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# Towards supported bolaamphiphile membranes for water filtration: Roles of lipid and substrate

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## ABSTRACT

Supported biomimetic membranes hold potential for applications such as biosensors and water purification by filtration. The current paper reports on the preparation of a supported bolaamphiphile membrane on two polymeric nanofiltration membranes: NF-270 made of polyamide with carboxylic surface charges and NTR-7450 made of sulfonated polyethersulfone with sulfonic surface charges. The results, obtained using a proprietary microfluidic device, confirmed that the supported membrane coverage on the polymeric membranes was governed by the double-layer interactions, in agreement with previous studies on silica, mica, and gold. Remarkably, the formation of the biomimetic membrane was more favorable on the sulfonated polyethersulfone than on the polyamide surface, although both surfaces exhibit a similar surface charge density. It is suggested that the higher dissociation constant of sulfonic groups was the main reason for the higher coverage of NTR-7450. Finally, spinach aquaporins, which are trans-membrane proteins that facilitate the water transport, were incorporated into a supported membrane on NTR-7450. The incorporated aquaporin resulted in enhanced pressure-driven water transport through the membrane, however, for still unclear reasons, the transport was not selective. Despite this failure, the results provide new insights into formation of biomimetic membrane on water permeable polymeric substrates, as a generic approach towards biomimetic water filters.

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## 1. Introduction

Supported biomimetic membranes (SBMs) are commonly used to study biological membranes and biosensors [1–3]. Vesicle fusion is probably the most straightforward SBM preparation technique. In this method a hydrophilic substrate is a solution containing vesicles or micelles, allowing the vesicles or the micelles to adhere to the surface and eventually rupture and self-assemble to SBM [4–6]. The preparation of SBM using vesicle fusion on flat, hydrophilic, and densely charged surfaces, such as silicon wafers, mica, and gold has been studied extensively [7–9].

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Recently, it was proposed to use SBMs as the basis for next generation membranes for water purification [2,10–15]. Previous reports showed that the osmotic permeability of vesicles with incorporated aquaporins, a group of trans-membrane proteins found in biological cell membranes, was significantly higher than that of vesicles without aquaporins [12,16]. Based on these reports it was estimated that a defect-free SBM with incorporated aquaporin (lipid to protein molar ratio (LPR) of ~50) would be at least an order of magnitude more permeable and significantly more selective than available commercial reverse osmosis membranes [2,10–14,17–19]. The exceptional performance of aquaporin-based SBM might open up new avenues for membrane-based separations, particularly for small-scale processes in microfluidic devices [20,21].

For water purification or analytical membrane-based separations, the use of mechanical pressure as a driving force may be preferred over osmotic or electric gradients that drive the transport of water and ions in most biological systems. However, the

**Table 1**The lipids, the NF membranes, and their  $\zeta$ -potentials as measured in 1 mM and 150 mM NaCl, respectively, at neutral pH 30 °C.

Lipid	Structure	$\zeta$ -Potential (mV)	
		1 mM	150 mM
GLH-20		46±3	21±1
DMTAP		61±1	30±3
DMPC		0±6	0±6
DMPA		-80±2	-35±2
Rh-PE			
Membrane	Top-layer	$\zeta$ -Potential (mV)	
		1 mM	150 mM
NF-270		-55±5	-25±10
NTR-7450		-47±5	-25±10

preparation of a macro-scale ( $\text{mm}^2$ ) defect-free SBM that withstands pressure gradients is still challenging. Herein we try to address that problem.

Previously, several researchers have tried to prepare a pressure durable SBM. Vogel et al. prepared a free-standing lipid membrane suspended within micropores [22], and Wang and coworkers employed for this purpose a substrate with  $\sim 60$  nm nanopores [23]. Neither study reported membrane permeability or selectivity

in pressure-driven mode. Instead of a planar lipid membrane, Sun et al. proposed to affix intact proteoliposomes (vesicles with incorporated membrane proteins) to modified ultra-filtration membranes, with still smaller, several nm large nanopores [24]. Finally, Zhao et al. embedded aquaporin-laden proteoliposomes within a conventional dense polyamide layer of a reverse osmosis membrane prepared by interfacial polymerization [14]. In the latter two studies the membranes could filter aqueous solutions

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