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Multi-chamber electroosmosis using textile reinforced agar membranes – A promising concept for the future of hemodialysis

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ABSTRACT

Renal replacement therapy options are limited to hemodialysis and peritoneal dialysis (70% of US patients) or renal transplantation. Diffusion processes are the main physico-chemical principle behind hemodialysis. An alternative way to achieve liquid flow through membranes bases on the electroosmotic flow which is observed as electrokinetic phenomenon in porous membranes which bear surface charges. Agar consists of the non-ionic agarose and the negatively charged agaropectine thus an electroosmotic flux is observed in analytical electrophoresis. In this study the potential electroosmosis on textile reinforced agar membranes as separation method was investigated. Using a five-chamber electrolysis cell and an agar membrane/cellulose fabric composite an intensive electroosmotic flow of 1–2 ml cm^2 h^{-1} at 100 mA cell current could be observed. The movement of cations in the negatively charged agar structure led to an intensive electroosmotic flux, which also transported uncharged molecules such as urea, glucose through the membrane. Separation of uncharged low molecular weight molecules is determined by the membrane characteristic. The transport of ions (K^+, PO_4^{3-}, creatinine) and uncharged molecules (urea, glucose) in electroosmotic separation experiments was monitored using a pH 5.5 phosphate electrolyte with the aim to assess the overall transport processes in the electrochemical cell. The results demonstrate the potential of the method for filtration of biological fluids in the absence of external pressure or high shear rates.

1. Introduction

The prevalence of chronic kidney disease in the U.S. is estimated to be approximately 14%, which corresponds to 39 million adults (Levey et al., 2009; United States Renal Data System, 2010). Patients with end stage renal disease (ESRD) and requiring renal replacement therapy (RRT) have increased from approximately 10,000 in 1973 to 615,000 in 2011 (United States Renal Data System, 2013). Renal replacement therapy options are limited to hemodialysis and peritoneal dialysis (70% of US patients) or renal transplantation (Fissell, Roy, & Davenport, 2013). Diffusion processes are the main physico-chemical principle behind hemodialysis. High flux dialyzers provide clearance rates of 70–90% of the blood flow through the capillaries (e.g. creatinine clearance of 270 ml/min at 300 ml/min of blood and 500 ml/min of dialysate flow) (Fresenius Medical Care, 2012).

Hemodialysis utilising classical separation membranes requires high volumes of dialysate (approx. 120–200 l per 4 h session) to maintain a high concentration gradient for diffusion. Additionally, as during hemodialysis excess fluid is removed from the patient, convective clearance is achieved; the magnitude of which depends on the technique used and ranges from about 10 ml/min during conventional dialysis (2.5 l during a 4 h dialysis session) up to 90 ml/min during hemodiafiltration (fluid turnover about 20 l/4 h). Amongst others this basic requirement of hemodialysis necessitates non-portable devices and significantly contributes to the annual direct treatment costs of approximately US$ 87.000 per patient (United States Renal Data System, 2010). Furthermore, despite all efforts, only the current hemodialysis is able to provide 10–15% of clearance rates achieved by the native kidneys

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and consequently the prognosis of affected individuals is massively reduced. As outlined above during dialysis solute clearance has to be achieved through transfer of low molecular weight products such as urea and creatinine from the blood into the dialysate by diffusion and to a lesser extent by convection.

Additionally, pressure is applied to achieve sufficient filtration volumes, which not only results in solute transport via convection, but also requires accurate control of volumes and solutes status in the patient, in the dialysate compartment as well as the substitution fluid used. At the same time the concentrations of ions such as calcium, hydrogen-carbonate, sodium and potassium have to be controlled, which is performed by adjusting the respective concentrations in the dialysate in response to changes in patients serum concentrations.

An alternative way to achieve liquid flow through membranes bases on the electroosmotic flow which is observed as electrokinetic phenomenon in porous membranes which bear surface charges (Li, Ghanem, & Higuchi, 1999; McLaughlin & Mathias, 1985).

When an electrical current is applied to a polymer gel which bears negatively charged groups an electroosmotic flow can be observed as a result of the movement of the mobile cations through the porous membrane structure (Polevoi, Bilova, & Shevtsov, 2003; Van Woude & Davis, 1963). In case of a polymer with carboxylic or sulphate groups a plug of liquid constantly moves through the pores of the polymer gel in direction of the cathode. Electroosmosis has been observed at number of biologic membranes (Mudd, 1925; Weaver, 2003) and has been proposed as method for various applications such as purification of biologic substances (Bier, 1972), or decontamination of soil (Asadi, Huat, Nahazanan, & Keykhah, 2013; Cameselle & Reddy, 2012; Kali, Das, & Shekhar, 2012; Lima, Kleingeld, Heister, & Loch, 2012). However, applications in real life are rare as often more simple techniques are sufficient to achieve the required separation.

The potential to achieve high membrane flux without any pressure difference combined with the ability to control ion concentrations, makes electroosmosis an appealing alternative technique to the separation methods used in current hemodialysis.

Agar consists of two main constituents, the non-ionic linear agarose which consists of repeating units of agarobiose (a disaccharide of D-galactose and L-3,6-anhydro-α-galactopyranose) and the slightly branched agaropectine which bears negative charges through sulphate and pyruvate acid side groups (Akelah, 2013). Agar gels thus represent a swollen porous structure which bears immobilised negative charges (sulphate and pyruvate side groups) and mobile positive counter ions (e.g. sodium ions). When a transport of positive ions through the gel structure is induced by an external electrical field a parallel flow of liquid, the electroosmotic flux is observed.

In this article the electroosmotic flow though mechanically more stable textile reinforced agar membranes was investigated. Agar gels were chosen as model for negatively charged gel membranes to study separation of low molecular weight substances by electroosmosis. In lab-based electroosmotic batch experiments ion separation and transport of uncharged molecules through the membrane structure were investigated to assess the potential of the technique. An electroosmotic filter could lead to the design of smaller and more efficient blood purification units (Degim et al., 2011; Kofler et al., 2013).

2. Experimental

2.1. Materials

All chemicals used were of analytical grade (KH_2PO_4, K_2HPO_4, KCl, HCl, CsCl, H_2SO_4, ammonium heptamolybdate-tetrahydrate, potassium antimony(III)oxide tartrate hydrate, glucose monohydrate, urea, L-ascorbic acid,). Creatinine (Merck, Darmstadt) and agar (M_r 3000–9000, gel strength 1.5% sol. > 300 g/cm^2, gel temp. ~35 °C, Fluka, Buchs, Switzerland) were biochemistry grade.

A bleached plain weave cotton fabric with mass per area of 129 g/m^2 was used as 100% cellulose based textile reinforcement for the agarose gel.

For preparation of the textile reinforced agar a sheet, a piece of fabric first was tailored to fit into a plastic bag, e.g. 15 cm × 20 cm. The fabric then was placed in the plastic bag and an excess of hot agar solution was added to the fabric into the bag. The excess of hot agar solution was wiped out of the bag. Thereby the height of the agar layer was reduced to the thickness of the fabric. The agar then was allowed to gel. Photographs and micrographs of the reinforced agar are shown as Supplementary material.

2.2. Electroosmotic cell

The electroosmotic cell stack was built up from plexiglass, using cylindric filter-press design (cell diameter 20 mm; 3.14 cm^2) with silicon ring sealings. Titanium expanded metal with platinum coating was used as inert cathode and anode (width 15 mm). Pictures of the five chamber electroosmosis cell are shown in Fig. 1a and b.

Electrode chambers (1, 5; vol. 6 ml) were separated from the recipient dialysate chambers (2, 4; vol. 4.25 ml) by Nafion type cation exchange membranes (M1, M2). Biological model fluid was filled into the treatment chamber (3; vol. 4.2 ml), which was separated from the recipient chambers (2, 4) by means of textile reinforced agar-membranes. Agar-membranes were prepared by coating a bleached 100% cotton fabric (126 g/m^2) with 1.5% (w/w) agar solution. For comparison also a viscose type cellulose film (Muscozell Einsiedehaut rund Konfitürenverschluß, Spar, Dornbirn, Austria 35.8 g/m^2) was mounted instead of the agar-membranes, to study a separator system with low surface charge. In the case of the viscose membrane no significant electroosmotic flow was observed.

For an electroosmotic experiment all cell chambers were filled with 50 mM pH 5.5 phosphate buffer (PB) (3.4 g KH_2PO_4 and 4.3 g K_2HPO_4 in 500 ml deionised water) or 100 mM PB. The biological model fluid in chamber 3 additionally contained low molecular weight components as representatives for solutes present in blood.

Urea, creatinine and glucose were used as model systems and their respective concentrations in the different chambers were analysed at the end of a batch.

Electrochemical experiments were performed batchwise applying a fixed voltage between 30.5 and 31.1 V using a bench type power supply (Farnell Instruments LTD, L30-1, stabilised power supply 0–30 V, 1 A). Current flow was recorded and overall electroosmotic flow was determined from the gravimetic analysis of the additional volume transported into chamber 2 of cell. The experiments were executed for a predefined time and the concentration of marker substances, potassium and phosphate, glucose, urea and creatinine were determined.

Two repetitions of the experiments were performed. Results are given as mean values and standard deviation (error bars).

2.3. Analytical procedures

Potassium analysis was performed with AAS (Analytik Jena contrAA 300, wavelength 404 nm) using KCl as calibration standard (According DIN 38 406 Teil 13, E13).

Phosphate analysis was performed by photometry of the blue phosphor-molybdato complex at 880 nm (Hitachi U-2000, Tokyo, Japan; double beam spectrophotometer, 10 mm cuvette) according to DIN EN ISO 6878, D11, using KH_2PO_4 for calibration.