

Supplementary Data

BIOELECTROCATALYTIC REDUCTION OF OXYGEN AT A SUPRAMOLECULARLY ASSOCIATED LACCASE ELECTRODE

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The electrochemical confirmation of the formation of a SAM of FcH and also a mixed SAM of FcH and MUA-Tyr was assessed by the linear dependence of the intensities of the anodic and cathodic peaks ( $i_{pa}$  and  $i_{pc}$ , respectively) on the scan rate applied (Figure S1).

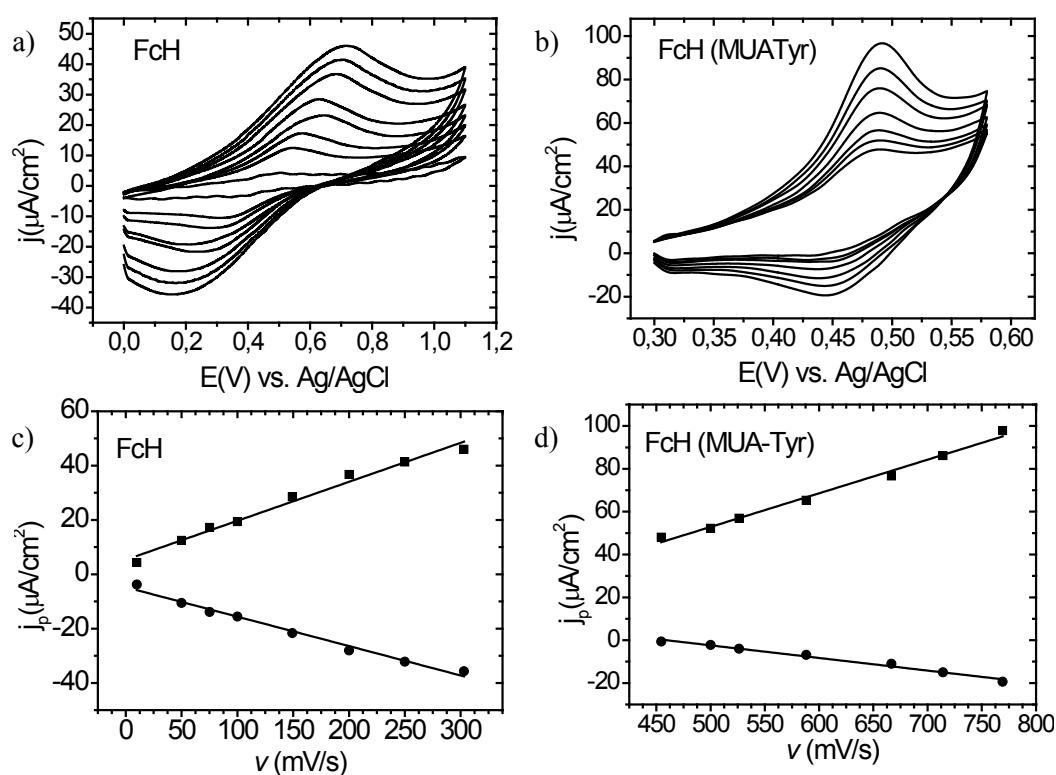


Figure S1. (a) Voltammogram of SAM of FcH and (b) of FcH in the mixed SAM with MUATyr at different scan rates; (c) linear fit between the intensity of the peaks and the scan rate applied for the SAM of FcH and (d) for the mixed SAM of FcH with MUA-Tyr.

Information about the organization of the mixed SAM of FcH and MUATyr can be obtained from the full width at half of the maximum height (fwhm). The theoretical value of fwhm for an electroactive species in a SAM is  $90.6/n$  mV, while differences from this value have been attributed to lateral interactions between electroactive species in SAM. The voltammogram of SAM of FcH (Figure S2 a, dashed) exhibited a broad shape (fwhm = 280 mV), testimony of strong lateral interactions between contiguous FcH moieties. However, when FcH is in the presence of MUATyr the voltammogram has a narrower shape (Figure S2 a, solid) (fwhm = 80 mV). This almost ideal value of fwhm indicates that lateral interactions between FcH have been significantly diminished by the presence of MUATyr, which can isolate individual FcH moieties. Hence, it may be assumed that the mixed SAM of FcH and MUATyr is characterized by a homogeneous distribution of both components.

In Figure S2 b, the cathodic branch of the reductive desorption experiment of FcH (dashed) and of FcH MUATyr (solid) may be appreciated.

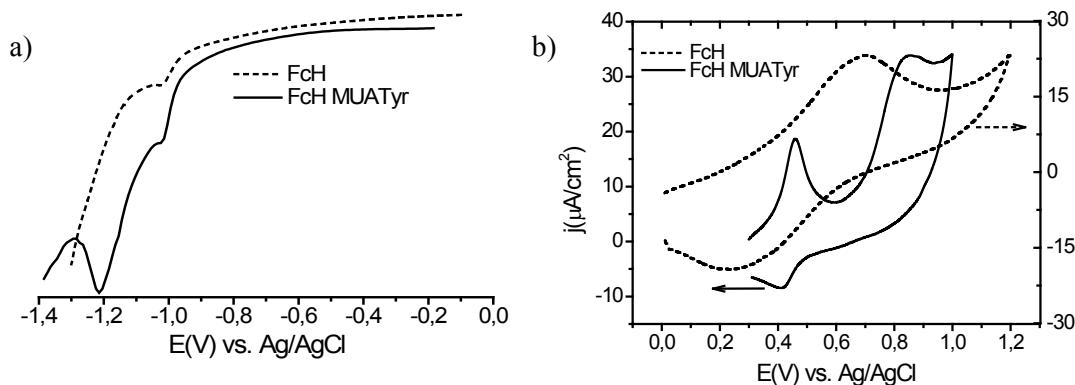


Figure S2. a) Cathodic branch of the reductive desorption of SAM of FcH (dashed), and of the mixed SAM of FcH and MUATyr (solid) b) Voltammogram of SAM of FcH (dashed), and of mixed SAM of FcH and MUATyr (solid). All scans were performed in the anodic direction; scan rate = 100 mV/s.

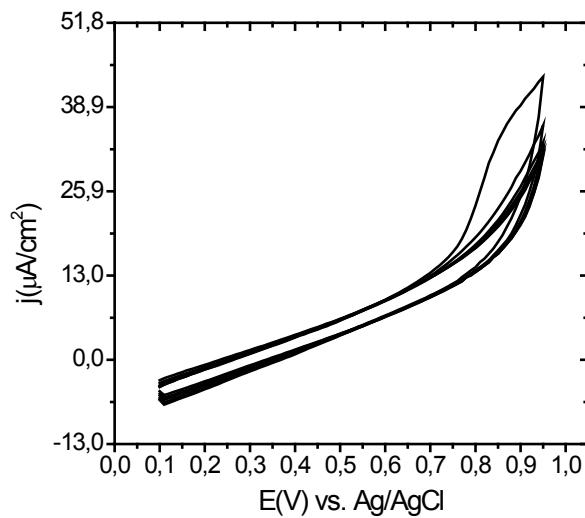


Figure S3. Voltammogram of the electrode modified with MUATyr and laccase supramolecularly immobilized, where no cathodic wave for dioxygen reduction was observed. All scans were performed in the anodic direction; scan rate = 100 mV/s.