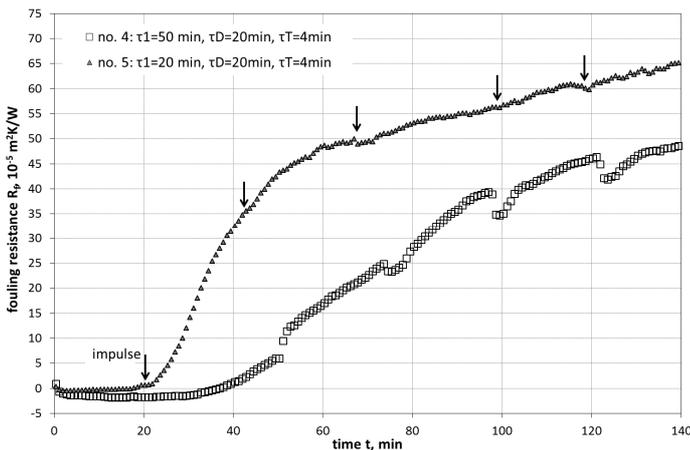


Fig. 7 Fouling resistance over time, comparison of direct ultrasonic experiments; decreasing cleaning efficiency due to problems with coupling of the ultrasonic sources.

## CONCLUSIONS

The experimental investigations showed that CIP using ultrasound in microstructured heat exchanger for crystalline deposits is possible.

1. The best results were obtained for cleaning with the indirect ultrasound method.
2. The first impulses should start at the time when the induction period begins. The duration should be chosen in a way that a sufficient cleaning is achieved.
3. An extension of the operation time, without significant deterioration of thermal and hydrodynamic behaviour of the heat exchanger is possible.
4. The method of introducing the ultrasound in the microstructured heat exchanger has a big impact (direct and indirect) and leads to different results.

## OUTLOOK

In the future, further investigation should focus to:

1. Modulation of the frequency in order to avoid local node points of poor cleaning.
2. Improving the coupling of ultrasound into the microstructure with a better connection between the sonotrodes and the microstructured heat exchanger.
3. Design of an optimized system of heat exchanger and ultrasonic transducer based on simulations.
4. Investigation of combined mechanical and chemical cleaning with ultrasonic and chemical agents.

## NOMENCLATURE

$A$	heat transfer area, $m^2$
$b_C$	width of microchannel, $m$
$c_i$	concentration, $mol/L$
$c_P$	heat capacity, $J/(kg K)$
$h_f$	overall heat transfer coefficient under fouling conditions, $W/(m^2 K)$
$h_0$	overall heat transfer coefficient under clean conditions, $W/(m^2 K)$
$L_C$	length of microchannel, $m$
$m$	mass flow, $kg/h$
$M$	molecular weight, $g/mol$
$n_C$	number of microchannels
$R_f$	fouling resistance, $(m^2 K)/W$
$R_{f,a}$	fouling resistance after impulse, $(m^2 K)/W$
$R_{f,b}$	fouling resistance before impulse, $(m^2 K)/W$
$t$	time, $min$
$T_{f,in}$	inlet fouling temperature, $^{\circ}C$
$T_{f,out}$	outlet fouling temperature, $^{\circ}C$
$T_{h,in}$	inlet heating temperature, $^{\circ}C$
$T_{h,out}$	outlet heating temperature, $^{\circ}C$
$T_{sol}$	fouling solution temperature in tank, $^{\circ}C$
$\Delta T_{log}$	logarithmic temperature difference, $K$
$\tau_1$	time for first impulses, $min$
$\tau_D$	time interval between impulses, $min$
$\tau_T$	duration of ultrasonic impulses, $min$
$\theta$	quantity of the ultrasound cleaning, $\%$

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