

EXPERIMENTAL INVESTIGATIONS ON PERFORMANCE IMPROVEMENT OF HYDROGEN-FUELLED PEM FUEL CELL USING OXYGEN-ENRICHED AIR AND GRAPHENE ELECTRODES

ROHAN KUMAR



**DEPARTMENT OF ENERGY SCIENCE AND ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY DELHI
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CELL USING OXYGEN-ENRICHED AIR AND GRAPHENE
ELECTRODES**

by

Rohan Kumar

Department of Energy Science and Engineering

Submitted

in fulfilment of the requirements of the degree of Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

May 2026

Dedicated

To

My Eternal Parents

And My loving family

Certificate

This is to certify that the thesis entitled "**Experimental Investigations on Performance Improvement of Hydrogen-Fuelled PEM Fuel Cell using Oxygen-Enriched Air and Graphene Electrodes**", being submitted by **Rohan Kumar** to the Indian Institute of Technology Delhi for the award of the degree of Doctor of Philosophy is a record of the bonafide research carried out by him which has been prepared under my supervision in conformity with the rules and regulations of the Indian Institute of Technology Delhi. He has fulfilled all requirements for the submission of this thesis, which has attained the standard required for a Ph.D degree of the Institute. The research reports and results presented in the thesis have not been submitted for any degree or diploma in any other university or institute.

Place: New Delhi

Date:

Prof. K. A. Subramanian

Professor

Department of Energy Science and Engineering

Indian Institute of Technology Delhi

New Delhi-110016

India

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Abstract

Fuel cells generate electricity with high energy efficiency and zero emissions. Among various types of fuel cells, the proton exchange membrane (PEM) fuel cell is more suitable for the automotive sector due to its better transient operation. The power density improves with an increase in current density; however, its energy efficiency decreases. Simultaneous improvement of both energy efficiency and power density in a PEM fuel cell is a technical challenge in fuel cells.

This research work is mainly focused on the characterization of materials for electrodes, and the enhancement of energy efficiency and power density using graphite-graphene electrode combinations and oxygen-enriched air from 23% oxygen (base) to 100%. It is observed from the Raman Spectra result indicates the sp^2 hybridised lattice, showing that the graphene material has a higher electrical conductivity (10^4 S/cm) compared to graphite (100 S/cm). The higher contact angle for graphene (154.8°) compared to graphite (107.5°) shows the superior hydrophobicity, facilitating the efficient water removal for avoiding water flooding and pore blocking.

In the next phase, the experimental tests were conducted on a hydrogen PEM fuel cell with different graphite-graphene electrode (anode and cathode) combinations, and the final phase with oxygen-enriched air (23% to 100% by mass). The experimental results indicate that the performance of the fuel cell is the highest with graphene (anode) - graphite (cathode) electrode combinations compared to other material combinations (graphite-graphite, graphite-graphene, graphene-graphene). The voltage efficiency at power output of 1 W improved from 47.96 % with graphite-graphite to 52.84 % with graphene-graphite due to a decrease in losses, as it is observed from the Nyquist plot that ohmic resistance and capacitance decreased from $0.113\ \Omega$ to $0.065\ \Omega$ and $3.8\ F$ to $1.57\ F$. The Tafel slope of $81.09\ mV/dec$ for graphene-graphite is lower than $119.25\ mV/dec$ for graphite, resulting in lower activation energy loss. The Bode plot shows the phase angle that is -26.5° for the graphite-graphite electrode and -18.5° for graphene-graphite, indicating a shift toward more resistive behavior. The maximum power density is $79.2\ mW/cm^2$ compared to base graphite-graphite electrodes ($53\ mW/cm^2$).

In the final phase, the performance of the fuel cell with all electrode combinations with oxygen-enriched air and 100% oxygen is analysed. The performance with graphene-graphite electrodes with 100% oxygen is the highest due to a reduction in losses (ohmic losses and activation losses). The voltage efficiency at power output of 1 W improved from 52.84% with 23% oxygen

in air to 66.87% with 100% oxygen with graphene-graphite due to a decrease in losses such as ohmic resistance ($0.101\ \Omega$ to $0.096\ \Omega$) and charge transfer resistance ($0.326\ \Omega$ to $0.203\ \Omega$). The maximum power density increased to $132.92\ \text{mW/cm}^2$, which is the highest. A notable conclusion emerged from this study is that voltage efficiency and power density with graphene (anode) -graphite (cathode) and 100% oxygen could be improved significantly.

सार

ईंधन सेल उच्च ऊर्जा दक्षता और शून्य उत्सर्जन के साथ बिजली उत्पन्न करते हैं। विभिन्न प्रकार के ईंधन सेल में, प्रोटॉन एक्सचेंज मेम्ब्रेन (PEM) ईंधन सेल अपने बेहतर क्षणिक संचालन के कारण ऑटोमोटिव क्षेत्र के लिए अधिक उपयुक्त है। धारा घनत्व में वृद्धि के साथ शक्ति घनत्व में सुधार होता है; हालाँकि, इसकी ऊर्जा दक्षता कम हो जाती है। प्रोटॉन एक्सचेंज मेम्ब्रेन (PEM) ईंधन सेल में ऊर्जा दक्षता और शक्ति घनत्व दोनों में एक साथ सुधार ईंधन सेल में एक तकनीकी चुनौती है।

यह शोध कार्य मुख्य रूप से इलेक्ट्रोड के लिए पदार्थों के लक्षण-निर्धारण, और ग्रेफाइट-ग्रेफीन इलेक्ट्रोड संयोजनों और ऑक्सीजन-समृद्ध वायु का उपयोग करके ऊर्जा दक्षता और शक्ति घनत्व को 23% ऑक्सीजन (आधार) से 100% तक बढ़ाने पर केंद्रित है। रमन स्पेक्ट्रा के परिणाम से यह देखा गया है कि sp^2 संकरित जालक दर्शाता है, जो दर्शाता है कि ग्रेफाइट (100 S/cm) की तुलना में ग्रेफीन पदार्थ की विद्युत चालकता (10^4 S/cm) अधिक है। ग्रेफाइट (107.5°) की तुलना में ग्रेफीन (154.8°) का उच्च संपर्क कोण इसकी उच्च जलभीति दर्शाता है, जिससे जल-बाढ़ और छिद्रों के अवरुद्ध होने से बचने के लिए कुशल जल निष्कासन संभव होता है।

अगले चरण में, विभिन्न ग्रेफाइट-ग्रेफीन इलेक्ट्रोड (एनोड और कैथोड) संयोजनों वाले हाइड्रोजन पीईएम ईंधन सेल पर, और अंतिम चरण में ऑक्सीजन-समृद्ध वायु (द्रव्यमान के अनुसार 23% से 100%) पर प्रायोगिक परीक्षण किए गए। प्रायोगिक परिणामों से पता चलता है कि अन्य पदार्थ संयोजनों (ग्रेफाइट-ग्रेफाइट, ग्रेफाइट-ग्रेफीन, ग्रेफीन-ग्रेफीन) की तुलना में, ग्रेफीन (एनोड) - ग्रेफाइट (कैथोड) इलेक्ट्रोड संयोजनों वाले ईंधन सेल का प्रदर्शन सबसे अधिक है। 1 वाट के पावर आउटपुट पर वोल्टेज दक्षता ग्रेफाइट-ग्रेफाइट के साथ 47.96% से बढ़कर ग्रेफीन-ग्रेफाइट के साथ 52.84% हो गई, ऐसा नुकसानों में कमी के कारण हुआ, जैसा कि नाइकिस्ट प्लॉट से देखा गया है कि ओमिक प्रतिरोध और धारिता 0.113Ω से घटकर 0.065Ω और $3.8 F$ से घटकर $1.57 F$ हो गई। ग्रेफीन-ग्रेफाइट के लिए 81.09 mV/dec का टैफेल ढलान ग्रेफाइट के लिए 119.25 mV/dec से कम है, जिसके परिणामस्वरूप कम सक्रियण ऊर्जा हानि होती है। बोड

अरेख कला कोण दर्शाता है जो ग्रेफाइट-ग्रेफाइट इलेक्ट्रोड के लिए -26.5° और ग्रेफीन-ग्रेफाइट के लिए -18.5° है, जो अधिक प्रतिरोधक व्यवहार की ओर बदलाव का संकेत देता है। बेस ग्रेफाइट-ग्रेफाइट इलेक्ट्रोड (53 mW/cm^2) की तुलना में अधिकतम शक्ति घनत्व 79.2 mW/cm^2 है।

अंतिम चरण में, ऑक्सीजन-समृद्ध वायु और 100% ऑक्सीजन वाले सभी इलेक्ट्रोड संयोजनों वाले ईंधन सेल के प्रदर्शन का विश्लेषण किया जाता है। 100% ऑक्सीजन वाले ग्राफीन-ग्रेफाइट इलेक्ट्रोड के साथ प्रदर्शन, हानियों (ओमिक हानियाँ और सक्रियण हानियाँ) में कमी के कारण सर्वोच्च है। 1 वाट विद्युत उत्पादन पर वोल्टेज दक्षता, 23% ऑक्सीजन वायु के साथ 52.84% से बढ़कर ग्राफीन-ग्रेफाइट के साथ 100% ऑक्सीजन के साथ 66.87% हो गई, ऐसा ओमिक प्रतिरोध (0.101Ω से 0.096Ω) और आवेश स्थानांतरण प्रतिरोध (0.326Ω से 0.203Ω) जैसी हानियों में कमी के कारण हुआ। अधिकतम शक्ति घनत्व बढ़कर 132.92 mW/cm^2 हो गया, जो कि सर्वाधिक है। इस अध्ययन से एक उल्लेखनीय निष्कर्ष यह निकला कि ग्राफीन (एनोड)-ग्रेफाइट (कैथोड) और 100% ऑक्सीजन के साथ वोल्टेज दक्षता और शक्ति घनत्व में उल्लेखनीय सुधार किया जा सकता है।

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Abbreviations and Nomenclatures

List of Abbreviations

BET	Brunauer–Emmett–Teller
CA	Chronoamperometry
CCM	Catalyst Coated Membrane
CTR	Charge Transfer Resistance
CL	Catalyst Layer
CNT	Carbon Nanotubes
CPE	Constant Phase Element
2D	Two Dimensional
DI	Deionized Water
EIS	Electrochemical Impedance Spectroscopy
FESEM	Field Emission Scanning Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy
HOR	Hydrogen Oxidation Reaction
HFR	High Frequency Resistance
IPA	Isopropanol
KBR	Potassium Bromide
G	Graphite
GDE	Gas Diffusion Electrode

GDL	Gas Diffusion Layer
GCG	Graphene Coated Graphite
LTPEM	Low Temperature Polymer Electrolyte Membrane
LIG	Laser-Induced Graphene
LSV	Linear Sweep Voltammetry
MEA	Membrane Electrode Assembly
MPL	Micro-porous layer
MWCNTs	Multi-Wall Carbon Nanotubes
NMP	N- Methyl Pyrrolidone
OCV	Open Circuit Voltage
ORR	Oxygen Reduction Reaction
PEM	Polymer Electrolyte Membrane
PEMFC	Polymer Electrolyte Membrane Fuel Cell
PFSA	Perfluoro Sulfonic acid
PTFE	Poly Tetra Fluoro Ethylene
RE	Reference Electrode
SWCNT	Single Wall Carbon Nano Tube
WE	Working Electrode
XRD	X-Ray Diffraction

List of Nomenclature

C	Capacitance
C_{dl}	Double layer capacitance
I	Current (A)
i	Current density (A/cm ²)
p_{H_2O}	Partial pressure of water
p_{O_2}	Partial pressure of oxygen
p_{H_2}	Partial pressure of hydrogen
F	Faraday's Constant
RH	Relative Humidity
R	Universal gas constant
R_s	Electrolyte Resistance
R_{Ct}	Charge transfer Resistance
r_p	Pore radius
T	Temperature
Δg_{rxn}^0	Gibbs Free energy of reaction
Δh	Change in enthalpy
V_{actual}	Actual circuit voltage
E_{nernst}	Nernst Potential
V_{act}	Activation overpotential
V_{ohmic}	Ohmic overpotential

V_{conc}	Concentration overpotential
$\eta_{thermo,fc}$	Fuel cell thermodynamic efficiency
n	Number of moles/electrons
Q	Charge (coulombs)
ω	Angular frequency
W	Warburg
Z_{real}	Real Impedance
Z_{imag}	Imaginary Impedance

