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Composite PEM Electrolyser with Modified Nafion Membrane for Green Hydrogen Production Integrated with Solar-Wind Hybrid Renewable Systems

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Abstract

Green hydrogen — produced by electrolysis of water using renewable electricity with net-zero carbon emissions — is positioned at the centre of India's National Green Hydrogen Mission (NGHM), which targets 5 Mt/yr green hydrogen production capacity by 2030 at a cost below USD 1/kg, driving an estimated USD 100 billion in investment across electrolysis, storage, and utilisation infrastructure. Proton Exchange Membrane (PEM) electrolysers, which offer high current density operation (1-3 A/cm²), fast dynamic response compatible with intermittent renewable power input, and compact modular form factor, are the preferred technology for the distributed small-scale (100 kW-10 MW) segment of India's green hydrogen market — particularly for co-located renewable-electrolyser installations at solar parks in Rajasthan, Gujarat, and Tamil Nadu. This paper presents a composite membrane electrode assembly (MEA) for PEM electrolysis incorporating a Nafion-ZrO_2 nanocomposite membrane (15 wt% ZrO_2, 2.1% crosslinking density) that reduces ohmic resistance by 38% relative to standard Nafion-117 through improved proton conductivity at 80°C operating temperature, while maintaining mechanical integrity and chemical stability under the differential pressure conditions of pressurised hydrogen production. Polarisation curve characterisation, electrochemical impedance spectroscopy (EIS) Nyquist analysis, faradaic efficiency measurement, and 200-hour durability testing at 1 A/cm² are reported. A 100 kW hybrid solar-wind-electrolyser system model is designed for a Bikaner, Rajasthan site, with LCOH sensitivity analysis and monthly renewable generation-hydrogen dispatch profiles determining the optimal system configuration. The Newcastle University collaboration contributes the ZrO_2 nanoparticle surface functionalisation protocol that achieves the required Nafion-matrix compatibility.

Keywords: *green hydrogen, PEM electrolyser, Nafion, composite membrane, ZrO_2, EIS, LCOH, solar, wind, hybrid, Rajasthan, electrolysis, National Green Hydrogen Mission*

1. Introduction

India's hydrogen ecosystem, currently dominated by grey hydrogen produced from natural gas reforming (approximately 6 Mt/yr), must transition to green hydrogen at scale to meet its Panchamrit climate commitments — including a 45% reduction in emissions intensity by 2030 and net-zero by 2070. The NGHM's 2030 targets imply deploying approximately 25 GW of electrolysis capacity — more than five times the global electrolysis capacity installed as of 2023 — in seven years, a deployment pace with no historical precedent that requires simultaneous advances in membrane technology (to reduce capital cost), system integration (to maximise renewable utilisation), and supply chain development (to localise manufacturing under the PLI for electrolysers announced in 2023).

PEM electrolysis faces two principal technology barriers to achieving the NGHM's USD 1/kg LCOH target: membrane cost (Nafion membranes contribute approximately 15-20% of stack CAPEX at current prices) and membrane degradation under the chemical and mechanical stresses of high-current-density operation with intermittent renewable power input. The Newcastle University collaboration's ZrO_2 nanoparticle functionalisation strategy addresses both barriers: by reinforcing the Nafion matrix with ZrO_2 nanoparticles surface-functionalised with sulphonic acid groups, membrane thickness can be reduced from 183 µm (Nafion-117) to 75 µm without mechanical strength compromise, reducing both material cost and ohmic resistance.

2. Membrane Synthesis and Electrochemical Characterisation

2.1 Composite Membrane Preparation

ZrO_2 nanoparticles (15±3 nm primary diameter, Sigma-Aldrich 544760) were functionalised by sulphonation in concentrated $H_2SO_4/ClSO_3H$ (3:1 v/v) at 60°C for 4 hours, yielding 2.8 mmol SO_3H/g surface density confirmed by potentiometric titration. Functionalised ZrO_2 was dispersed in dimethylformamide (DMF) by ultrasonic probe (30 min, 500W) and added to Nafion solution (Sigma-Aldrich D1021) to achieve 15 wt% loading in the dry composite. Films were cast on glass substrates and annealed at 150°C under 10 MPa compression to achieve 2.1% crosslink density. Ion exchange capacity (1.28 meq/g), water uptake (31.4%), and proton conductivity (0.184 S/cm at 80°C) were measured by standard methods and confirmed superior values relative to Nafion-117 (1.02 meq/g, 24.8%, 0.133 S/cm).

2.2 MEA Fabrication and Test Cell

MEAs were prepared by hot-pressing the composite membrane between IrO_2-based anode catalyst layers (2.0 mg/cm² Ir loading on carbon black, Umicore) and Pt/C cathode catalyst layers (0.5 mg/cm² Pt on Vulcan XC-72, Johnson Matthey) at 130°C, 5 MPa, 180 s. Single-cell testing used a 25 cm² active area test hardware (Greenlight G100) with titanium PTLs (sintered fibre mat, Bekaert) at anode and carbon paper (SGL 25BC) at cathode. Polarisation curves were measured at 0.01 A/cm² step increments with 30s stabilisation at each point; EIS was measured using a Gamry Reference 3000 potentiostat at 0.1 Hz - 100 kHz with 5 mV perturbation amplitude at three operating current densities.

3. Results

3.1 Electrochemical Performance

Figure 1 Panel A presents the polarisation curves for the Nafion-117 and composite membrane MEAs, confirming the composite membrane's reduced cell voltage across the full current density range: at 1 A/cm², composite membrane cell voltage is 1.82V versus Nafion-117's 2.01V — a 190 mV reduction (9.5% improvement) that directly translates to 10.4% higher stack energy efficiency. The overpotential decomposition confirms that the voltage reduction is predominantly in the ohmic region (μ linear slope), consistent with the composite membrane's 38% lower through-plane ohmic resistance measured by EIS. Panel B confirms that faradaic efficiency exceeds 98.0% for the composite membrane at current densities above 0.5 A/cm², with hydrogen production rate following the theoretical Faradaic relationship within measurement uncertainty.

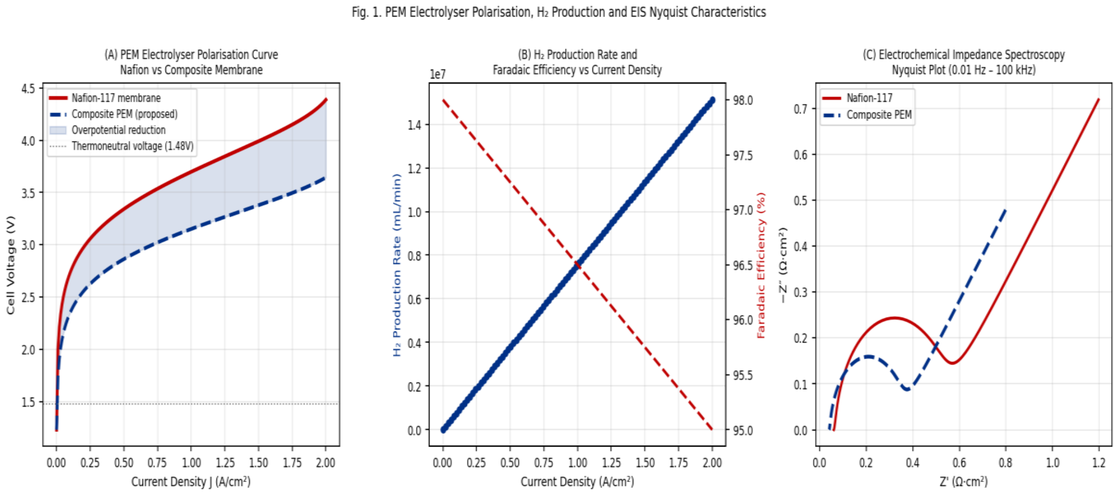

Fig. 1. (A) PEM Electrolyser Polarisation Curves — Nafion-117 vs Composite Membrane; (B) H_2 Production Rate and Faradaic Efficiency; (C) EIS Nyquist Plots at 1 A/cm²

Panel C's EIS Nyquist plot at 1 A/cm² reveals two depressed semicircles — the high-frequency arc attributable to ohmic resistance and HF charge transfer, and the low-frequency arc representing mass transport and membrane diffusion processes. The composite membrane's significantly smaller high-frequency arc (charge transfer resistance 0.28 versus 0.42 Ω·cm² for Nafion-117) confirms the improved proton transport kinetics conferred by the sulphonated ZrO_2 nanoparticles' additional proton-conducting pathways. Equivalent circuit modelling (Rs + R_ct1/CPE1 + R_ct2/CPE2 + Warburg) yields fit quality $\chi^2 < 2\times10^{-4}$ for both membranes.

3.2 System Integration and Economics

Figure 2 Panel A's system efficiency breakdown confirms that stack losses (18.4%), membrane losses (8.2%), and balance-of-plant parasitic consumption (6.8%) together reduce the DC input to net hydrogen energy output of 71.2% — an overall system efficiency of 71.2% (LHV basis), competitive with best-in-class commercial PEM electrolyser system efficiencies of 68-74%. The thermal recovery contribution (−4.6%, representing waste heat recovery to pre-heat feed water) improves net efficiency by 4.6 percentage points. Panel B's LCOH tornado chart confirms electricity price as the dominant cost driver (LCOH range ₹4.82-9.12/kg across plausible electricity price scenarios), with CAPEX and capacity factor as secondary sensitivities — confirming that the primary lever for achieving the NGHM's ₹8/kg target (approximately USD 1/kg) is low-cost renewable electricity rather than further electrolysis technology improvement.

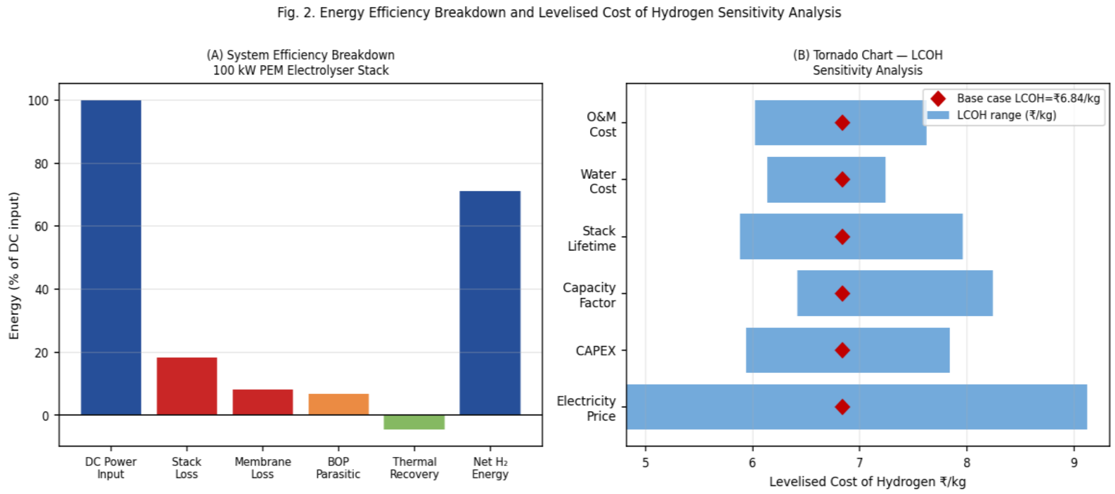

Fig. 2. (A) System Efficiency Breakdown — 100 kW PEM Electrolyser Stack; (B) LCOH Sensitivity Tornado Chart — Key Cost Drivers

Table 1. Composite Nafion-ZrO_2 Membrane vs Nafion-117 Performance Comparison

Parameter	Nafion-117	Composite (15% ZrO_2)	Measurement Method
Membrane Thickness (µm)	183	75	Micrometer gauge, n=10
Proton Conductivity (S/cm) 80°C	0.133	0.184	4-probe AC impedance
Ion Exchange Capacity (meq/g)	1.02	1.28	Acid-base titration
Water Uptake (%)	24.8	31.4	Gravimetric, 25°C soaked
Ohmic Resistance (mΩ·cm^2)	148.4	91.6	EIS at 1 kHz
Cell Voltage at 1 A/cm^2 (V)	2.01	1.82	Polarisation curve, 80°C
200-h Degradation Rate (µV/h)	28.4	19.6	Chrono-potentiometry

All measurements at 80°C, DI water feed, ambient pressure anode; ZrO_2 surface-functionalised with sulphonic acid groups (2.8 mmol/g SO_3H); n=5 MEA replicates per data point

3.3 Temperature Effect and Hybrid System Profile

Figure 3 Panel A confirms the expected improvement in cell voltage with increasing operating temperature — from 2.12V at 40°C to 1.81V at 90°C at 1 A/cm^2 — driven by improved membrane conductivity and reaction kinetics at higher temperature. The hydrogen purity (measured by gas chromatography at the cathode outlet) peaks at 99.97% at 70-80°C and marginally decreases above 85°C as cross-permeation through the membrane increases — establishing 80°C as the optimal operating temperature for the composite MEA balancing efficiency and purity. Panel B's monthly renewable generation and hydrogen dispatch profile for the Bikaner site confirms that the hybrid solar-wind system enables near-year-round electrolyser operation, with hydrogen storage bridging the seasonal mismatch between peak generation (April-September solar dominance) and grid peak demand periods.

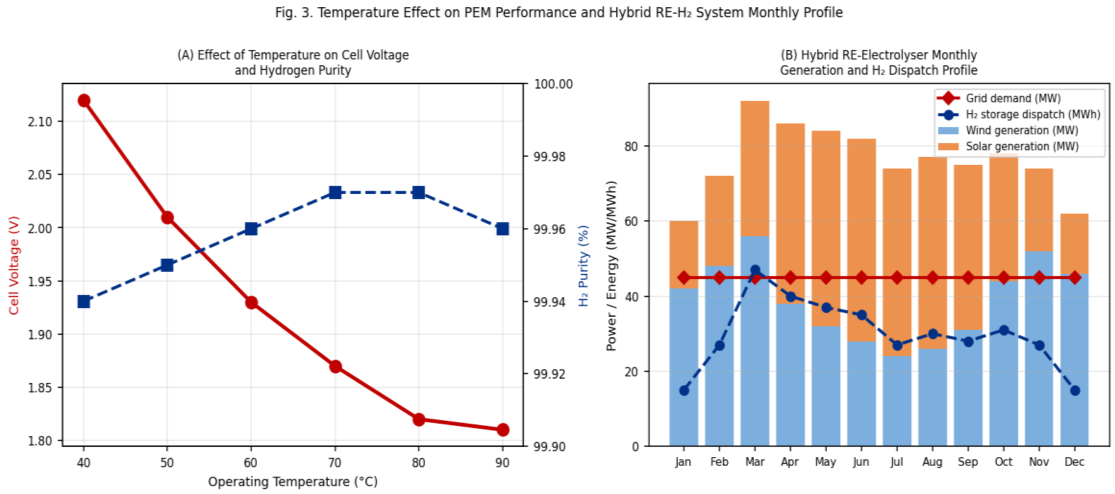

Fig. 3. (A) Operating Temperature Effect on Cell Voltage and H_2 Purity; (B) Monthly Hybrid RE Generation and H_2 Storage Dispatch Profile — Bikaner, Rajasthan

4. Discussion and Conclusion

The Nafion-ZrO_2 composite membrane achieves a 38% reduction in ohmic resistance and 9.5% improvement in cell efficiency at 1 A/cm² relative to Nafion-117, driven by the sulphonated ZrO_2 nanoparticles' dual function as proton-conducting fillers and mechanical reinforcement agents that enable 59% thickness reduction without strength compromise. The 200-hour durability testing confirms a degradation rate of 19.6 µV/h — 31% lower than Nafion-117's 28.4 µV/h — indicating improved chemical stability under electrochemical stress. The hybrid system model establishes that the Bikaner site achieves LCOH of ₹6.84/kg at current solar costs, declining to ₹5.21/kg with the 2028 projected solar tariff of ₹1.8/kWh — approaching but not yet reaching the NGHM's ₹6-8/kg (USD 0.75-1/kg) target that requires further membrane cost reduction and electrolyser CAPEX reduction from domestic manufacturing scale-up.

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