

Supporting Information

Recovery of expensive Pt/C catalysts from end of life membrane electrode assembly of proton exchange membrane fuel cells

*Abha Bharti, †and Rajalakshmi Natarajan, †**

†Centre for Fuel Cell Technology, International Advanced Research Centre for Powder Metallurgy and New Materials, IITM-Research Park, Chennai-600113, India

E-mail: rajalakshmi@arci.res.in

Experimental Section

Materials

The recovery process was carried out using 5-layer end-of-life (EoL) membrane electrode assembly (MEA) of proton exchange membrane fuel cell (PEMFC) composed of catalyst (20 wt. % Pt/C) coated membrane (Nafion membrane 212) sandwiched between two polytetrafluoroethylene (PTFE) treated carbon cloth gas diffusion layer (GDL), iso-propanol (IPA) and deionized (DI) water. The recovered products were compared with commercial (fresh) 20 wt. % Pt/C (DURA-LYST) and 5 wt.% Nafion ionomer dispersion (Dupont).

Recovery process of Pt/C catalyst and Nafion membrane from EoL MEA of PEMFC

The 5-layer EoL MEA was physically delaminated to separate the catalyst coated membrane (CCM) from the carbon cloth GDL. The CCM (3.23 g containing 0.75 g of 20 wt.% Pt/C) was further sized into smaller chunks and dispersed in 50:50 v/v of DI water: IPA with a total solid content of 10 wt.%. Subsequently, it was sonicated for 60 min to aid catalyst detachment resulting in a viscous slurry. The complete detachment and dissolution of membrane was

achieved through hydrothermal treatment of the slurry at 200 °C for 24 h under ambient pressure. The Pt/C catalyst and ionomer enriched solution was recovered through vacuum filtration in form of retentate and filtrate solution, respectively. The recovered Pt/C powder was subsequently washed numerous times with DI water and acetone to remove any organic moieties and oven dried at 80 °C for 12 h. The recovered ionomer enriched solution was also subjected to heat treatment at 80 °C for 12 h to aid evaporation of the volatile solvents to yield ionomer powder and is designated as IP-R. The recovered catalyst, ionomer enriched solution is designated as Pt/C-R and IES-R, respectively.

Materials characterization

The functional group analysis of the recovered ionomer solution was carried out through Fourier transform infrared (FTIR) spectroscopy using Shimadzu IRAffinity-1S Fourier-Transform infrared spectrometer. The structure of the recovered Pt/C catalyst and ionomer powder was studied through X-ray diffraction (XRD) patterns recorded using Rigaku SmartLab X-ray diffractometer. The surface morphology of the recovered Pt/C catalyst was investigated using field emission scanning electron microscopy (FESEM, Carl Zeiss at 20 kV) and transmission electron microscopy (TEM, Technai G20 at 100 kV). The Pt content in the recovered catalyst was determined through thermal gravimetric analysis (TGA) using Netzsch thermal analyzer (STA 449 F1 Jupiter®) at heating rate of 10 °C min⁻¹ under air atmosphere.

Preparation of catalyst ink and working electrode

2 mg of Pt/C-R catalyst was mixed with 20 µL Nafion dispersion (5 wt.%) and 180 µL of DI water and sonicated to form homogenous catalyst ink dispersion. Subsequently, 2.5 µL of the catalyst ink was drop casted on glassy carbon (GC, geometrical surface area of 0.071 cm²) and air dried to form the working electrode. Similar procedure was carried out for the preparation

of working electrode with commercial Pt/C catalysts for comparison of the performance with the recovered Pt/C catalyst.

Electrochemical measurements

The electrochemical experiments were conducted in a standard three-electrode configuration using Biologic potentiostat (Vmp3). The experiments were carried out by employing catalyst coated working electrode, Pt counter electrode and saturated calomel electrode as reference electrode. All the potentials are converted with respect to reversible hydrogen electrode (RHE). Cyclic voltammetry (CV) experiments were conducted in N₂ purged 0.5 M H₂SO₄ electrolyte in the potential window 0 and 1 V vs. RHE at a scan rate of 50 mV s⁻¹. The stability of the recovered catalyst was accessed by cycling the working electrode from 0.6-1.0 V vs. RHE at a scan rate of 100 mVs⁻¹ for 5,000 cycles in N₂ saturated 0.5 M H₂SO₄.

Single cell PEMFC measurements

MEA for single cell PEMFC was made using teflonized carbon paper which acts as GDL and catalyst ink coated CCM. The catalyst ink for both the anode and cathode was prepared by ultrasonically mixing Pt/C-R catalyst, IPA, DI water and 10 wt% Nafion solution. CCM was prepared by ultrasonically coating the anode and cathode catalyst ink on membrane (Nafion 212) using ultrasonic coating machine (*USI Prism 500*). MEA with active geometrical area of 30 cm² was prepared by hot pressing CCM in between two teflonized carbon paper at 130 °C for 150 s. The Pt loading at the anode and cathode was maintained at 0.25 mg cm⁻² and 0.5 mg cm⁻², respectively. Subsequently, single cell PEMFC was assembled using MEA packed in between two graphite bi-polar plates which was further sandwiched between Au coated Cu current collectors and aluminium endplates. The fuel cell polarization curves were recorded at cell temperature of 60 °C with humidified, pure H₂ and air gases fed to the anode and cathode

compartment, respectively. The H₂ and air gases were fed at the constant flow rate of 1 lpm and 2 lpm, respectively.

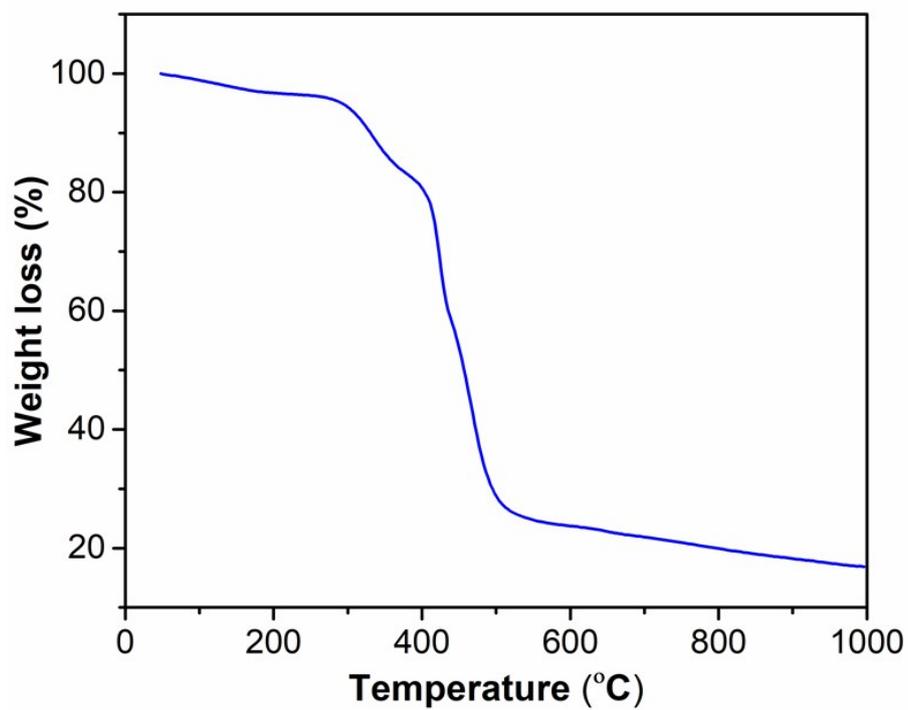


Fig. S1. Thermogram of Pt/C-R catalyst.