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Ultrathin Polydopamine Films with Phospholipid Nanodiscs Containing a Glycophorin A Domain

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Introduction

Inspired by mussels adhesive proteins, polydopamine (PDA) films gained attention in the last years because of their adhesive properties on virtually any substrate and the presence of surface hydroxyl groups from the catechol moieties but also amine and imine groups [1], which make them relatively easy to functionalize. Electropolymerization has been proved to be an effective technique to control dopamine polymerization on a conductive substrate, giving the possibility to form compact film with very controlled thickness, high reproducibility and long term stability [2]. Together with their reducing and pH responsive features, Polydopamine films are suitable candidates for the development of biocompatible nanocomposite films with catalytic properties. Embedding nanomaterials and tuning film shapes are only some of the advantages given by electropolymerization that together with the active properties of polydopamine lead to the possibility of a new approach in the production of nanocomposite materials. Phospholipids nanodiscs are a class nano entities that retain the structural features of cell membranes allowing the incorporation of transport proteins and receptors [3]. The implementation of such nanomaterial into the PDA film would allow applications ranging from functional coatings for bioelectronics devices to mimicking of cell membranes.

Film preparation and Nanodiscs

Highly controlled deposition of polydopamine can be achieved by cyclic voltammetry. The potential is changed cyclically to alternate oxidation on the working electrode and reduction on the counter one leading to the deposition of polymerized dopamine on the surface of the working electrode (**a**,**b**).

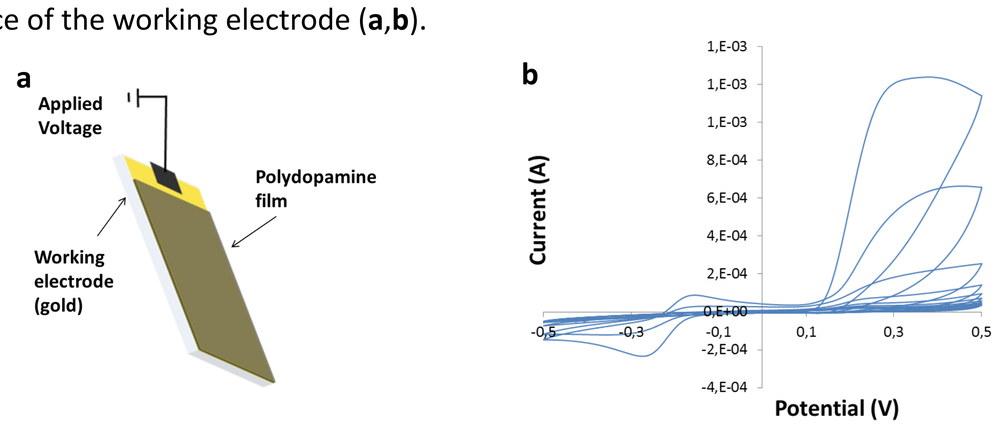

Figure1: a. Scheme of polydopamine film formation at gold working electrode b. Cyclic voltammogram polydopamine.

Polydopamine-nanodiscs hybrid films

Phospholipids nanodiscs with glycophorin domain were assembled and deposited on the gold substrate by drop casting, prior surface functionalization using DTSSP to bind them to the electrode. The electropolymerization process was then applied to embed them into the PDA matrix (Figure 3).

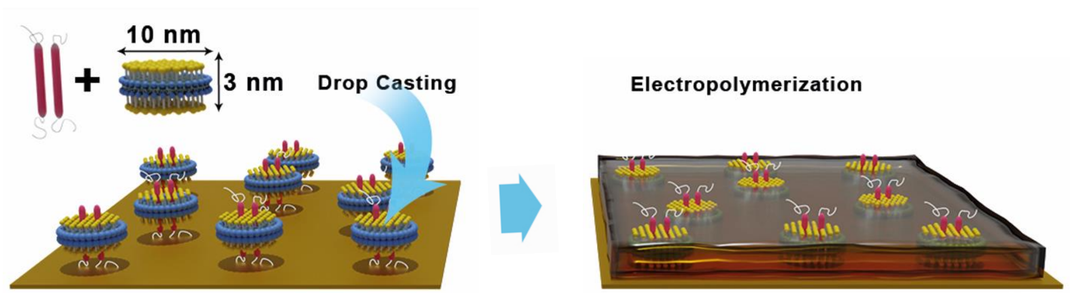

Figure 3: Scheme of deposition and embedding of nanodiscs into polydopamine matrix.

The hybrid film was characterized using AFM-IR to prove the successful embedding of the nanodiscs both chemically and mechanically as shown in figure 4 [4].

Electrochemical characterization

PDA-Nanodiscs hybrid films were characterized electrochemically running CV using two different redox probe, positively and negatively charged (Figure 5). Proving the nanodiscs were available on the surface of the film by selectively increasing the positive charges transport (Figure 6 a and b).

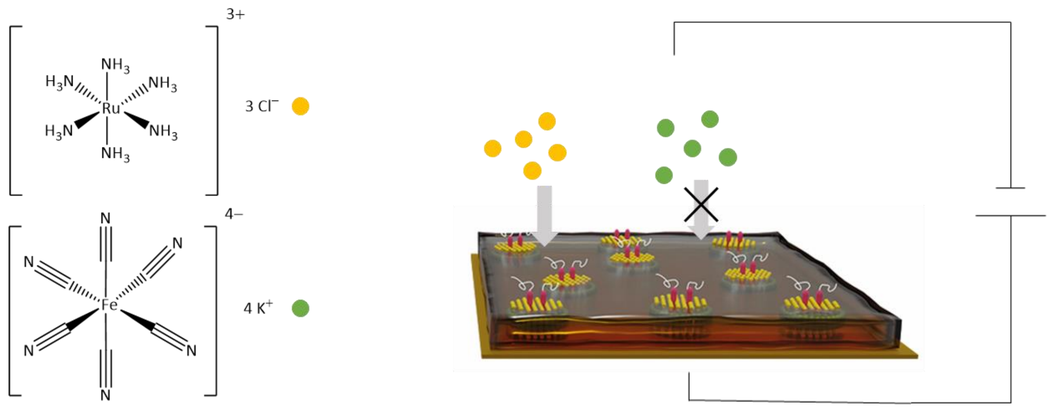

Figure 5: Scheme of the electrochemical characterization of the hybrid film. On the left the two redox probes: $Ru(NH_3)_6{\cdot}Cl_3$ in yellow , $K_3[Fe(CN)_6]/K_4[Fe(CN)_6]$ in green. On the right scheme showing the permeation of the positively charged ions while the retention of the negatively charged ones.

Electropolymerized polydopamine films are smooth and homogeneous as shown by AFM (Figure 2 a) with an average surface roughness of ~1,3 nm. The thickness for a 5 cycles polydopamine film is between 8 and 10 nm as calculated by AFM . In addition AFM image of phospholipid nanodiscs deposited on mica (Figure 2 b) showing ca 3 nm in height and 10 nm diameter.

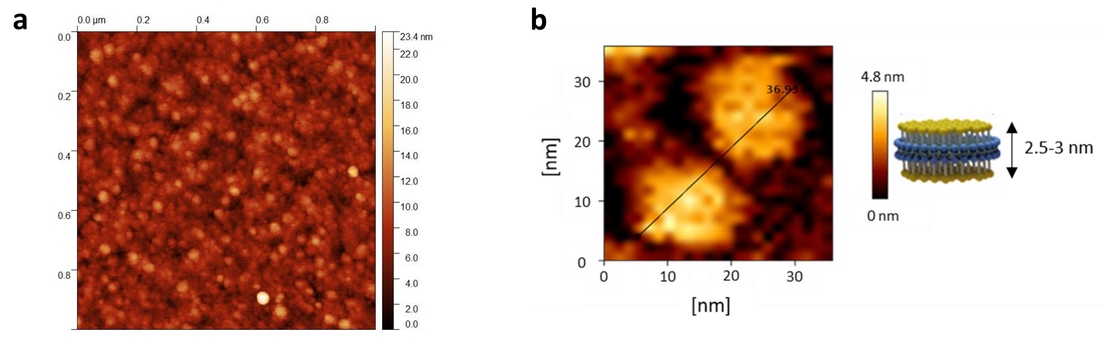

Figure2: a. AFM micrograph polydopamine film on gold b. AFM nanodiscs deposited on Mica .

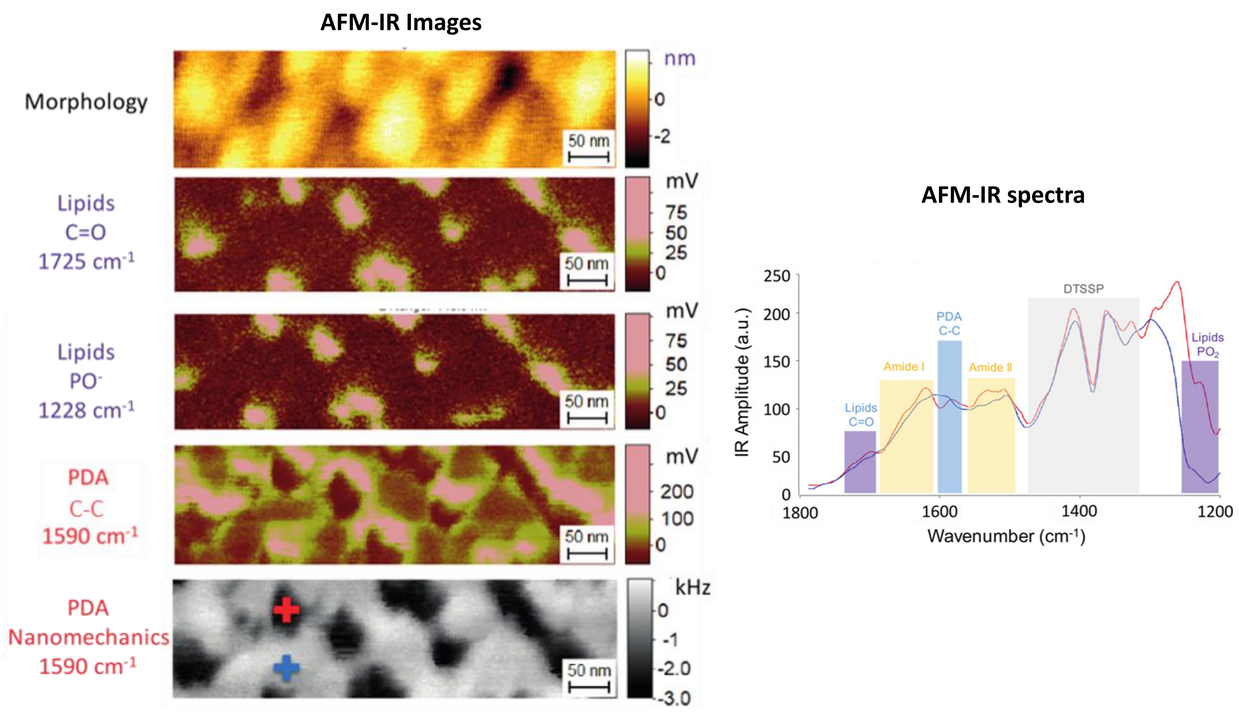

Figure 4: AFM-IR characterization of polydopamine-nanodiscs hybrid film on gold; on the left AFM micrographs at specific wavenumbers, morphology and nanomechanics; on the right AFM-IR recorded spectra for the spot were the nanodisc is present (red line) and were mostly polydopamine is present (blue line).

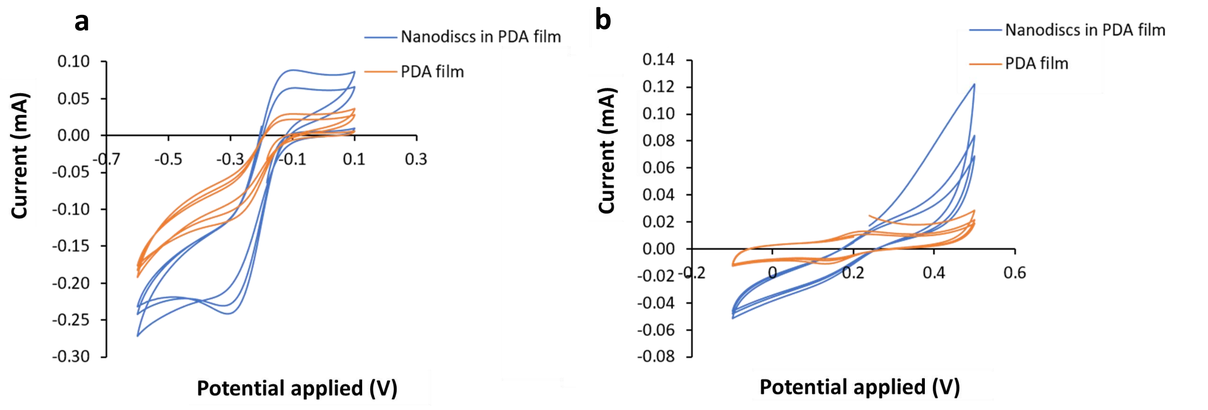

Figure 6: Cyclic voltammograms obtained from two redox probes. A. Ruthenium hexamine chloride (positively charged) B. Potassium ferrocyanide (negatively charged).

Conclusions and outlook

Non conductive Phospholipids nanodiscs were successfully embed in the polydopamine matrix retaining their shape and a certain availability. Proving electropolymerization is a mild process that allows fine deposition control opening the way to the production of new nanocomposite films with cell membranes mimicking features.

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