Ultrasound based Fouling Detection in Polymerization Processes

Marco Osenberg[1], Jan Förster[2], Stephan Westerdick[1], Jan Tebrügge[2], Esther von Grotthuss[3], Thomas Musch[1]
[1]Ruhr University Bochum, Chair of Electronic Circuits, 44780 Bochum, Germany,
[2]KROHNE Innovation GmbH, 47058 Duisburg, Germany
[3]Covestro Deutschland AG, 51365 Leverkusen, Germany
marco.osenberg@est.rub.de

Abstract

In polymerization processes, unpreventable fouling eventually occurs and results in potential product contamination and increased energy consumption. Formation of this fouling must be detected reliably in real-time to perform obligatory cleaning procedures in time. Unfortunately, measurement technique for this purpose is commercially not available yet. Thus, in the present work a measurement setup is developed that can reliably determine formation of fouling using ultrasound echo measurements.

1 Introduction

During polymerization reactions, undesirable deposits form on the walls of pipes, mixers and reactors. This leads to product contamination and has a heat-insulating effect in heat exchangers [1]. Both, the product contamination as well as the heat-insulating effect cause unnecessary waste of resources and energy and thus need to be avoided. Furthermore, if fouling in chemical plants growth to critical thicknesses, clogging of the whole plant might occur which results in unplanned downtime of the plants and can even cause permanent damage to components of the plants.
Up to now, plant operators try to prevent fouling by performing cleaning procedures on a regular basis. However, these regular cleaning procedures do not always prevent from unforeseen downtime of the plants. Furthermore, as these cleaning procedures are planed with a lot of safety margin, they offer a lot of potential for process optimization. In order to prevent downtime due to fouling and to optimize the cleaning procedures plant operators must know the state of fouling in their plants. Although there has been numerous research on such sensors [2-4], there is no industrially available measuring method to detect fouling in such processes yet.
Thus, this work aims on the integration of ultrasonic echo measurement technique into the process to allow for monitoring the beginning and continuous growth of fouling.

2 Polymerization Process

In order to investigate the concept of using ultrasound echo measurements to monitor fouling, trimerization of hexamethylene diisocyanate (HDI) was selected as an exemplary chemical process. This substance system is well suited for ultrasonic measurements, since neither does outgassing occur during the reaction nor do the raw materials contain solid particles. Therefore, less interference in the measurement signals and only a moderate attenuation of the signals are to be expected.
To detect fouling by means of ultrasound echo measurements, the fouling must differ in its acoustic impedance from the base material. This requirement is fulfilled by HDI as has already been demonstrated in previous work [5]. It was found that HDI has a speed of sound (SOS) of c_{HDI} = 1414 m/s at a temperature of T = 23.5 °C and a density of ρ = 1050 kg/m³. Therefore, according to formula (1), the acoustic impedance of this material system is Z_{HDI} = 1.5 MRayl.

$$Z = c \cdot \rho \tag{1}$$

Compared to the base material, the fouling has an almost similar density with an increased sound velocity, which results in a low reflection at the interface to the fouling.

3 Ultrasound Measuring Cell

The ultrasound measuring cell used in previous work [5] had some disadvantages. As it was made completely of polyether ether ketone (PEEK) and had a homogenous flow profile in its measuring chamber, only few or none fouling occurred in that measuring cell. Furthermore, the old cell design did not allow for temperature control of the fluids thus temperature effects had a much stronger influence on the obtained results than the effects due to fouling.

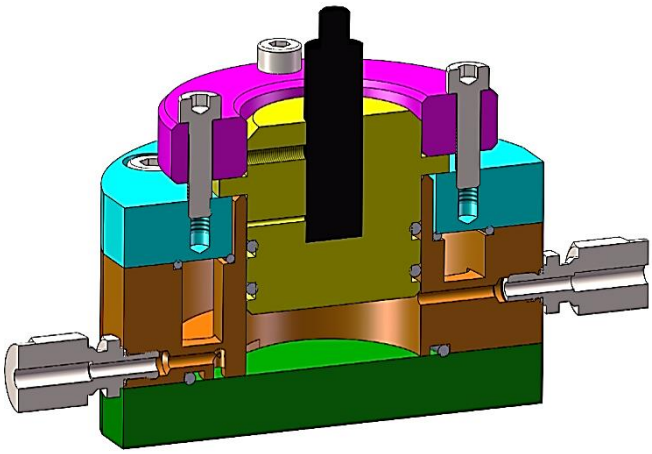

Figure 1 3D model of the new design of the ultrasonic measuring cell used in this work

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Therefore, in this work an improved version of the measuring cell was constructed as shown in Figure 1. First, the flow profile inside the measuring chamber is now designed in a way which introduces areas of long dwell time so formation of fouling in the chamber is to be expected. Second, the new cell design allows for temperature control thus temperature effects can be neglected. And third, despite the inlet section which is still made of PEEK the new measuring cell is completely made of stainless steel 1.4404. PEEK was chosen for the inlet section, because its acoustic impedance matches much better to the acoustic impedance of the fluid than the acoustic impedance of stainless steel. Thus, less ultrasonic losses are to be expected at the transition from the inlet section into the fluid.
The measurement channel is now of cylindrical shape with a height of d_C = 10 mm and a radius of r_C = 15mm. This prevents interference from the channel walls. For the measurements presented in this work a transducer with a center frequency of 19.2 MHz (3 mm aperture diameter, 12.2 MHz bandwidth, Olympus Corp., USA, model V316-SM) has been used.
Although this new cell design is temperature controlled, two PT-100 temperature sensors (class A) are used to monitor the temperature in the measurement cell to detect even very small temperature changes due to exothermal reactions during the formation of fouling.

4 Measurement Setup

The ultrasonic transducer is driven by an experimental MOSFT output stage, which generates a pulse with a frequency of 20 MHz and a voltage of UPA = 50 V. The generated pulse is transmitted to the ultrasonic transducer via passive decoupling. The reflection and excitation signals are transferred to the amplifier circuit through a limiter stage to a filter and amplification stage. For analog filtering, a 6th order bandpass with a lower limit frequency of f_u = 0.5 MHz and an upper limit frequency of f_o = 40 MHz was realized. The digitization of the reflection signals is performed with an oscilloscope (Rohde & Schwarz GmbH, Germany, model RTO 1004). The data is received, evaluated and stored with the software Matlab™.
To increase the signal-to-noise ratio (SNR), 20 consecutive measurements are taken and the average value is calculated. The measurements have a small time offset of approx. 150 µs, so that a static behavior in the measuring cell can be assumed during these measurements. The flow rate in the experiments is Q_c = 11 ml/min with a total volume of the measuring cell of V_c = 7.07 ml.
Figure 2 shows the received signal after average value calculation. The reflection point at the coupling from the PEEK into the fluid is clearly visible at 8.7 µs after excitation. The second multiple reflection of the coupling section occurs after 17.2 µs. The back-wall reflection for the cell filled with HDI occurs at 22.95 µs.

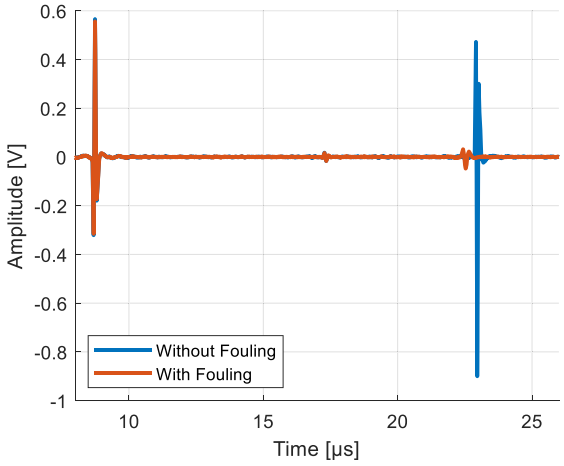

Figure 2 received signals, without fouling (blue) and with fouling (red)

5 Signal Evaluation

The measured reflection signals are again digitally filtered for evaluation, with the limit frequencies f_u = 2 MHz and f_o = 30 MHz, in order to minimize interference even further. Afterwards, the envelope of the signal is determined with the Hilbert transformation. At this point, the processed data is the basis for further evaluation of the measurement. In contrast to earlier work [5], multiple properties are taken into account to determine formation of fouling to improve the reliability of the measurement setup.
In the experiments, two influences of the fouling on the received reflection signals were analyzed. The fouling changes the average sound velocity (ASV) in the measuring cell as well as it attenuates the received signal power. A basic calibration is done by correlating the received signals with the mechanical measured fouling layer thickness after ending an exemplary experiment. Unfortunately, this calibration is subject to substantial uncertainty because the ending procedure includes an inerting phase, which alters the properties of the fouling layer.
To determine the ASV, the local maxima of the envelope are searched to find the reflection points. To determine the reflection time as accurately as possible, a centroid analysis is applied via the maxima. The ASV is then determined via the coupling time t_1 and the back wall reflection time t_2 using formula 2. The ASV changes over the growing layer formation as can be seen in **Fig. 3**.

$$c_a = \frac{d_C}{t_2 - t_1} \tag{2}$$

The dependence of the sonic velocity on the temperature can also be clearly seen in this graph. At the beginning of the reaction, about 6 minutes after the start of the test, the temperature increases due to the exothermic chemical reaction. This has the consequence that the ASV in the measuring cell decreases.

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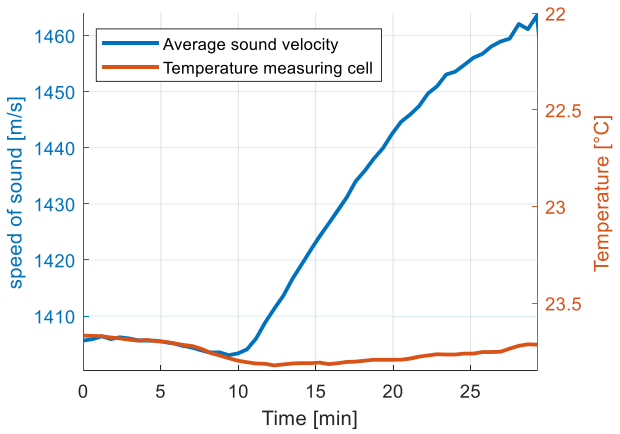

Figure 3 Average of the measured sound velocity (blue) measured temperature (red)

To calculate the thickness of the fouling from the previous information and the ASV, formula 4 is used. For this purpose, the previously determined sound velocity for the fouling of c_F = 1853 m/s is assumed. This was analyzed in previous tests for a cured fouling. However, in its formation phase, the sound velocity of the fouling will be significantly different from that value. In order to eliminate the temperature effects from the reconstruction, the speed of sound as a function of temperature is used for the HDI. For this purpose, a linear approximation according to formula 3 was established around the operating temperature of 23.5 °C.

$$c_{HDI}(T) = 2422\frac{m}{s} - 42.9\frac{m}{s\cdot°C}\cdot T \qquad (3)$$

$$d_{ASV}(T) = \frac{\frac{d_C}{c_a} - \frac{d_C}{c_{HDI}(T)}}{\frac{1}{c_F} - \frac{1}{c_{HDI}(T)}} \qquad (4)$$

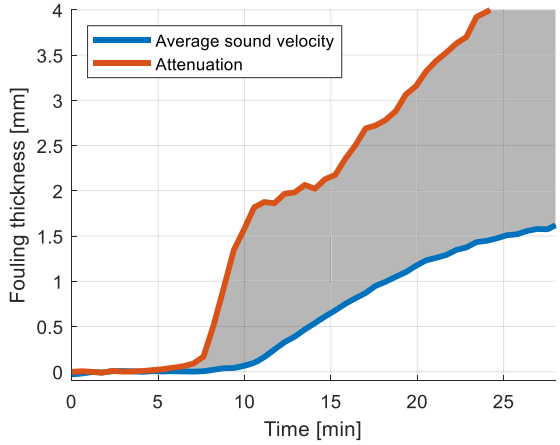

Figure 4 Estimation of the fouling thickness, calculated via average sound velocity (blue) and attenuation (red)

The evaluation of the signal attenuation was calculated on the basis of the envelopes. The maximum value of the reflection points is compared with the reflection without fouling. In previous tests it was found that an attenuation of approximately a_F = 10 dB/mm occurs. In the attenuation curve, a very rapid increase in attenuation can be clearly seen at the start of the reaction. This can be attributed to the uneven growth of the fouling and the surface roughness. This causes the ultrasonic signals to be scattered and therefore attenuated.

However, **Fig. 4** shows the estimated fouling layer thickness based on the changes of the ASV and the attenuation. Due to the systematic uncertainties during calibration, the fouling layer thickness is underestimated by the method based on the ASV. In contrast, the fouling layer thickness is overestimated by the method based on the attenuation due to scattering and refraction effects. Therefore, the actual fouling thickness is between these two curves. In the experiment, an average fouling thickness of 3 mm was determined after the test, which lies in the region between these two curves. Thus, it can be clearly stated that growth of fouling can be detected and quantitatively determined by this measuring system.

6 Results

The growth of the fouling in this polymerization process can be observed with good accuracy using ultrasound echo measurements. The acoustic properties of the monitored chemical system must be very well known to allow for reliable reconstruction of fouling thickness. With the presented methods based on ASV and attenuation, upper and lower limits for the fouling thickness can be estimated accurately. A combination of both reconstruction methods increases the reliability of the system.

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