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## Online monitoring of biofouling using coaxial stub resonator technique

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#### ABSTRACT

Here we demonstrate the proof-of-principle that a coaxial stub resonator can be used to detect early stages of biofilm formation. After promising field tests using a stub resonator with a stainless steel inner conductor as sensitive element, the sensitivity of the system was improved by using a resonator of shorter physical length, implying higher resonance frequencies (and by that a higher frequency range of operation) and improved sensitivity towards dispersion. In addition, the space between inner and outer conductor was filled up with glass beads, thereby exploiting the larger surface area available for biofilm formation.

Analysis of the biofilm and the stub resonator signal, both as function of time, indicates that the sensor allows detection of early stages of biofilm formation. In addition, the sensor signal clearly discriminates between the first stages of biofilm formation (characterized by separated, individual spots of bacterial growth on the glass beads) and the presence of a nearly homogeneous biofilm later on in time. Model simulations based on the transmission line theory predict a shift of the sensor response in the same direction and order of magnitude as observed in the biofouling experiments, thereby confirming the operating principle of the sensor.

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#### 1. Introduction

Biofouling, i.e., the colonisation of an interface by a diverse array of organisms, affects surfaces and by that may have detrimental effects on the operation of processes in the field of water technology such as, raw water pre-treatment, drinking water production and distribution, wastewater treatment, industrial water cooling and water quality analysis [1-6].

Because of the high impact of biofouling on process operation and by implication high economic cost, in recent years there has been an increasing interest in developing an on – line sensor able to monitor biofilm formation in real time, especially in an early stage [7–9]. Despite all the efforts to engineer such a sensor, discussed in detail in [10–16], reliable detection technology for (the onset of) biofouling is still lacking. Existing technologies rely on pressure drop changes [14,17], differential heat transfer [10,19] or differential turbidity [20]. Actually, none of these methods can reliably detect biofouling in an early stage. Changes are detected when it is already too late and the system operation already suffers from serious impairment.

Of all the different detection technologies to track biofouling, actuators that are either acoustic [21], optical [22] or electromagnetic [23] in nature are most reliable and most sensitive [10,24]. A drawback of all these devices is however that the actual detector required is rather expensive whether that is e.g., an optical sensor [25], an analyser for scattering (S) parameters [18] or an impedance analyser [26].

The motivation to develop a new type of biofouling sensor was based, firstly, on the realization that we really need the detection of biofouling in a much earlier stage than currently available and, secondly, to offer a more cost effective alternative for existing technology. In the present study we demonstrate the feasibility of a (flow-through) coaxial stub resonator as a sensitive element of a biofouling sensor. Such resonator systems and their amplitude-frequency or AF response has been characterized, simulated and reported by the authors previously [27–31]. We discuss two different designs of such resonators. The first one has an inner





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and outer conductor separated by a fluid. The formation of a biofilm on the surface of the inner conductor (and on the surface of the outer conductor but to a much lesser extent) affects the skin effect of the inner conductor as well as the dielectric between inner and outer conductors thereby changing the AF response of the resonator. In the second type of resonator, the space in between both conductors is filled up with glass beads (Fig. 1).

The changes in AF response are caused by both the formation of a biofilm on the surface of the glass beads and the reaction of the inner conductor surface to the amount of nutrients in the feed stream. In this case, the response is more related to an (apparent) change in composition of the feed solution. The sensor geometry and dimensions were designed such that the sensor can be operated at flow conditions that are relevant for process operation in industrial equipment and piping and that the required electronics equipment can be produced in a cost effective way. Additional advantages of our sensor compared to currently existing ones are that it operates as an early warning system and is low in maintenance.

#### 2. Materials and methods

#### 2.1. Sensor description

Fig. 1 shows a schematic outline and the basic elements of a sensor based on a stub resonator coaxial transmission line, discussed in detail previously [27–31].

The resonator itself consists of an inner and outer conductor separated by a fluid of certain dielectric permittivity. A change in this (effective) permittivity of the fluid, e.g., due to a change in fluid composition, will alter the resonator characteristics. Formation of a biofilm on the surface of the inner and/or outer conductor will also change the behaviour (i.e., resonant frequency and quality factor) of the resonator. In general, the system is more sensitive to changes at the surface of the inner conductor than of the outer conductor. Obviously, the larger the surface area covered with biofilm mass, the higher the volume fraction of biofilm dielectric between inner and outer conductor. As explained in a previous study [28], an inner conductor of larger diameter will however not result in a more sensitive sensor, an effect due to stronger converging electric field lines near an inner conductor of smaller diameter. There is however a way to enlarge the effective surface area without compromising the sensor's sensitivity. Surface area enhancement can also be accomplished by filling up the space in between both conductors e.g., with glass beads (see Fig. 1). A schematic cross section of such system is shown in Fig. 3. The formation of a biofilm on the surface of the glass beads (in red) introduces a dielectric permittivity that differs from the permittivity of the glass and the fluid. As a result, the resonant frequency and quality factor (amplitude ratio) shift upon biofouling of the glass beads surface.

## 2.2. The dielectric properties of a coaxial resonator filled with glass beads covered with a biofilm

The effective dielectric permittivity  $\varepsilon_{eff}$  and the effective loss tangent tan  $\delta_{eff}$  of a coaxial resonator with multiple concentric lay-



**Fig. 1.** Schematic 3D image of the coax sensor with a length of 30 cm and an outer conductor diameter of 25.4 mm filled with glass beads. Also indicated are the input and output ports used for fluid flow-through.

ers of different dielectric permittivity has been described in [33,34] and is expressed by:

$$\mathcal{E}_{eff} = f(\mathcal{E}_{r1}, \mathcal{E}_{r2}, \dots, \mathcal{E}_{rm}) \tag{1}$$

$$\tan \delta_{eff} = f(\tan \delta_1, \tan \delta_2, \dots, \tan \delta_n) \tag{2}$$

In an ideal resonator without any losses, the resonance frequency  $f_{res}$  of an open ended ( $\lambda/4$ ) and closed end ( $\lambda/2$ ) resonator are given by Eqs. (3a) and (3b), respectively. In this special case, the dielectric constant  $\varepsilon_{re}$  can be determined directly from Eqs. (3a) and (3b) [27,28].

$$f_{res} = \frac{2n-1}{2 \cdot \pi \cdot \sqrt{LC}} = \frac{c \cdot (2n-1)}{4l \sqrt{\varepsilon_{re} \varepsilon_0 \mu_{re} \mu_0}}$$
(3a)

$$f_{res} = \frac{n}{2 \cdot \pi \cdot \sqrt{LC}} = \frac{c \cdot n}{2l\sqrt{\varepsilon_{re}\varepsilon_0 \mu_{re}\mu_0}}$$
(3b)

where *c* represents the speed of light in vacuum (m/s), *n* the order number of  $f_{res}$  (Hz), *l* the length of the resonator (m),  $\mu_r$  relative magnetic permeability of the dielectric between inner and outer conductors (–),  $\mu_0$  the absolute vacuum permeability (H/m),  $\varepsilon_0$  the absolute vacuum permittivity (F/m) and  $\varepsilon_{re}$  the real part of the relative effective dielectric constant. Note that the capacitance *C* in Eqs. (3a) and (3b) is determined by the real part  $\varepsilon_{re}$  of  $\varepsilon_r$ .

For a lossy resonator, polarization and conductivity losses in the dielectric under investigation, as well as resistance losses in the inner and outer conductors, must be taken into account. A detailed model accounting for these losses, essentially based on telegrapher's equations, is explained in [35].

In order to describe the behaviour of the biofouling sensor i.e., a lossy resonator packed with glass beads on which a film of biofouling can grow, the model described in [35] was extended with expressions for both the effective dielectric permittivity  $\varepsilon_r$  and the effective conductivity of the composite dielectric consisting of glass beads with biofilm, immersed in a feed substrate.

For a lossy dielectric, complex dielectric permittivity can be described as:

$$\varepsilon_r = \varepsilon_{re} - j\varepsilon_{im} \tag{4}$$

where  $\varepsilon_{re}$  and  $\varepsilon_{im}$  represent the real and imaginary parts of  $\varepsilon_{rr}$ , respectively.

The effective loss tangent tan  $\delta_{eff}(-)$ , which is a measure for the dielectric losses in the system, is expressed by Eq. (5):

$$\tan \delta_{eff} = \frac{\omega \varepsilon_{im} + \sigma_{eff}}{\omega \varepsilon_{re}}$$
(5)

where  $\varepsilon_{im}$  and  $\sigma_{eff}$  reflect the polarization losses and the conductivity losses in the dielectric, respectively, and  $\omega = 2\pi f$  the angular frequency, in rad/s.

In the following, two models for the effective dielectric permittivity of the composite dielectric will be discussed.

For this, we consider the coaxial resonator packed with glass beads with dielectric permittivity  $\varepsilon_{gb}$  and volume fraction  $\varphi_{gb}$  [–] (see Fig. 3). The glass beads are covered with a biofouling layer with dielectric permittivity  $\varepsilon_l$  and volume fraction  $\varphi_l$ . The free space in between the beads is occupied by feed substrate with dielectric permittivity  $\varepsilon_m$  and volume fraction  $\varphi_m$ .

The first model is known as Lichtenecker's logarithmic law and is based on the assumption that the individual components in the mixture are randomly distributed over the total volume of that mixture [34].

According to Lichtenecker's logarithmic law the effective permittivity  $\varepsilon_{ceff}$  of the (composite) space between inner and outer conductor is given by:

$$\log \varepsilon_{ceff} = \sum_{i=1}^{n} \varphi_i \cdot \log \varepsilon_i \tag{6}$$

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