DEVELOPMENT OF ANODE ELECTRO-CATALYST FOR DIRECT ETHANOL PEM FUEL CELL

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by

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CERTIFICATE

This is to certify that the thesis entitled **Development of Anode Electro-Catalyst for Direct Ethanol PEM Fuel Cell** submitted by **Mrs. Jyoti Goel** to the Indian Institute of Technology Delhi, for the award of Degree of Doctor of Philosophy, is a record of the original bonafide research work carried out by her. She has worked under my supervision and has fulfilled the requirements, which to my knowledge, has reached the requisite standard for the submission of this thesis. The results contained in this thesis have not been submitted in part or full to any University or Institute for the award of any degree or diploma.

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ABSTRACT

State of the art Pt/C catalysts suffer from high cost and low catalytic activity towards ethanol oxidation, because of incomplete oxidation of ethanol in direct ethanol fuel cells (DEFCs). Ethanol is hard to oxidize completely into CO_2 and H_2O due to the difficulties in breaking the C-C bond at low temperature and the formation of CO-intermediates which poison the Pt catalyst. Thus it is necessary to develop catalyst with high selectivity towards ethanol oxidation reaction (EOR) in DEFC. The objective of this thesis is to study the performance and behavior of platinum based binary and ternary metal anode catalysts towards EOR. Binary Pt-Ir/C (20:20), Pt-Sn/C (20:20), Pt-Re/C (20:20) electro catalysts and ternary Pt-Ir-Sn/C and Pt-Re-Sn/C electro-catalysts with wt % (20:5:15), (20:10:10), (10:15:15) are prepared using co-impregnation reduction method. The prepared electro catalysts are physically characterized using scanning electron microscope (SEM), transmission electron microscope (TEM), energy dispersive X-ray spectroscope (EDX) and X-ray diffraction (XRD) technique. XRD and TEM study reveals that the prepared nano-particles are in the range of 10-20 nm. The prepared catalysts are electrochemically characterized by cyclic voltammetry (CV), chronoamperometry (CA), and linear sweep voltammetry (LSV) methods. Electrochemical characterization points out that small addition of third metal Ir or Re to Pt-Sn/C based catalysts enhances their activity towards ethanol oxidation.

DEFC results show that the ternary Pt-Ir-Sn/C (20:5:15) and Pt-Re-Sn/C (20:5:15) catalysts exhibit higher catalytic activity than binary Pt-Sn/C, Pt-Ru/C Pt-Ir/ C, Pt-Re/C and Pt/C anode catalysts. The effect of various operating parameters like ethanol fuel concentration, ethanol flow rate, operating temperature, anode diffusion layer, and oxygen flow rate on

DEFC are studied using Pt-Re-Sn (20:5:15)/C metal catalyst. Analysis of products obtained by the oxidation of ethanol in DEFC shows that acetic acid and acetaldehyde are the major products.

To study the effect support material on EOR, mesoporous carbon nitride (MCN) and treated-multiwalled carbon nanotubes (t-MWCNTs) are prepared and tested. The physical properties of different support materials e.g., MWCNTs, t-MWCNTs and MCN are studied using TGA, TEM, FTIR and XPS technique. Using MWCNTs, t-MWCNTs and MCN as support, Pt-Re-Sn and Pt-Ru metal anode catalysts are tested in DEFC. The results suggest that Pt-Ru/MCN catalyst show a maximum power density of 63.2 mW/cm^2, using 1 M ethanol as feed at 100 ^oC.

A comprehensive one dimensional (1-D) steady state mathematical model is developed using tafel kinetic relations for ethanol oxidation and mass transport of different species throughout the cell. Overall polarization of cell is obtained by systematically addressing the various useful parameters such as ethanol cross over, mixed potential effect due to ethanol crossover, limiting current behaviour and variation in anode and cathode over-potential in the catalyst layer. The mathematical model developed for DEFC predicted well the experimental data of Pt-Re-Sn/MCN (20:5:15), Pt-Ru/MCN (20:20) and Pt-Re-Sn/C (20:5:15) anode catalysts.

Based on the experimental and theoretical analyses, Pt-Ru/MCN found to be most suitable anode catalyst for DEFC in terms of cell performance.

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