Use of Ultrasonic Sensors for Characterization of Membrane Fouling and Cleaning

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ABSTRACT

The modern challenges for membrane separations in a wide range of processes require more sophisticated approaches for the detection and remediation of fouling, i.e., the association of solutes, particulate matter, and colloids on and/or within a membrane. Most commonly, fouling is assessed from inferred measurements of permeation rate and/or permeate quality. The use of acoustic techniques for direct observations of membrane fouling was introduced over 10 years ago. We summarize here, recent developments in ultrasonic reflectometry that use both time-domain and frequency-domain spectra for noninvasive, real-time assessments of fouling in a variety of module configurations and geometries. In addition, we describe recent developments and applications of scanning acoustic microscopy (SAM) for post-mortem characterization of membranes with particular emphasis on biofouling.

INTRODUCTION

Many different filter media are currently available for use in a broad variety of liquid- and gas-based separations. Of these media, nonwovens and membranes have been gaining increased attention, where the latter include polymeric, ceramic, metallic or composite materials. Although microporous membranes have been successfully utilized in industrial micro- and ultrafiltration applications, membrane fouling continues to be the key challenge which limits the operations of membrane-based liquid separations.

Fouling occurs when the component(s) filtered from the feedstream collect near the membrane/fluid interface. The earliest stage of the fouling process is characterized by concentration polarization (CP) associated with a boundary layer, in which a gradient of excluded products forms near the membrane surface [1]. Under some conditions the excluded product can associate with the membrane surface or membrane pores, forming what is generically known as a fouling layer (*Figure 1*). Membrane fouling is a complex phenomenon that depends upon the type of foulant(s), the feed concentration, oxidation/reduction conditions, temperature, pH, ionic strength, and separation system hydrodynamics [2].

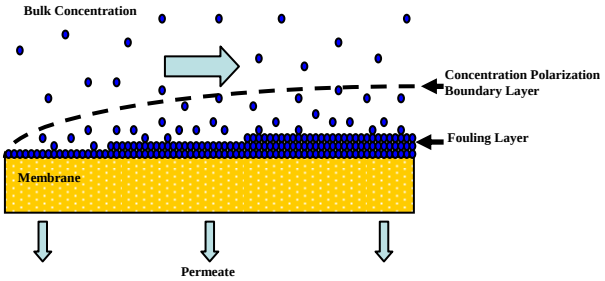

FIGURE 1. Schematic representing the development of a concentration polarization boundary layer (CPBL) and subsequent fouling of a membrane during a cross-flow filtration.

This fouling layer usually has an adverse effect on separation performance by decreasing flux and selectivity as well as serving as a source of contamination. Inorganic fouling or scaling occurs when local concentrations exceed saturation and solid phases precipitate from solution near the membrane surface, or in the membrane pores [3]. Other common types of fouling often occur during the separations of select biopolymers such as proteins [2], as well as with the retention and selection of microorganisms which that form biofilms [4].

This review summarizes developments regarding the use of non-destructive acoustic testing (NDT) as a novel *in-situ* method to monitor inorganic fouling and biologically associated fouling (commonly referred to as "biofouling") of membranes during liquid separation processes. Recent acoustic studies have reported considerable success in real-time monitoring of inorganic scaling processes on a variety of membranes. In contrast, monitoring of biofouling by

acoustic means has proven more challenging due to the fact that many biological foulants present amorphous dynamic morphologies when associating with membranes. Further, the acoustic properties of biopolymer conglomerates are often very similar to the water environment in which they grow [4]. However, improved acoustic detection methods are being developed that may enable these difficulties to be overcome.

Membrane-based liquid-separation processes are generally monitored by the measurement of permeate-flow volume (flux). However, this approach reflects a performance condition as a conglomerate of the entire membrane surface. This flow-based approach and at best provides delayed information on the extended effects of foulants associating with membranes, and cannot provide any information on local membrane conditions. Delays between the actual interactions of membranes and foulants and performance degradation, can adversely affect the quality of the filtration product(s), the efficiency of the process, as well as the useful life of membrane materials themselves. In addition, performing isolated flux decline measurements is not always a productive strategy to gage the extent of fouling, since permeate flux decline may be due to other factors such as membrane compaction and concentration polarization [5].

Thus, it is desirable to develop a method that can detect, in real time, the earliest interactions between foulants and membrane materials during the formation of fouling layers, so that remedial measures, such as membrane cleaning or replacement, can be more efficiently implemented. Ultrasonic time-domain reflectometry (UTDR) as well as ultrasonic frequency-domain reflectometry (UFDR) can be used for this purpose in a manner that does not damage the membrane or alter the function of a liquid separation system. Ultrasonic methods can also be correlated and verified with standard post-mortem characterization techniques, such as microscopic and gravimetric analysis. Recent work has also demonstrated that ultrasonic methods can be applied for post-mortem characterization via scanning acoustic microscopy (SAM). This novel approach leveraging both *in-situ* and post-mortem analysis, provides the basis for a comprehensive monitoring methodology for fouling formation, as well as characterizing the distribution and abundance of conglomerate fouling layers.

ACOUSTIC FUNDAMENTALS: REFLECTION AND TRANSMISSION OF SOUND WAVES

Ultrasonic NDT has been well-documented for a wide range of applications, including the inspection of metal welds to the characterization of a wide variety of material properties. When a longitudinal sound wave propagating in a medium encounters an interface with a different medium, part of the energy of the wave will be transmitted forward through the interface, and part of the energy will be reflected, resulting in an echo. Such an interface can be the location where two thin layers meet (e.g. thickness measurements of industrial films), a flaw in a material (weld inspection), or pores in a porous substrate (geological mapping). The proportions of acoustic wave energy that are transmitted or reflected are determined by the acoustic impedance mismatch of the two media at the interface. Simple expressions describing the physical situation include:

$$R = \frac{Z_2 - Z_1}{Z_2 + Z_1}; \quad T = \frac{2Z_2}{Z_2 + Z_1}; \quad R + T = 1, \tag{1}$$

where Z_1 and Z_2 are the respective acoustic impedance of the two media, R is the reflection ratio, and T is the transmission ratio of the wave. Note that these ratios apply for a sound wave at normal incidence, and that oblique incidence requires more complicated relationships. Acoustic impedance (Z) is typically measured in Rayls (N s/m^3) and physically represents the ratio of sound pressure to particle velocity at a point in a given material. The acoustic impedance of a material is defined as the product of density (ρ) and acoustic velocity (c) in the medium:

$$Z = \rho c. \tag{2}$$

The concept of impedance in acoustics is similar to the relative index of refraction in optics. The higher the impedance mismatch, the more is reflected. Thus, it is often advantageous to attempt to reduce the impedance mismatch between a transducer and the loading medium or specimen being inspected. A coupling medium (gel or grease) is often applied between a transducer and the "loading" medium to avoid unwanted reflections from small air pockets. Air has a very large acoustic impedance mismatch with most solid and liquid materials (dry air at 0°C: 0.4286 kRayl; liquid water at 20°C: 1.494 MRayl; aluminum: 17.33 MRayl; tungsten: 101.0 MRayl [6]). This coupling or matching medium improves transducer efficiency, sensitivity and signal-to-noise ratio.

REAL-TIME MONITORING OF MEMBRANE FOULING

Based on the key concepts of acoustic impedance, transmission and reflection, we now consider applications of using real-time ultrasonic acoustic

spectra for monitoring membrane fouling processes in liquid separation processes. UTDR is one of the fundamental modes of operation in much of ultrasonic NDT technology. In this technique, an ultrasonic pulse is generated and then reflected from an interface between two different media. As described above, a portion of the wave is transmitted, and a portion reflected. If the transducer is oriented normal to the interface, the reflected wave will return to the transducer as an echo that can be recorded for analysis. Knowledge of sound velocity in all pertinent media allows the determination of the distance from the interface to the acoustic transducer [7, 8]. This is particularly useful when trying to determine the thickness of material layers [8-10].

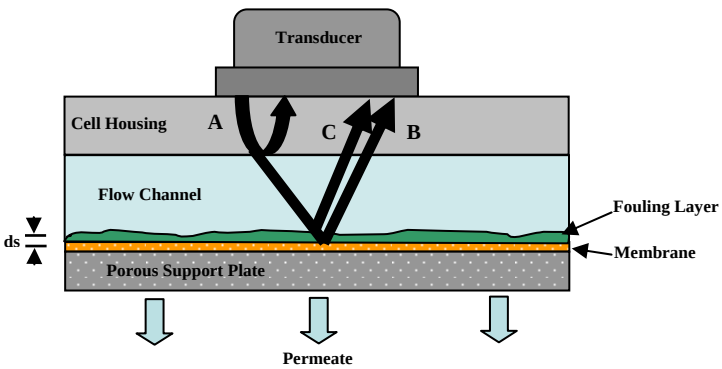

FIGURE 2. Schematic of an externally mounted ultrasonic transducer and the echoes used to monitor the formation and growth of a fouling layer during cross-flow filtration.

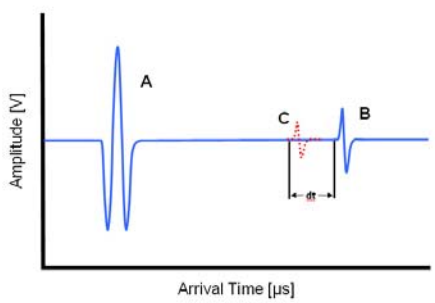

FIGURE 3. Schematic representation of time-domain echo waveforms recorded by an oscilloscope for the arrangement shown in Figure 2. The arrival time of the echo shifts to smaller values (left) after formation and growth of the fouling layer.

Figure 2 presents a schematic cross-section of a flat sheet cross-flow membrane filtration cell. An ultrasonic transducer is placed in contact with the external shell of the flow cell, and a pulse is generated. The pulse is first reflected from the interface at the inside of the shell (reflection A in *Figure 2*). This reflection is recorded, typically with an oscilloscope that is connected to the transducer; a characteristic spectra is presented in *Figure 3*. Part of the pulse moves forward through the flow channel, and then reflects from the membrane surface (reflection B in *Figure 2*). When the membrane is clean, this reflection is recorded as the echo waveform B (*Figure 3*). When a fouling layer forms, however, the interface moves closer to the transducer by a distance, *ds*, and the result is a departure of the echo in the time domain (to the left); thus, echo C can be observed, shifted from echo B by an amount *dt*

$$dt = \frac{2ds}{v} \tag{3}$$

where *v* is the velocity of sound in the fluid medium. This time domain echo shift is observed by overlaying the recorded signal from a fouled membrane over that of an otherwise clean membrane under the same process conditions.

In addition to echo time shift, the amplitude of the echo is also of interest for the analysis of the acoustic spectra. The relative echo amplitudes are dependent on the combination of the order of the reflections (echo A occurred before B, and B was an echo from the wave transmitted at A) and the relative acoustic impedance mismatch (larger mismatch, larger echo). In *Figure 3*, echoes C and B are ideally seen to be distinct and separate in time-domain spectra (signal from fouled membrane on signal from clean membrane). If the fouling layer is relatively thin or diffuse, however, the start time of the echo may actually only shift slightly to the left.

Fouling Detection via UTDR

Previous reports on the monitoring fouling growth with UTDR has been performed using a flat-sheet cross-flow separation system (*Figure 2*). Bond et al. [11] first described use of UTDR to study membrane compaction and fouling with high-frequency transducers. Paving the way for future researchers, Mairal et al. [12] demonstrated the efficacy of the acoustic technique to observe non-biological fouling, and successfully interpreted real-time acoustic spectra to monitor the effectiveness of cleaning procedures on membrane surfaces. In this pioneering study, UTDR was applied in a high-pressure flat sheet module containing reverse osmosis (RO) membranes fouled with calcium sulfate ($CaSO_4$). Here, multiple transducers were used to monitor fouling at different locations along the flow axis of

the cell. The feed pressure and feed concentration were maintained constant, but axial velocity was varied. A decline in ultrasonic echo amplitude as a scaling layer formed corresponded well with a decline in permeate flux, which was also monitored along axial locations corresponding to the ultrasonic transducers. Mairal et al. [5] also demonstrated that relative amplitude changes of ultrasonic time-domain signals were indicative of the inception and maturation of an inorganic fouling layer on RO membranes. The amplitude values either increased or decreased with respect to a baseline waveform, depending on the extent of the fouling layer.

Li et al. [10] utilized UTDR methodology to monitor fouling during ultrafiltration (UF) of paper mill effluent. Advanced fouling formation was detected by comparison of a reference echo waveform from a clean membrane and a time-shifted echo waveform from a fouled membrane. The authors reported that changes in the echo signal were also observed after cleaning of the fouled membrane. More recent reports have described the ability of real-time UTDR for detection of fouling layers on the surfaces of flat sheet [13, 14] and hollow-fiber membranes [15] used for drinking-water treatment. Li et al. [16] also indicated successful application of real-time UTDR for the detection of protein fouling on polysulphone UF membranes and a subsequent work reported that real-time UTDR successfully detected protein fouling on tubular UF membranes [17].

A membrane module geometry distinct from those described above is the spiral-wound arrangement (*Figure 4*); spiral-wound modules are often the "work horses" for industrial membrane-separation applications due to their much larger surface area to volume ratio. The geometry of a spiral-wound membrane module presents significant challenges to the application of UTDR. First, a more complicated echo signal will result from the superposition of the multiple reflections from the module housing (not shown in *Figure 4*) and multiple membrane layers. Second, the ultrasonic pulse is significantly attenuated by multiple reflections, since only a fraction of the initial wave energy passes through from one interface to the next. This makes observation of the innermost layers particularly difficult. Despite these limitations, a series of studies reported that UTDR was successfully implemented for the detection of inorganic scaling as well as subsequent cleaning in spiral-wound membrane modules [5, 18, 19]. Of particular importance is that the ultrasonic results were confirmed with the use of independent techniques including post-mortem gravimetric measurements and scanning electron microscopy (*Figure 5*).

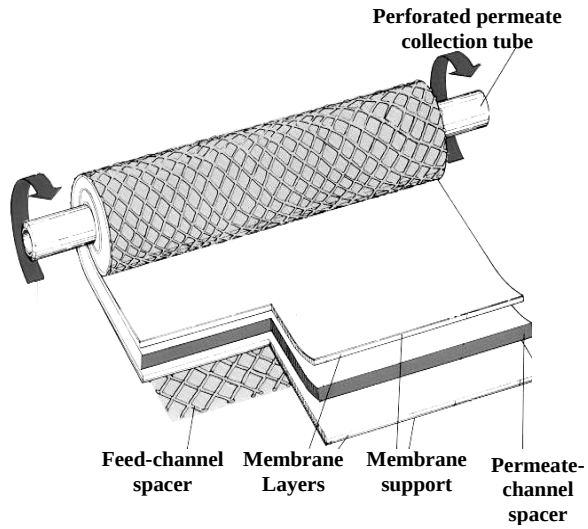

FIGURE 4. Schematic drawing of a spiral-wound membrane module showing the concentric geometry and multiple layers consisting of feed-channel spacers, membrane layers and permeate-channel spacers.

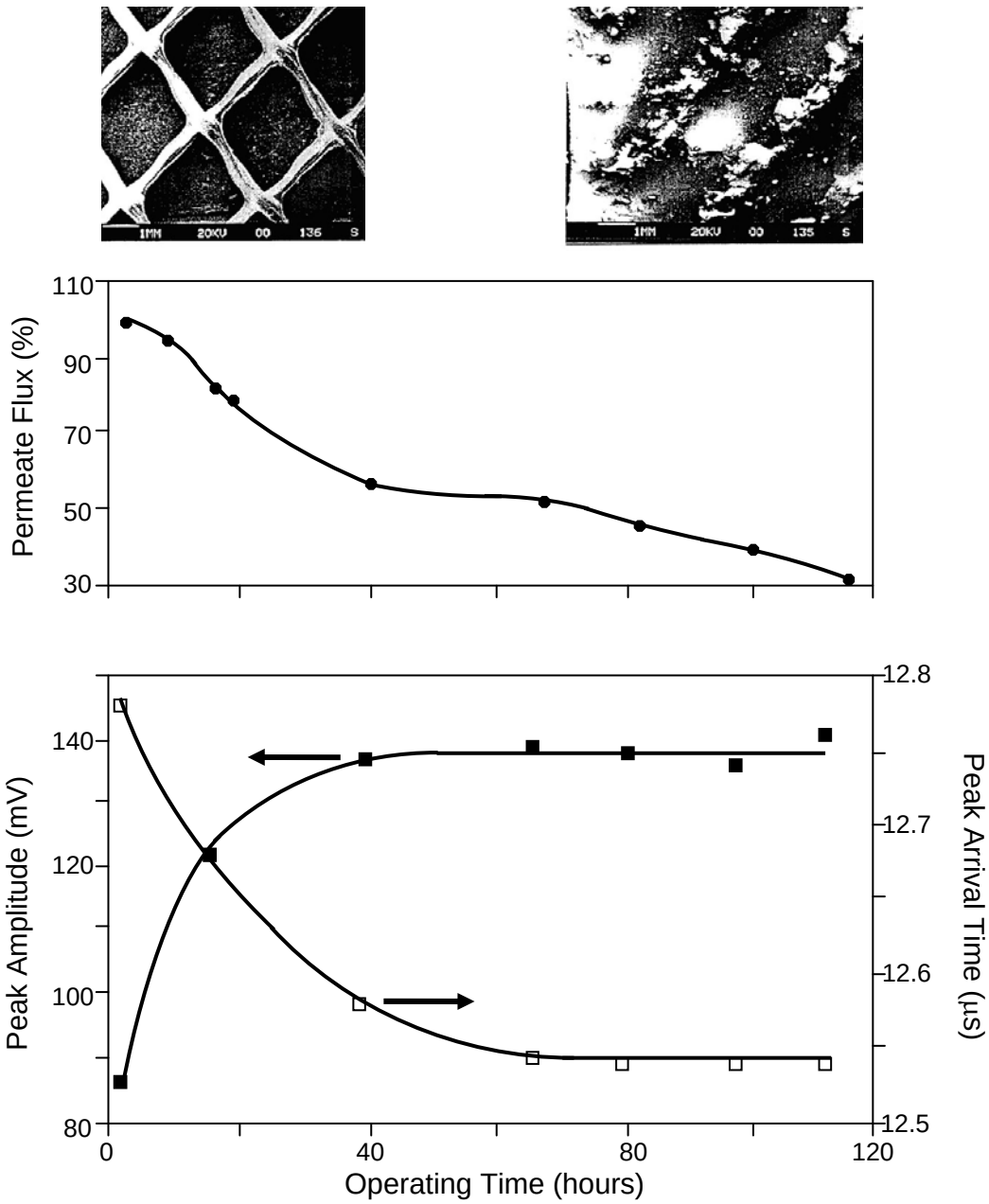

FIGURE 5. Representative results obtained from experiments employing a spiral-wound membrane module and ultrasonic sensors. The data indicate an overall decrease in permeate flux due to inorganic scaling (calcium sulfate) and a corresponding monotonic increase and decrease in ultrasonic peak amplitude and arrival time, respectively. Inset: low-magnification micrographs showing clean (initial) and fouled (final) surface conditions of spacer in agreement with flux and ultrasonic data.

Fouling Detection via UFDR

In addition to time-domain methods described above, an approach based upon frequency-domain analysis has proven useful for membrane fouling determination, in particular, in those situations where multiple reflections cause spectral overlap, or where significant noise is present. In brief, the square of a Fourier transform is assigned the power spectrum of the signal. The overall magnitude of the power spectrum, also known as total reflected power (TRP), can be represented by the peak magnitude or the area under the spectrum, and is a quantitative measure of the amplitude or power contained in the signal as a whole. Changes in TRP due to the presence of a

fouling layer may be observed even if a corresponding change in the time-domain signal may not be apparent. Recent work [20] has suggested that ultrasonic measurement using TRP is particularly effective for the difficult task of monitoring the early-stages of biofouling.

Biofouling is a generic term referring to the flux decline caused by the association of microbes and/or biopolymers associating with (or in) membrane surfaces; this can include but is not limited to biofilms, which consist predominantly of polymerized sugars excreted by bacteria. These exocellular polysaccharides (EPS) are dynamic hydrogels, which not only entrain bacterial cells, but also encapsulate a host of organic and inorganic particulate matter near the critical surface(s) performing separation.

In a series of biofouling observations [21], commercial polyvinylidene fluoride (PVDF) microfiltration membranes (nominal pore size of 0.1 µm) were fouled under laminar conditions, in a cross-flow filtration cell. The microfiltration (MF) membrane module was initially operated with ultrapure water to enable complete membrane setting (compaction). Subsequently, a feed solution conducive to biofilm growth was introduced and fouling was monitored for 70 hr. Fouling rates were quantified by ultrasonic reflectometry using a planar 10-MHz ultrasonic transducer with an acoustic field (sampling area) of ~8 x 10^6 μm^2. The permeation flow-rate was simultaneously monitored. The arrival time and amplitude of the reflected sound waves were recorded and compiled into frequency distributions by Fourier transform using methods previously developed and described by Ramaswamy [22, 23].

The total reflected power from each acoustic observation was determined by integrating the amplitude over the frequency spectrum of the reflected sound waves through the range between 0-10 MHz. Those TRP values obtained from fouled membranes, were compiled and normalized to their initial value and compared with a normalized permeate flow-rate (*Figure 6*). Results indicated a rapid initial increase (>7%) in TRP, which was followed by a gradual increase such that after 70 hr of exposure, a total TRP increase of > 20% was observed. Over the same period, the permeate flow-rate decreased by more than 70%. Significant variability in the TRP values was evident during the course of this and other biofouling tests, and may be attributed to the fact that biofilms are dynamic in nature and their morphology constantly changes depending on microbial community structure, growth rates, age, substrate concentrations, and local hydrodynamic conditions. In addition, a portion of TRP variability where biofouling is concerned may be attributed to capricious growth, movement and sloughing. Findings of the study presented here suggest that the use of UFDR is effective for monitoring membrane biofouling; however, many different modes of biofouling exist, and further studies are required for optimization of the technique.

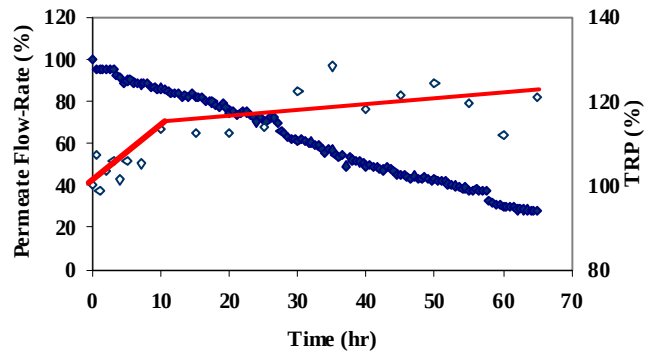

FIGURE 6. Permeate flow-rate (primary y-axis, ♦) and TRP (secondary y-axis, ◊) changes (%) plotted as a function of time (hr) during membrane fouling using a feed solution conducive to biofilm growth.

POST-MORTEM CHARACTERIZATION OF MEMBRANE BIOFOULING USING SCANNING ACOUSTIC MICROSCOPY (SAM)

In order to validate results obtained from ultrasonic reflectometry, a number of standard post-mortem techniques must be used to analyze a membrane after biofouling. Although biofilms have been observed using various microscopic methods, optical quantification is quite difficult and the degree of accuracy is uncertain. Common means for the morphological observation of biofilms have been epifluorescence microscopy (EFM), environmental scanning electron microscopy (ESEM), and confocal laser scanning microscopy (CLSM). ESEM and CLSM can also be used to characterize inorganic scaling layers. Each of these techniques has associated operating and interpretation obstacles. EFM involves staining microbial cells with fluorescence dyes in which the membrane may interfere with the observations (e.g. high backgrounds); in addition, EFM requires the removal of microorganisms before their analytical observation. ESEM and CLSM are expensive and

labor intensive, and often result in destruction of the biological foulants themselves.

Given the limitations of these microscopic methods, scanning acoustic microscopy represents a promising tool that provides an accurate picture of the structure of biological foulants deposited on membrane surfaces. Confirmation of biofilm occurrence, particularly in the early stages of growth, is critical to the efficient and cost-effective operation of many industrial and medical systems, and may be used to initiate appropriate counter measures to prevent its maturation. Only a limited number of studies have reported results using SAM for the post-mortem characterization of biofouling on polymeric substrates.

Kujundzic et al. [20] used UFDR to monitor early-stage biofouling on porous PVDF MF membranes (nominal pore size: 0.65 µm). Membrane coupons were placed in a biologically active annular reactor for up to 300 days, and subjected to a constant shear field (0.12 N m^{-2}), which induced sessile microbial growth from acetate-amended municipal tap water. Ultrasonic monitoring was non-destructively performed by traversing coupons in a constant temperature water bath using a spherically focused 20-MHz immersion transducer. This semi-automated system was configured to obtain reflections from 50 regions (sampling area of 120×10^3 μm^2) distributed evenly near the centerline of each coupon. The total reflected power from each sampled region was determined as previously described, and frequency histograms were compiled. The data were tested for normality, and compared using widely accepted statistical metrics.

Results indicate that the TRP spectra from the PVDF membranes are not normally distributed as judged by Anderson–Darling test (*Figure 7*). The reflected power distributions were compared (90% confidence level) with a standard biochemical assay that describes surface-associated biofilms [24]. Using exocellular polysaccharides (EPS) as a surrogate measure of total biofilm mass, UFDR was able to detect biofilms developing on membrane material tested at surface-averaged masses of ≤150 µg/cm^2. Above these threshold levels, increasing amounts of exocellular polysaccharides correlated with a significant decrease in total reflected power (*Figure 7*). When compared to clean (virgin) conditions, biofilms growing on coupons induced consistent attenuations in reflection amplitude, which caused statistically significant shifts in the reflected power ($p<0.1$). These results suggest that UFDR may be used as a non-destructive, post-mortem tool to monitor biofouling in a wide variety of applications.

SAM has also been used to characterize protein fouling on MF membranes [25]. PVDF MF membranes (nominal pore size: 0.2 µm) were fouled with a commercial protein, bovine serum albumin (BSA), in dead-end flow cells at a transmembrane pressure of 13.8 kPa (2 psi). A BSA solution with a concentration of 1g L^{-1} was prepared and then filtered through the MF membranes, and coupon sections were obtained for SAM analysis.

The TRP from the clean and protein-fouled membrane coupons are presented in *Figure 8*. Total reflected power values from clean and protein-fouled membrane coupons presented a non-normal distribution. The statistical standards in this study were established by choosing a 95% confidence level. When compared to clean conditions, BSA-fouled membrane coupons induced consistent attenuations in reflection amplitude, which caused statistically significant departures in reflected power ($p<0.05$). The fouled membrane coupons showed a TRP departure towards lower values when compared to the clean membrane coupons. The findings based on the acoustic response of the protein-fouled membranes showed that UFDR can differentiate between clean and protein-fouled membranes.

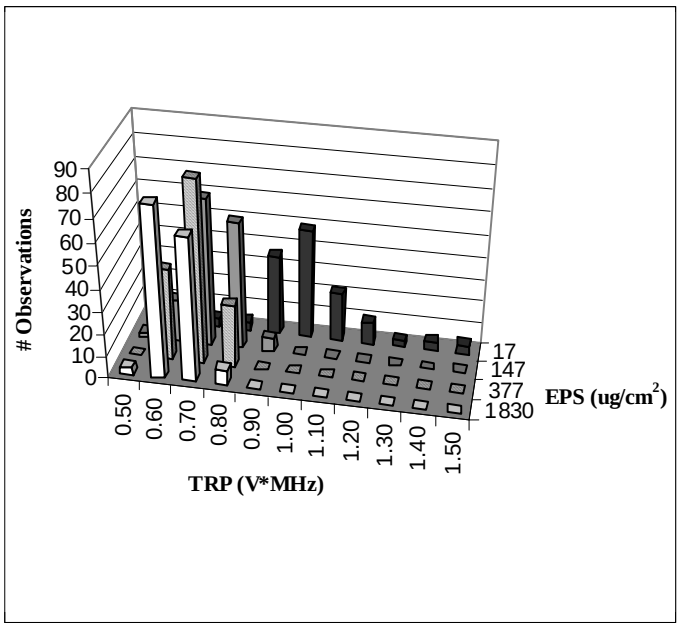

FIGURE 7. Distributions of the total reflected power (TRP) from polyvinylidene fluoride (PVDF) MF membrane coupons. TRP spectra are systematically altered in response to an increasing mass of exocellular polysaccharides (EPS).

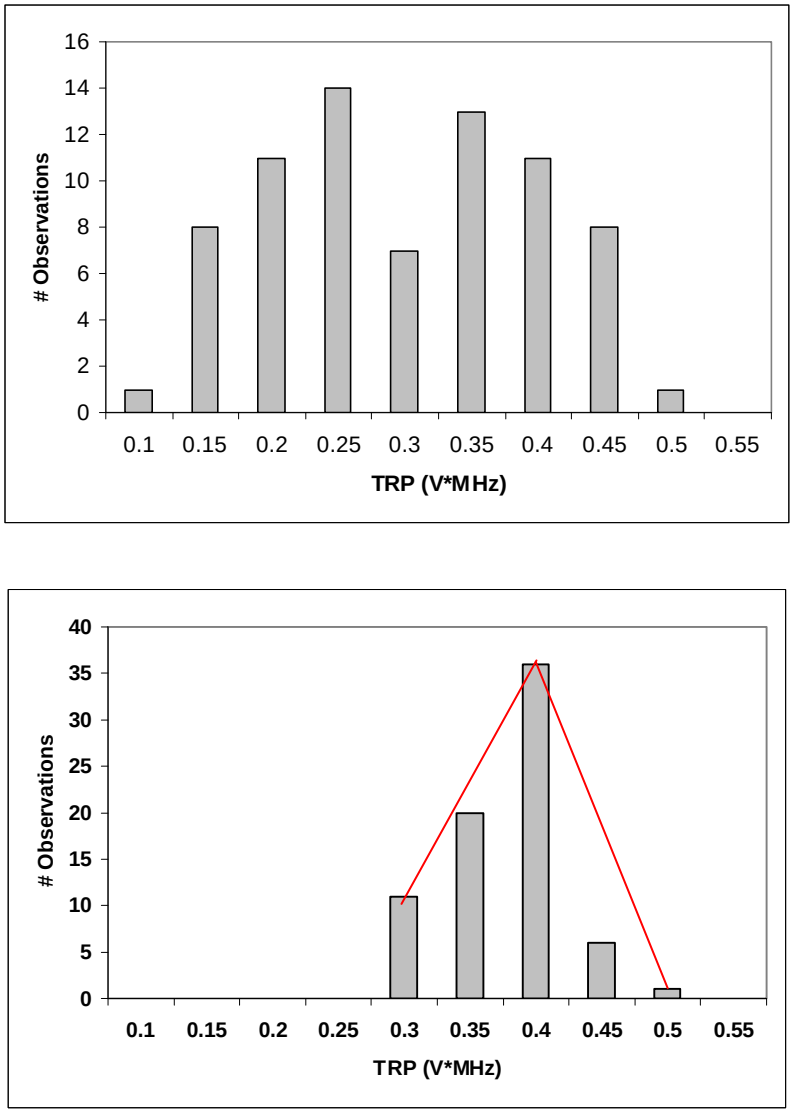

FIGURE 8. Total reflected power (TRP) distribution of protein-fouled (top), and clean (virgin) membrane coupons (bottom) showing a significant change in response to protein fouling.

SUMMARY

The studies summarized here suggest advantages of real-time, nondestructive characterization of membrane fouling. Despite the success of the ultrasonic reflectometry approach in detecting the presence or absence of a fouling layer, it is clear that more specific information regarding the onset, chemical nature and thickness of fouling layers would be of great value in optimizing module operation including the evaluation of particular cleaning strategies. We believe that recent advances in sensor technology can now be utilized to provide next-generation sensors that will lead to "smart" membranes. Similar approaches will undoubtedly be extended to other filter media including nonwovens. Successful development of such sensors will enable the more cost-effective use of filter media in an even wider range of commercial separation applications.

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