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IMMOBILIZATION OF NITROGENASE FOR BIOELECTROCHEMICAL AMMONIA SYNTHESIS

Janki H. Patel, Rong Cai, Shelley D. Minteer*

Department of Chemistry

Abstract

Ammonia has a variety of uses from cleaning products to fertilizer. Most commonly, ammonia is produced from hydrogen and nitrogen through the Haber-Bosch process, an energy-intensive process requiring high temperature and high pressure. In nature, N_2 is reduced by an enzyme called nitrogenase, at room temperature and ambient pressure. Nitrogenase the energy efficient reducing N_2 enzyme occurs using electrochemistry. Electrochemistry serves as a powerful tool to promote redox transformation by supplying or accepting electrons. Real-time observation of the 8-electron transfer and consumption of ATP allows insight to the nitrogenase mechanism. Limitations of this process arises with enzyme turnover, which occurs between the diffusional contact of the enzyme and electrode. Efficient electrons transfer can be achieved by immobilizing catalytic enzyme on the electrode surface. Here we apply a polypeptide (KDDD) modified pyrene as an anchor to immobilize nitrogenase on the electrode surface. The polypeptide segment is capable of coordinating with Ni ion, which could further bind with the His-tag region of nitrogenase. On the other end of the molecule, pyrene has been known to absorb on multiwall carbon nanotubes through pi-pi interactions. Successful immobilization of nitrogenase onto the electrode surface using the polypeptide (KDDD) modified pyrene allows the evaluation of the nitrogenase pathway for ammonia reduction. This mechanistic approach for nitrogenase could contribute to enhance electrocatalytic efficiency for ammonia production.