Electrochemical generation of ferrylmyoglobin during oxidation of styrene with films of DNA and a poly(ester sulfonic acid) ionomer

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Abstract:
The chemistry of electrochemically-driven myoglobin-catalyzed oxidation of styrene was investigated in films of DNA or Eastman AQ ionomer on optically transparent electrodes. Conversion of styrene to styrene oxide proceeded via a ferrylmyoglobin radical intermediate. Ferrylmyoglobins were clearly detected by spectroelectrochemistry in films of 1–4 mm thick. The ferrylmyoglobin radical is produced by reaction of metmyoglobin (Mb) in the films with hydrogen peroxide formed by electrochemical catalytic reduction of oxygen catalyzed by Mb. Thus, electrochemically-driven styrene oxidation with these films proceeds by a ‘doubly catalytic’ electrode-driven reduction–oxidation pathway. Ferrylmyoglobin formation during electrolysis of Mb–DNA films in aerobic solutions was much faster, and styrene oxidation occurred with less Mb decomposition compared to the Mb–AQ films. The better performance of Mb–DNA films is correlated with a larger fraction of electroactive Mb and better stability than for the Mb–AQ films.