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Self-assembled polysaccharide-based multilayer nanofilms of xanthan gum and diethylaminoethyl dextran on gold substrate and their interaction with model biomacromolecules

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INTRODUCTION & AIM

Layer-by-layer (LbL) self-assembled multilayer nanofilms comprised of oppositely charged polysaccharides have emerged as potential candidates in the fields of biomedical and food sciences due to their physicochemical and biocompatible properties. The LbL technique itself, which is driven primarily by electrostatic attraction and secondarily by weaker supramolecular forces, allows the precise engineering of nanoscale architectures that can emulate and interact with the structural complexity of the extracellular matrix. In the present study, the formation and characterization of xanthan gum (XG) and diethylaminoethyl dextran (DD) nanofilms on gold (Au) substrates under neutral pH conditions are investigated. The nanofilms are tested for their stability against increased ionic strength, pH change, as well as for their interactions with bovine serum albumin (BSA) and porcine gastric mucin (PGM). These self-assembled nanofilms hold significant promise for applications as drug delivery systems, wound healing scaffolds, and biosensors.

MATERIALS & METHODS

Materials and Sample Preparation: XG was purchased from CP Kelco, while DD, PGM, BSA, citric acid (CA) and sodium chloride (NaCl) were purchased from Sigma-Aldrich. Stock solutions of XG, DD, PGM and BSA of 0.2 mg/mL and of NaCl of 0.15 M were prepared using water for injection. CA was used to decrease the pH from 7 to 2.5.

Protocols: XG/DD nanofilms were prepared on Au substrates using the LbL method under neutral pH conditions. The resulting nanofilms consisted of 10 alternatingly deposited monolayers (5 bilayers) of the oppositely charged biopolymers, contractually starting and ending with a DD and a XG monolayer, respectively. Stability against NaCl 0.15 M solutions and acidic environments and interaction experiments with BSA and PGM were performed after the formation of the respective XG/DD nanofilms.

Surface Plasmon Resonance (SPR): A custom-made SPR apparatus based on the Kretschmann configuration was used. Au layers were formed on the flat surfaces of SF11 equilateral prisms. A He-Ne laser beam with a wavelength of 632.8 nm was employed as the light source for SPR. Solutions were placed in a PTFE cell sealed onto the Au-coated surface of the prism.

Quartz Crystal Microbalance with Dissipation (QCM-D): An openQCM Q-1 device from OpenQCM equipped with an Au-coated quartz crystal was utilized for conducting QCM-D experiments. The resonators possessed a fundamental frequency of 10 MHz and a mass sensitivity constant of 4.42 ng·cm^{-2}·Hz^{-1} for high-precision frequency measurements. The sensors were mounted in a fluid cell with one side exposed to solution.

RESULTS

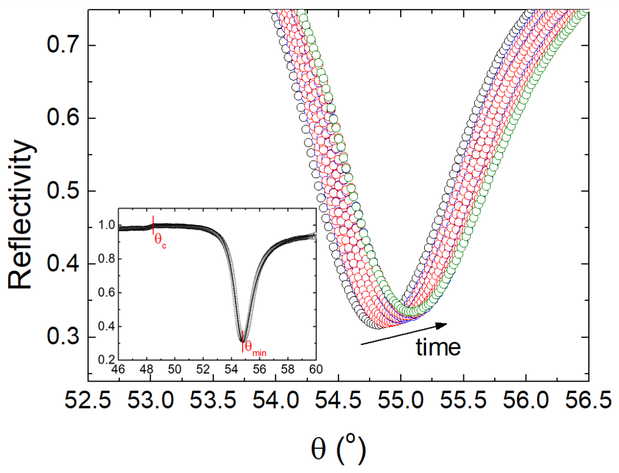

Fig. 1: Characteristic SPR curves from the XG (red) and DD (blue) layers and the resulting multilayer nanofilm (olive) in contact with Au substrate (black). The arrow indicates time evolution of the XG/DD multilayer. Inset: Full-scale experimental SPR curve from the Au/water interface (continuous line is the fitting profile).

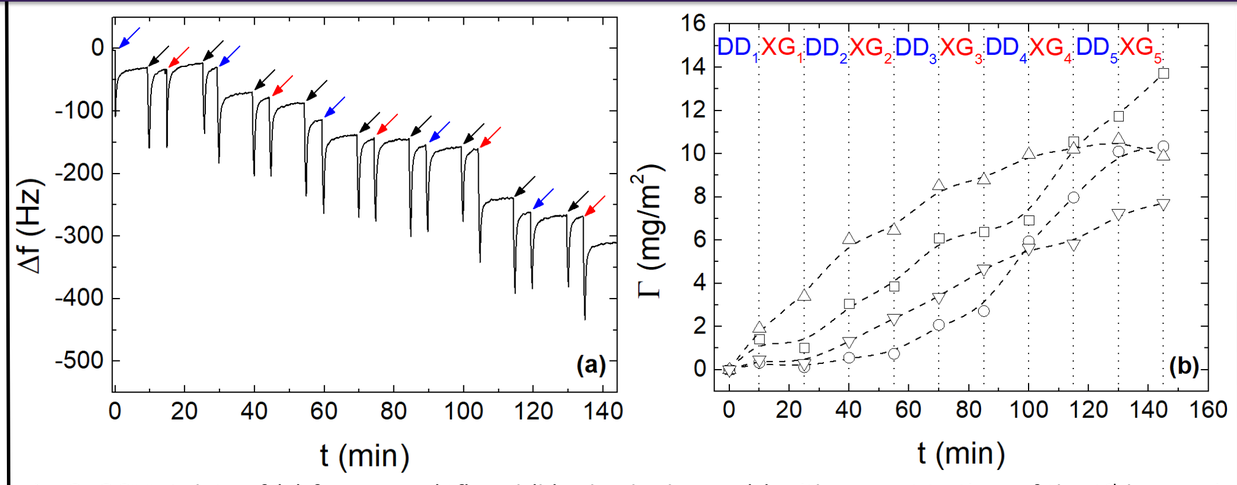

Fig. 2: QCM-D data of (a) frequency (Δf) and (b) adsorbed mass (Γ) with respect to time of the 1st harmonic of the sensor across different preparations (different symbols) were obtained using the Sauerbrey model. Arrows in graph (a) indicate the introduction of DD (blue) and XG (red), as well as the intermediate water rinsing steps (black). Sequential adsorption of the oppositely charged polysaccharides in graph (b) is separated by vertically dotted lines and is denoted by labels DD_k and XG_k for the k^{th} layer of DD and XG, respectively. The dashed lines connecting the Γ values in graph (b) are used solely as guides to the eye.

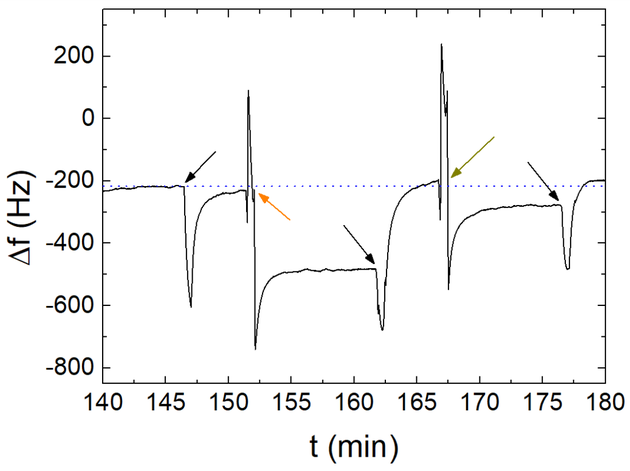

Fig. 3: Representative QCD-D data of frequency (Δf) in relation to time for XG/DD nanofilms after their preparation interacting with saline environments. Arrows indicate rinsing with water at pH 7 (black), NaCl 0.15 M at pH 7 (orange) and 2.5 (dark yellow). The dotted lines represent the initial Δf values prior to saline exposure.

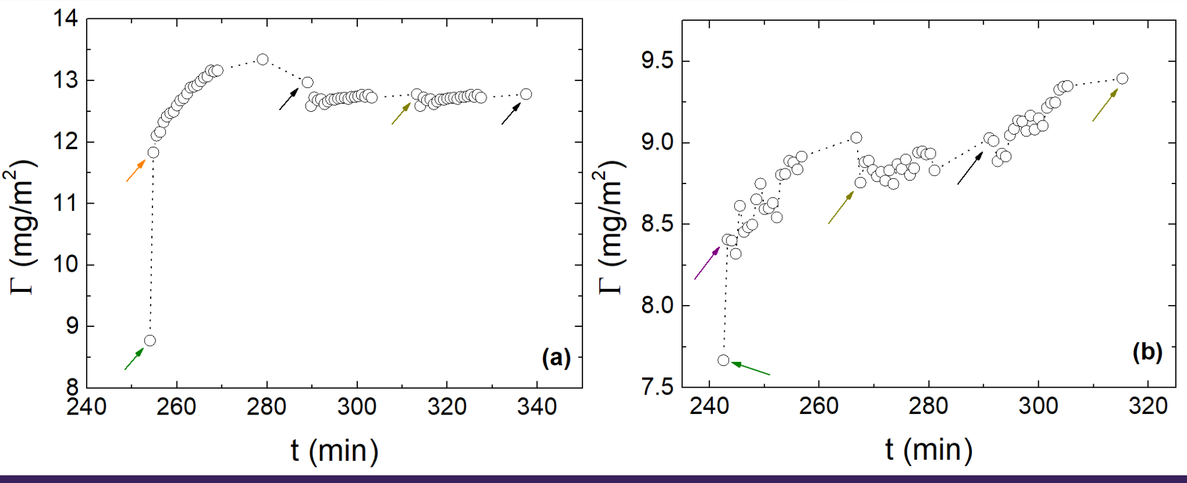

Fig. 4: Adsorption kinetics from SPR measurements during the interaction of BSA with XG/DD nanofilms at pH (a) 7 and (b) 2.5. Arrows indicate the introduction of BSA at pH 7 (orange) and 2.5 (purple), as well as the prepared XG/DD nanofilm (olive) and the intermediate water rinsing steps at pH 7 (black) and 2.5 (dark yellow) for stability. The dotted lines are guides to the eye.

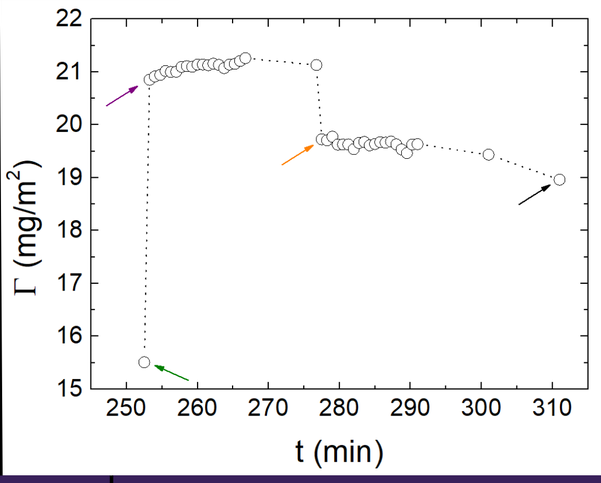

Fig. 5: Adsorption kinetics from SPR measurements during the interaction of PGM with XG/DD nanofilms at pH 7. Arrows indicate the introduction of PGM at pH 7 (purple) and NaCl 0.15 M at pH 7 (orange), as well as the prepared XG/DD nanofilm (olive) and the intermediate water rinsing steps at pH 7 (black) for stability. The dotted lines are used solely as guides to the eye.

CONCLUSIONS

- Electrostatic self-assembly of XG/DD nanofilms comprised of XG and DD on Au substrates was achieved under neutral pH conditions.
- XG/DD nanofilms remained stable in neutral, acidic, and physiological ionic environments.
- pH-Dependence of interactions between BSA and XG/DD nanofilms allowed tunable and stable binding of BSA across neutral and acidic conditions.
- Mucoadhesive properties of XG/DD nanofilms enabled PGM adsorption at neutral and release at physiological conditions.
- XG/DD nanofilms exhibited tunable characteristics which render them promising candidates for biomedical, biotechnological and food industry uses.

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