

The Location of Ubiquinone in a Hybrid Bilayer Membrane Studied by Neutron Reflectometry

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Studies of redox molecules in lipid bilayers are of interest for the development of biochemical sensors, one important example is the role of ubiquinone (UQ, coenzyme Q₁₀) in the electron transport chain. Ubiquinone consists of a redox-active quinone ring with a hydrophobic isoprenoid side chain. It is found in all cells and in particular the mitochondrial inner cell matrix. The role of the isoprene domain in ubiquinone mobility, redox reactions, and the location within the membrane have not been unequivocally explained. A biomimetic phospholipid hybrid bilayer membrane (HBM) supported on a gold coated quartz substrate with a proximal leaflet consisting of an octadecanethiol self assembled monolayer (SAM) and a distal leaflet formed through fusion of ubiquinone containing DMPC lipid vesicles was employed in this work.

The present study aims to provide new insight about the location of ubiquinone in a HBM and the influence of the redox state of UQ on the organization of the lipid matrix.

Reflectivity curves were collected and modelled for each layer of the system including the supporting metal layer on the quartz single crystal, the SAM in D₂O/NaF electrolyte, and distal lipid leaflets with and without

ubiquinone. The experiments were performed in a custom built liquid cell equipped with temperature and electrochemical control. The HBM were studied at 37°C and at both open circuit and reducing potentials.

In the figure the reflected intensity from an octadecanethiol SAM and a DMPC/octadecanethiol HBM and a DMPC/octadecanethiol HBM with 5% UQ incorporated is plotted on a log scale versus Q , the momentum transfer vector. The presence of UQ in the bilayer results in a reflected intensity higher than that from the HBM without UQ and was modelled with a larger thickness. With applied negative potential, the HBM without ubiquinone shows no change in reflectivity. With ubiquinone incorporated in the HBM, the applied negative potential reduces the ubiquinone and a small shift in the reflectivity curve results. This indicates that the bilayer remains intact and any change in the ubiquinone during the redox process do not significantly disrupt the structure of the bilayer. With supporting surface enhanced infrared adsorption spectroscopy (SEIRAS) experiments, our results suggest the ubiquinone molecules lie in the plane of the hydrophobic core of the HBM. This work is in preparation of publication.

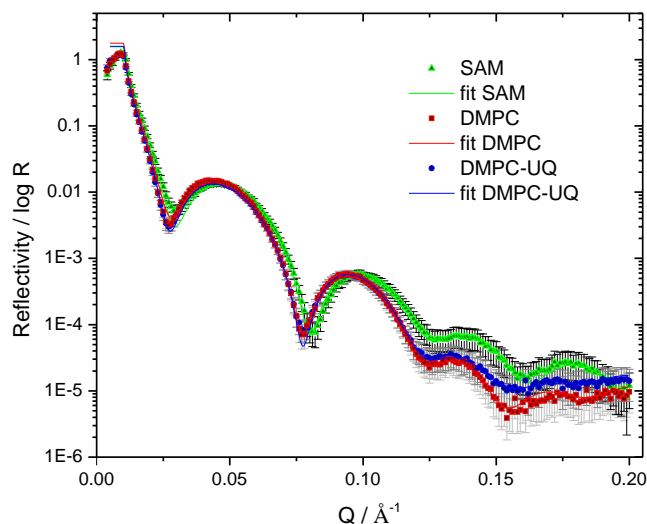


Figure 1: Neutron reflectivity curves with modelled fits for SAM (\square), DMPC/octadecanethiol HBM and UQ-DMPC/octadecanethiol HBM in 0.1 M NaF at 37°C.