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Tangential electrokinetic characterization of hollow fiber membranes: Effects of external solution on cell electric conductance and streaming current

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ABSTRACT

The methodology of tangential electrokinetic measurements with hollow fiber (HF) membranes was investigated. Two cell configurations were used, one configuration in which HF were immersed in the measuring solution and another one in which HF were embedded in a gel. The experiments were conducted with hydrophilic polysulfone HF of different lumen cross sections but of the same cross section of porous body in order to evaluate the contributions of porous fiber body and its lumen to both streaming current and cell electric conductance. The good linearity of streaming current/potential data vs. pressure difference gives evidence that expressions of the streaming current/potential derived in laminar flow are still valid for turbulent flux conditions (at least for experimental conditions used in the present study). Significant differences in both electric conductances and streaming current coefficients were obtained between the two cell configurations. The differences in the electric conductances show that the solution around fibers makes contribution to the cell electric conductance (as demonstrated previously). The observed differences in the streaming current coefficients could be due to either the contribution of the external surface of fibers to the measured streaming current or a non-negligible Starling recirculation flow. For tests conducted with HF embedded in the gel, linear variations of streaming current coefficient and cell electric conductance on channel cross section were obtained, as originally demonstrated by Yaroshchuk and Luxbacher for channels with porous walls (A.E. Yaroshchuk, T. Luxbacher, Interpretation of electrokinetic measurements with porous films: role of electric conductance and streaming current within porous structure, Langmuir, vol. 26, 2010, pp. 10882–10889). Surprisingly, it is found that the (negative) zeta potential of pore surface is higher (in absolute value) than that of the lumen surface. The contribution of the fiber body to both streaming current and cell electric conductance turns out to be considerable. It is shown that neglecting this additional path (i.e. the fiber body) for streaming current leads to an overestimation (in absolute value) of the zeta potential by a factor of 1.7–5.7 depending on the fiber. This is a clear indication that the contribution of the fiber body to the streaming current cannot be neglected in the case of standard HF ultra-filtration membranes otherwise the interpretation of their electrokinetic properties could be distorted. With such materials, it is therefore recommended not to convert the measurement of the streaming current into zeta potential of lumen surface.

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1. Introduction

Assessing the zeta potential of membranes is particularly attractive because this parameter is very sensitive to any change in surface charge properties. It can therefore serve as a probe in studies dealing with rejection mechanisms of charged solutes [1–3], membrane fouling [4–6], membrane ageing and cleaning [7–9], membrane functionalization [10,11], etc. In the case of fine-porous and asymmetric/composite membranes used in pressure-driven processes, the zeta potential is determined from the measurement of the tangential streaming potential and/or streaming current. In the case of flat membranes, the tangential mode consists in applying a pressure gradient along a slit channel formed by two identical membranes facing each other and measuring the resulting electric potential difference or electric current with a pair of reversible electrodes located on both sides of the channel, near the ends. Yaroshchuk and Luxbacher [12] have shown that in tangential electrokinetic measurements with porous substrates soaked with electrolyte solution, the pores could make contribution not only to the cell electric conductance (as first suggested by

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Yaroshchuk and Ribitsch [13] and further proved experimentally by Fievet et al. [14] and Sbai et al. [15] with ceramic and polymer membranes, respectively) but also to the streaming current. Due to these contributions, the classical Helmholtz–Smoluchowski (H–S) relation is not applicable to the interpretation of tangential electrokinetic measurements in terms of zeta potential. An experimental procedure based on both tangential streaming current and electric conductance measurements at various channel heights was therefore proposed by Yaroshchuk and Luxbacher [12] to make a correct interpretation of experimental data. This method allows the determination of zeta potentials of both external membrane and internal pore surfaces via linear regression. In a subsequent paper, Yaroshchuk et al. [16] investigated electrokinetic phenomena in undeveloped flows both numerically and experimentally. They showed that a sub-linear dependence of streaming current coefficient on the channel height occurs for undeveloped laminar flows. Therefore, they concluded that if the measurements are performed at larger channel heights only and the data are linearly extrapolated to zero channel heights, the nonzero y-intercept could then be wrongly interpreted as being the contribution of porous structure to the measured streaming current.

Up to now, measurements of tangential streaming current or streaming potential were mainly applied to the characterization of flat sheet membranes. However, some papers report their implementation for characterising hollow fiber (HF) membranes used in hemodialysers [17,18] and in bioprocessing applications [19]. Moreover, HF became increasingly attractive for other filtration and separation applications such as municipal wastewater treatment or in food and beverage industries [20]. Recent research focuses also on HF membranes for forward osmosis applications. The scarce electrokinetic characterization of HF is probably due to the fact that the large diameter of their lumen prevents the establishment of a laminar flow and the use of standard equations (e.g. the H–S equation) derived from the Hagen–Poiseuille equation (which is used to derive the expression of the streaming current or streaming potential). However, some researchers have paid particular attention to measurements in the turbulent flow. Rutgers et al. [21] worked on this subject and found that if the electric double layer is entirely included within the viscous sublayer (or laminar sublayer) adjacent to the wall, the turbulent flow does not influence the streaming potential whereas if the electric double layer expands into the turbulent core region, the streaming potential coefficient decreases. From streaming potential measurements performed with flat plate systems and capillaries, Van Wagenen and Andrade [22] showed that the phenomenon of turbulent flow does not appreciably alter the zeta potential. Igarashi and Nishizawa [23] studied the effect of turbulent flow condition on the streaming potential measurement using Teflon, Nylon and Pyrex glass capillaries with inner diameters from 1 to 5 mm and lengths from 10 to 50 cm. They concluded that the zeta potential can be measured in the turbulent flow using the streaming potential method. In a paper dealing with the electrokinetic characterization of multi-channel tubular ceramic membranes, the linear variation of the tangential streaming potential coefficient with pressure difference was observed even in turbulent flows [24]. In a recent paper [25], we also investigated the tangential electrokinetic behavior of a bundle of in/out HF by streaming current, streaming potential and cell electric conductance under conditions for which the flux was not fully laminar on the pressure range studied. It was shown that both tangential streaming potential and streaming current varied linearly with pressure difference. It was also found that in addition to the contribution of the fiber body to the cell electric conductance, the solution around fibers also made contribution.

In this work, two cell configurations were used, one configuration in which HF were immersed in the measuring solution and another one in which HF were embedded in a gel. It will be shown that not only the cell electric conductance but also the streaming current is affected by the solution around fibers. Experiments conducted with HF of different lumen cross sections but of the same cross section of porous body enabled us to assess the contributions of lumen and pore surfaces to the measured streaming current as well as their zeta potential.

2. Theory

Most membranes acquire an electric surface charge when brought into contact with an aqueous solution. This surface charge affects the spatial distribution of ions near the surface leading to the formation of the so-called “electric double layer”. If an hydrostatic pressure gradient is applied parallel to such a charged surface, the charges in the mobile part of the electric double layer are then carried towards the low pressure side, resulting in an electrical current in the direction of flow, called the streaming current. It can be measured by placing reversible electrodes in the high- and low-pressure compartments (close to the channel edges) and by connecting them to an electrometer with a low resistance (with respect to the channel resistance) so that the conduction current in the external circuit is almost equal to the streaming current. If the charges cannot be evacuated, their accumulation at one end sets up an electric field which acts to force the charges to move in the opposite direction of the streaming current. This generates an electric current called the conduction current. When this latter equals the streaming current, a steady state is achieved and the resulting electrical potential difference between the channel ends is the streaming potential. It is measured by connecting the electrodes to an electrometer with a high resistance so that the conduction current in the external circuit is negligible.

Let us consider a set of n identical HF (Fig. 1a). Each fiber is composed of a porous body with a cross-section $S_t - S_l$ and a

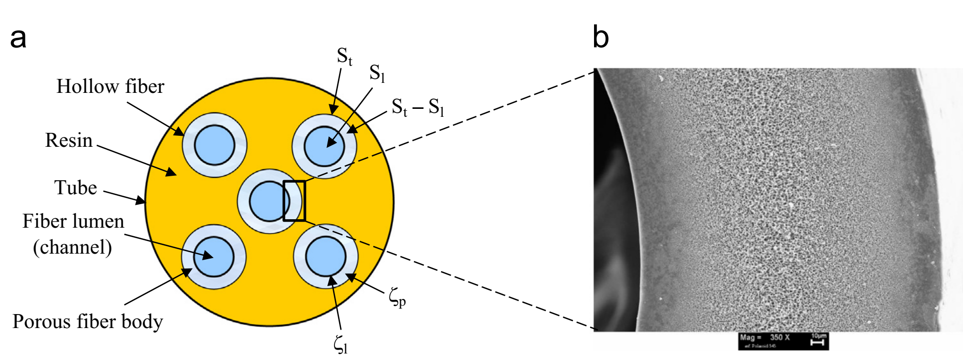

Fig. 1. Schematic cross section view of a bundle of HF (a) and SEM micrograph of the cross section view of a fiber body (b).

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