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Mitigating biofouling on thin-film composite polyamide membranes using a controlled-release platform

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ABSTRACT

Biofouling remains a major challenge for membrane processes. Several methods may curtail biofouling, including membrane surface modification with hydrophilic and antimicrobial polymers and nanomaterials. Although many of these modifications rely on compounds that leach from the membrane surface, the release of these compounds is not always characterized, understood, or controlled. Here, we adapt a technology used for drug delivery — controlled release — and apply it to the membrane biofouling problem. Capsules for controlled release can be designed using a number of polymers, and the loading and release rate of the capsules depends on a number of tunable variables. We have encapsulated two antimicrobial compounds, cinnamaldehyde and kanamycin, in biodegradable poly(lactic-*co*-glycolic acid), PLGA, particles. These capsules were then bound to the surface of thin-film composite polyamide membranes, targeting biofouling where it is most problematic. Cinnamaldehyde released from the capsules for ~2 days. Kanamycin, encapsulated in larger PLGA particles, showed continuous release over the 80 h period tested. Biofilm formation on modified membranes was assessed using model bacteria (*Escherichia coli*), and significant reductions in biofilm were observed on membranes modified with kanamycin capsules, indicating that sufficient kanamycin was released to curtail bacterial growth and biofilm development. This proof of concept study demonstrates that the controlled-release platform can be used to encapsulate a variety of compounds to slow membrane biofouling.

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1. Introduction

Biofouling, despite decades of research, remains one of the greatest challenges in membrane separations for water treatment and desalination [1–3]. Strong oxidizers (e.g. chlorine or ozone) are commonly used to curtail biofilm growth in several forms of water treatment technologies. However, in many membrane processes, including reverse osmosis (RO), forward osmosis (FO), and pressure retarded osmosis (PRO), these oxidizers cannot be used, as they significantly degrade the polyamide separation layer on thin-film composite (TFC) membranes. Therefore, alternative methods, such as control of operational conditions and modifications to membranes or membrane spacers, are sought to mitigate biofilm formation.

While many of the methods proposed for biofouling mitigation offer robust modifications to the membrane or spacer surface (e.g. chemicals, nanoparticles, or even enzymes) [4–7], some modifications act through the release of active compounds [8,9]. For example, it has been proposed that a membrane modified with silver nanoparticles acts through the release of silver ions. However, many modifications are tested only over a short period of time, and the duration of the modification and potential release of active compounds is not always well-characterized. When considering the true potential of antifouling modifications, it is important to know both their mode of action and their longevity. If a modification acts through the release of a particular compound, it is essential that this release be characterized, understood, and controlled. This is particularly critical if a modification is to be applied beyond the laboratory scale, as membrane modules may be operational for months between chemical cleaning intervals [10].

The diverse field of controlled release has developed over several decades in the medical community [11,12], and it may provide a useful tool for combating membrane biofouling for several reasons. Specifically, capsules containing targeted, active compounds can be designed to deliver a certain quantity of a compound at an engineered rate over a predetermined period of time (up to a month or more) [13]. Furthermore, capsules can be designed to respond to stimuli, for example pH, allowing them to only release targeted compounds when it is beneficial for the system, thereby conserving the active agents [14,15]. Additionally, capsules of similar formulations may house a variety of different active compounds, allowing for easy screening of agents for biofilm mitigation. Lastly, biodegradable capsules [13,16] may be fabricated with polymers that dissolve over time, allowing the targeted area to be reloaded with fresh particles.

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Here, we adapt a platform from the drug delivery field and apply it to combat membrane biofouling. We fabricated biodegradable poly(lactic-*co*-glycolic acid), PLGA, capsules with two very different compounds — hydrophilic kanamycin (Kan) and hydrophobic cinnamaldehyde (CA). These capsules release Kan and CA in a controlled manner, and the particular release kinetics can be tuned depending on the needs of a particular system. Although we have chosen two specific compounds to encapsulate, a wide variety of compounds, including quorum sensing inhibitors [17,18], could be incorporated. In addition to capsule fabrication, characterization, and binding to the surface of thin-film composite polyamide membranes, we explore the effect of this modification on membrane surface and transport properties, and we assess biofouling on the modified membranes. Finally, we discuss the potential applications and challenges of using this controlled-release platform for membrane biofouling redction.

2. Materials and methods

2.1. Bacteria and growth conditions

Escherichia coli MG1655 was obtained from the Yale Coli Genetic Stock Center (CGSC ♯7040) and maintained on Luria–Bertani (LB, American Bioanalytical) agar plates. Prior to experiments, a single colony of bacteria was placed in 25 mL sterile LB broth and grown overnight, shaking at 190 rpm, at 37 °C. To obtain a log-phase culture, 1 mL of the overnight culture was used to inoculate 24 mL fresh LB broth. The culture was then grown for 2 h at 37 °C shaking at 190 rpm to log-phase. Log-phase growth time was determined by a growth curve. Cell density was determined by a plate count. Optical density (OD) was measured at 600 nm (SPECTRA max 340PC) to determine cell growth. An OD_{600} of 1.0 corresponds to 10^9 CFU/mL.

2.2. PLGA capsule fabrication

Poly(D,L-lactide-*co*-glycolide) (PLGA) (85:15, M_w=50,000–75,000 Da, Sigma-Aldrich) capsules were fabricated using an emulsion–diffusion–evaporation method [19]. The hydrophobic PLGA core was stabilized with a layer of poly(vinyl alcohol) (PVA, 80 mol% hydrolyzed, M_w~6000 Da, Polysciences, Inc.) (Fig. 1a–d). We included 1 wt% poly(ethylene-*alt*-maleic anhydride) (PEMA, M_w 100,000–500,000, Sigma-Aldrich) (relative to PVA) in the aqueous phase to provide carboxyl groups necessary for particle binding [20]. A stock solution of poly(ethylene-*alt*-maleic acid) was made by stirring 1% poly(ethylene-*alt*-maleic anhydride) in deionized (DI) water (Milli-Q ultrapure water purification system, Millipore, Billerica, MA) for 3 days. For each capsule containing an active compound, i.e. CA-c and Kan-c (Fig. 1b and d), a blank capsule, i.e. sBl-c and dBl-c (Fig. 1a and c), was fabricated.

A single emulsion was used to encapsulate *trans*-cinnamaldehyde (CA, > 99%, Sigma-Aldrich), CA-c (Fig. 1b). First, an oil phase was made by dissolving 312.5 mg of PLGA in 4.75 mL acetone (ACS grade, J.T. Baker) and 3.125 mL ethanol (EtOH, 200 proof, anhydrous, PHARMCO-AAPER). The aqueous phase was made by dissolving 1.875 g PVA and 1.875 mL 1 wt% PEMA in 45 mL DI water. The oil phase was then added drop-wise to the aqueous phase under stirring. The emulsion formed spontaneously, as ethanol, which has less affinity for PLGA than acetone, exited the oil droplets and entered the water phase. The emulsion was left stirring for 4 h at room temperature. Capsules were then washed three times with DI water at 10,000*g* for 30 min (Thermo Scientific SorVALL RC 6+ Centrifuge).

Particles encapsulating hydrophilic kanamycin were made with a water–oil–water ($W_1/O/W_2$) double emulsion. First, W_1 was made by dissolving 50 mg/mL kanamycin sulfate (American Bioanalytical) in DI water. After dissolution, 3.3 mg of PVA was added and dissolved in 100 μL kanamycin sulfate solution. The oil (O) phase was made by dissolving 150 mg PLGA in 6 mL chloroform (> 99.9%, Sigma-Aldrich). 60 mL PVA (1.5 g/L) and 90 μL of 1 wt% PEMA were combined to form W_2. In order to make the first emulsion, W_1 was added to O, vortexed on the highest setting (Mini Vortexer VWR Scientific Products) for 20 s and bath sonicated (Aquasonic Model 150 T, VWR Scientific Products) for 30 s. During homogenization at 17,500 rpm (VWR VDE25), W_1/O was added dropwise to W_2 over a period of 3 min. The chloroform was removed with a rotary evaporator (Büchi Rotovapor R) for 15 min, and the capsules were washed 3 times using centrifugation (5000*g* for 5 min).

2.3. PLGA capsule characterization

Capsule size was determined by dynamic light scattering (DLS, ALV-GmbH, Germany). Particle suspensions were diluted to low concentrations using DI water and vortexed immediately before measurement to minimize sample settling. Measurements for each particle type were taken every 30 s for 20 cycles, with a scattering angle of 90° and an incident wavelength of 523 nm. For each type of particle, the correlation functions obtained from these measurements were analyzed in MATLAB and Fortran using the CONTIN algorithm [21,22] and then averaged into a single data set. The

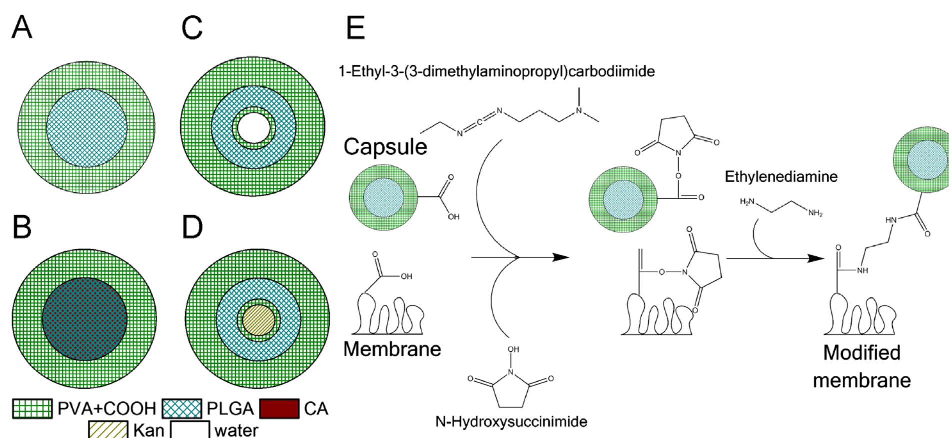

Fig. 1. Overview of capsules and binding process. Four different types of capsules were fabricated: (a) single emulsion blank capsules, sBl-c, (b) cinnamaldehyde-loaded single emulsion capsules, CA-c, (c) double emulsion blank capsules, dBl-c, and (d) kanamycin-loaded double emulsion capsules, Kan-c. To bind capsules to the membrane (e), carboxylic groups on both the particles and the polyamide active layer on the membrane surface were activated into amine-reactive esters through contact with an aqueous solution of EDC and NHS. ED was then used to form amide bonds on the membrane surface available for linking with the activated capsules.

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