

**BIOGAS-SOLID OXIDE FUEL CELL (SOFC) SYSTEMS:
EXPERIMENTAL AND THEORETICAL STUDIES
FOR INTEGRATED SYSTEM DEVELOPMENT**

BIJU I. K.



**CENTRE FOR RURAL DEVELOPMENT AND TECHNOLOGY
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Biogas-Solid Oxide Fuel Cell (SOFC) Systems: Experimental and Theoretical Studies for Integrated System Development

by

BIJU I. K.

Centre for Rural Development and Technology

Submitted

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CERTIFICATE

This is to certify that the thesis entitled “**Biogas - Solid Oxide Fuel Cell (SOFC) Systems: Experimental and Theoretical Studies for Integrated System Development**” being submitted by **Mr. Biju I. K.** to the Indian Institute of Technology Delhi for the award of the degree of **Doctor of Philosophy**, is a record of the bonafide research work carried out by him. He has worked under our supervision and has fulfilled the requirements for the submission of this thesis, which has attained the standard required for a Ph. D. degree of the Institute. The research results presented in this thesis have not been submitted elsewhere for any degree or diploma award.

(Dr. Virendra Kumar Vijay)

Professor

Centre for Rural Development & Technology

Indian Institute of Technology Delhi

Hauz Khas, New Delhi – 110 016, India

(Dr. P. V. Aravind)

Professor

Faculty of Science & Engineering

Energy & Sustainability Research Institute

University of Groningen, The Netherlands

&

Director of Hydrogen Economy

Wubbo Ockels School

University of Groningen, The Netherlands

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BIJU I. K.

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ABSTRACT

Adopting appropriate waste-to-energy technology generates renewable energy, solves many environmental issues, and most importantly, plays a key role in developing a sustainable circular economy. Wastewater treatment plants (WWTPs) play a significant role in protecting the environment and public health by removing biological and chemical pollutants. In most conventional wastewater treatment plants, the influent to effluent water quality is improved at the cost of enormous energy input, which accounts for around 25 – 40 % of the operating cost. A major chunk of this energy consumption in a conventional WWTP goes for aeration of mixed liquor, followed by primary and secondary settling with sludge pumping and sludge dewatering.

It is apparently evident that energy recovery through anaerobic digestion is a must for the sustainability of WWTPs. Anaerobic digestion coupled with combined heat and power (CHP) has been adopted in various WWTPs in countries such as America, Austria, Germany, Netherlands, etc. The electrical efficiency attained through conventional Internal Combustion Engines (ICEs) and Micro Gas Turbines (MGTs) operated in CHP mode ranges from 25 to 45%. Furthermore, it varies with the CHP size, and the energy and economic sustainability of smaller plants are greatly affected by the low efficiency of traditional CHP systems. On the other hand, the application of Solid Oxide Fuel Cells (SOFCs) seems to be a potential alternative to the traditional IC engines. SOFCs are capable of delivering high electrical efficiencies in the range of 50 – 60 % even at lower sizes, viz, tens of kW_e. Alternatively, this feature makes it possible to modulate the power of the CHP system according to the demand/requirement. Another attractive aspect of SOFC is the near-zero emissions to the environment in terms of CO₂, NO_x, SO_x, PM, VOC, etc.

A primary concern regarding the direct utilization of biogas in SOFC is the presence of trace impurities. The composition and quantity of such trace gases vary from site to site due to the variation in the feed/substrates, operating conditions, etc. The main challenges and hurdles for the biogas-SOFC systems are the Ni-based anode degradation due to carbon deposition, nickel re-oxidation, and trace contaminants in the fuel, as this will affect the stable long-term operation of SOFCs. So, the trace contaminant levels in biogas should be properly controlled, for which the upper tolerance limits are to be arrived at based on detailed experimental studies. As of now, quite a good number of research publications are available for contaminant tolerance studies with hydrogen sulphide (H₂S) in the fuel gas fed to SOFC. The influence of contaminants on direct internal reforming (DIR) process is not reported much and lacks experimental investigation results. Hence, more research focus is needed on such studies for a well-engineered integrated system development. It is understood that a large number of investigations on biogas-SOFC integration are concentrated on numerical models and very few on experiments. The polarization losses that result in reduced cell performance of SOFCs are challenging to model and, hence, need to be determined experimentally.

This research focuses on performing experimental and simulation studies to evaluate the energy recovery potential from sewage streams through biogas – SOFC systems. This work is related to research line 2 of the LOTUS^{HR} (LOcal Treatment of Urban Sewage streams for Healthy Reuse) project. LOTUS^{HR} aims to demonstrate a novel holistic (waste-) water management approach for recovering water, energy, and nutrients from urban wastewater. The Barapullah Drain, located in Sarai Kale Kahn in central New Delhi, is identified by the Department of Biotechnology of the Ministry of

Science & Technology (DBT) as an ideal demonstration site for this interdisciplinary project.

This study to determine the feasibility of direct electricity production by utilizing the biogas generated from sewage treatment plant in an SOFC is performed with particular focus on direct internal reforming of biogas, contaminant tolerance studies with selected contaminants that are expected in sewage biogas, thermodynamic analysis of the integrated biogas-SOFC system and evaluating the biogas generation and power production potential based on the estimated wastewater discharge at Barapullah site. As of now, there are only a few experimental studies pertaining to direct internal reforming of biogas in SOFC. Also, very few studies are performed with the real composition of biogas, viz, CH₄:CO₂ in 3:2 ratio. Hence, this research used biogas with the prior mentioned composition to perform all the experiments (DIR, Contaminant tolerance studies) in a biogas-SOFC system.

The results from the theoretical and experimental studies on DIR revealed that complete conversion of methane is possible when the Ni-YSZ SOFC is operated at 850 °C or above. The contaminant tolerance studies with the expected level of HCl traces in biogas generated from WWTPs do not affect the system, while the methyl mercaptan traces in ppb(v) levels itself are impacting both DIR and the stable long-term operation of SOFC. This indicates the need for a stringent gas cleaning unit (GCU) for the cleaning of methyl mercaptan traces prior to feeding biogas to the SOFC unit. Finally, the biogas generation potential and system thermodynamics study explored in this research reveals the potential for waste recovery and power generation, which in turn gives a pivotal/guiding direction towards improving the sustainability of WWTPs.

सार

उपयुक्त अपशिष्ट-से-ऊर्जा तकनीक अपनाने से नवीकरणीय ऊर्जा उत्पन्न होती है, कई पर्यावरणीय समस्याएँ हल होती हैं, और सबसे महत्वपूर्ण बात, यह एक स्थायी चक्रीय अर्थव्यवस्था विकसित करने में महत्वपूर्ण भूमिका निभाती है। अपशिष्ट जल उपचार संयंत्र (डब्ल्यू. डब्ल्यू. टी. पी.) जैविक और रासायनिक प्रदूषकों को हटाकर पर्यावरण और जन स्वास्थ्य की रक्षा में महत्वपूर्ण भूमिका निभाते हैं। अधिकांश पारंपरिक अपशिष्ट जल उपचार संयंत्रों में, बहिःस्राव जल की गुणवत्ता में भारी ऊर्जा निवेश की लागत से सुधार होता है, जो परिचालन लागत का लगभग २५ - ४० % होता है। एक पारंपरिक डब्ल्यू. डब्ल्यू. टी. पी. में इस ऊर्जा खपत का एक बड़ा हिस्सा मिश्रित द्रव के वातन में जाता है, जिसके बाद स्लज पंपिंग और स्लज डीवाटरिंग द्वारा प्राथमिक और द्वितीयक निक्षेपण होता है।

यह स्पष्ट है कि डब्ल्यू. डब्ल्यू. टी. पी. की संधारणीयता के लिए अवायवीय पाचन के माध्यम से ऊर्जा की पुनर्प्राप्ति आवश्यक है। संयुक्त ताप और शक्ति (सीएचपी) के साथ अवायवीय पाचन को अमेरिका, ऑस्ट्रिया, जर्मनी, नीदरलैंड आदि जैसे देशों में विभिन्न डब्ल्यू. डब्ल्यू. टी. पी. में अपनाया गया है। सीएचपी प्रणालियों को नियोजित करके पारंपरिक आंतरिक दहन इंजनों (आई.सी.ई) और सूक्ष्म गैस टर्बाइनों (एम.जी.टी) के माध्यम से विद्युत दक्षता २५ से ४५ % तक होती है। इसके अलावा, यह सीएचपी के आकार के अनुसार अलग होता है और छोटे आकार के संयंत्रों की ऊर्जा और आर्थिक संधारणीयता पारंपरिक सीएचपी प्रणालियों की कम दक्षता से बहुत प्रभावित होती है। दूसरी ओर, सॉलिड ऑक्साइड फ्यूल सेल्स (एस. ओ. एफ. सी.) का अनुप्रयोग पारंपरिक आई. सी. इंजनों का एक संभावित विकल्प प्रतीत होता है। एस. ओ. एफ. सी. कम आकारों में भी ५०-६० % की सीमा में उच्च विद्युत दक्षता प्रदान करने में सक्षम हैं, अर्थात्, दसियों किलोवाट। वैकल्पिक रूप से, यह सुविधा मांग/आवश्यकता के अनुसार सीएचपी प्रणाली की शक्ति को संशोधित करना संभव बनाती है। एस. ओ. एफ. सी. का एक अन्य आकर्षक पहलू CO₂, NO_x, SO_x, PM, VOC, आदि के संदर्भ में पर्यावरण के लिए लगभग शून्य उत्सर्जन है।

एस. ओ. एफ. सी. में बायोगैस के प्रत्यक्ष उपयोग के संबंध में एक प्रमुख चिंता सूक्ष्म अशुद्धियों की उपस्थिति है। ऐसी सूक्ष्म गैसों की संरचना और मात्रा फीड/सब्सट्रेट, परिचालन स्थितियों आदि में भिन्नता के कारण अलग अलग जगहों पर भिन्न होती है। बायोगैस-एस. ओ. एफ. सी.

प्रणालियों के लिए मुख्य चुनौतियों और बाधाओं में कार्बन जमाव, निकल पुनः ऑक्सीकरण और ईंधन में सूक्ष्म संदूषकों की उपस्थिति के कारण निकल आधारित एनोड का क्षरण शामिल है, जो सभी एस. ओ. एफ. सी. के स्थिर दीर्घकालिक संचालन को प्रभावित कर सकते हैं। इसलिए, बायोगैस में सूक्ष्म संदूषकों के स्तरों को ठीक से नियंत्रित किया जाना चाहिए, जिसके लिए विस्तृत प्रयोगात्मक अध्ययनों के आधार पर ऊपरी सहिष्णुता सीमाओं पर पहुंचा जाना चाहिए। वर्तमान में, एस. ओ. एफ. सी. को दी जाने वाली ईंधन गैस में हाइड्रोजन सल्फाइड (H₂S) के साथ संदूषक सहिष्णुता के अध्ययन के संबंध में काफी मात्रा में शोध पत्र प्रकाशित किए गए हैं। प्रत्यक्ष आंतरिक सुधार (डी. आई. आर.) प्रक्रिया पर संदूषकों के प्रभाव के बारे में अधिक जानकारी उपलब्ध नहीं है और प्रायोगिक जाँच परिणामों का अभाव है। नतीजतन, एक सुसंचालित एकीकृत प्रणाली विकास के लिए ऐसे अध्ययनों पर अधिक शोध केंद्रित करने की आवश्यकता है। यह समझा जाता है कि बायोगैस-सॉलिड ऑक्साइड फ्यूल सेल (एसओएफसी) एकीकरण पर बड़ी संख्या में जांच संख्यात्मक मॉडल पर केंद्रित है और प्रयोगों पर बहुत कम है। धुवीकरण हानि, जिसके परिणामस्वरूप एस. ओ. एफ. सी. के सेल कार्यक्षमता में कमी आती है, का प्रतिरूपण करना कठिन होता है और इसलिए इसे प्रयोगात्मक रूप से निर्धारित करने की आवश्यकता होती है।

यह शोध बायोगैस-सॉलिड ऑक्साइड फ्यूल सेल (एसओएफसी) प्रणालियों के माध्यम से सीवेज धाराओं से ऊर्जा पुनर्प्राप्ति क्षमता का मूल्यांकन करने के लिए प्रयोगात्मक और अनुकरण अध्ययन दोनों करने पर केंद्रित है। यह कार्य लोटस^{एच. आर.} (स्वस्थ पुनः उपयोग के लिए शहरी सीवेज धाराओं का स्थानीय उपचार) परियोजना की अनुसंधान लाइन २ से संबंधित है। लोटसएच. आर. का उद्देश्य शहरी अपशिष्ट जल से पानी, ऊर्जा और पोषक तत्वों की पुनर्प्राप्ति के लिए एक नवीन समग्र (अपशिष्ट) जल प्रबंधन दृष्टिकोण का प्रदर्शन करना है। नई दिल्ली के मध्य भाग में सराय काले खां में स्थित बारापुला नाले की पहचान विज्ञान और प्रौद्योगिकी मंत्रालय के जैव प्रौद्योगिकी विभाग (डीबीटी) द्वारा इस अंतःविषय परियोजना के लिए एक आदर्श प्रदर्शन स्थल के रूप में की गई है।

यह अध्ययन एक सॉलिड ऑक्साइड फ्यूल सेल (एसओएफसी) में सीवेज ट्रीटमेंट प्लांट से उत्पन्न बायोगैस का उपयोग करके प्रत्यक्ष बिजली उत्पादन की व्यवहार्यता निर्धारित करने के लिए किया गया है, जिसमें बायोगैस के प्रत्यक्ष आंतरिक सुधार, सीवेज बायोगैस में अपेक्षित चयनित संदूषक के साथ संदूषक सहिष्णुता अध्ययन, एकीकृत बायोगैस-एसओएफसी प्रणाली के ऊष्मागतिकी

विश्लेषण और बारापुला साइट पर अनुमानित अपशिष्ट जल निर्वहन के आधार पर बायोगैस उत्पादन और बिजली उत्पादन क्षमता का मूल्यांकन करने पर विशेष ध्यान दिया गया है। वर्तमान में, सॉलिड ऑक्साइड फ्यूल सेल्स (एस. ओ. एफ. सी.) में बायोगैस के प्रत्यक्ष आंतरिक सुधार के संबंध में केवल सीमित संख्या में प्रयोगात्मक अनुसंधान हैं। इसके अलावा, बायोगैस की वास्तविक संरचना, अर्थात्, 3:2 अनुपात में CH_4 : CO_2 के साथ बहुत कम अध्ययन किए गए हैं। नतीजतन, इस अध्ययन ने बायोगैस-एस. ओ. एफ. सी. प्रणाली के भीतर सभी परीक्षणों (डी. आई. आर., संदूषक सहिष्णुता परीक्षण) का संचालन करने के लिए पूर्व उल्लिखित संरचना के साथ बायोगैस का उपयोग किया।

डी. आई. आर. पर सैद्धांतिक और प्रायोगिक अध्ययनों के परिणामों से पता चलता है कि मीथेन का पूर्ण रूपांतरण तब संभव है जब Ni-YSZ SOFC को ८५० डिग्री सेल्सियस या उससे अधिक पर संचालित किया जाता है। डब्ल्यू. डब्ल्यू. टी. पी. से उत्पन्न बायोगैस में एच. सी. एल. ट्रेस के अपेक्षित स्तर के साथ संदूषक सहिष्णुता अध्ययन प्रणाली को प्रभावित नहीं करते हैं जबकि पीपीबी (वी) स्तरों में मिथाइल मर्केप्टान ट्रेस स्वयं डी. आई. आर. और एस. ओ. एफ. सी. के स्थिर दीर्घकालिक संचालन दोनों को प्रभावित कर रहे हैं। यह एस. ओ. एफ. सी. इकाई को बायोगैस देने से पहले मिथाइल मर्केप्टान के अंशों की सफाई के लिए एक सख्त गैस सफाई इकाई (जी. सी. यू.) की आवश्यकता को इंगित करता है। अंत में, इस शोध में खोजे गए बायोगैस उत्पादन क्षमता और प्रणाली ऊष्मागतिकी अध्ययन से अपशिष्ट पुनर्प्राप्ति और बिजली उत्पादन की क्षमता का पता चलता है, जो बदले में डब्ल्यूडब्ल्यूटीपी की संधारणीयता में सुधार की दिशा में एक महत्वपूर्ण/मार्गदर्शक दिशा देता है।

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ABBREVIATIONS AND SYMBOLS

AD	anaerobic digestion
AD-DAF	anaerobic digestion dissolved air flotation
AnMBR	anaerobic membrane bioreactor
APU	auxiliary power unit
ASR	area specific resistance
AS-SOFC	anode-supported solid oxide fuel cell
ATR	auto thermal reforming
BOD	biochemical oxygen demand
CBG	compressed biogas
CEM	controlled evaporator mixer
CH ₄	methane
CH ₄ SH	methyl mercaptan / methanethiol
CHP	combined heat and power
CO	carbon monoxide
CO ₂	carbon dioxide
COD	chemical oxygen demand
COS	carbonyl sulphide
DAF	dissolved air flotation
DBT	department of biotechnology
DEMOSOFC	DEMONstration of a large SOFC system fed with biogas from wastewater treatment plants
DIR	direct internal reforming
DOR	dry oxidation reforming
DR	dry reforming
EDS	energy-dispersive spectroscopy
ER	external reforming
E _{rev}	reversible voltage
f(T)	volumetric correction factor due to temperature
FESEM	field emission scanning electron microscope
GC	gas chromatograph

GCU	gas cleaning unit
GDC	gadolinium-doped ceria
GHG	greenhouse gas
GWP	global warming potential
H ₂	hydrogen
HCl	hydrogen chloride
HCV	higher calorific value
HHV	higher heating value
HOR	hydrogen oxidation reaction
HRT	hydraulic retention time
H ₂ S	hydrogen sulphide
I _L	loss index of gas in the reactor due to leakage
ICE	internal combustion engine
ICDD	international centre for diffraction data
IPCC	intergovernmental panel on climate change
kW	kilo watt
kWh	kilo watt-hour
LFG	landfill gas
LCV	lower calorific value
LHV	lower heating value
LOTUS ^{HR}	Local Treatment of Urban Sewage streams for Healthy Reuse
LSCF	lanthanum strontium cobalt ferrite
LSM	lanthanum strontium manganite
MCFC	molten carbonate fuel cell
m ³	cubic metre
MFC	mass flow controller
MFM	mass flow meter
MGT	micro gas turbine
MJ	mega joule
MLD	million litres per day
MPa	mega pascal
MW	mega watt

MWh	mega watt-hour
N ₂	nitrogen
Ni	nickel
NiO	nickel oxide
NO _x	nitrogen oxides
OCV	open circuit voltage
OLR	organic loading rate
P _{fcell}	fuel cell pressure
PEMFC	polymer electrolyte membrane fuel cell
POX	partial oxidation
PREACT	reactor pressure
Q _{BGAR}	total biogas flow rate from the anaerobic reactor
Q _{SEW}	total sewage flow rate into the reactor
RSD	relative standard deviation
S	effluent COD in kg/m ³
S ₀	influent COD in kg/m ³
ScSZ	scandia-stabilized zirconia
SDC	samarium-doped ceria
SDG	sustainable development goal
SEM	scanning electron microscopy
SOFC	solid oxide fuel cell
SO _x	sulphur oxides
SR	steam reforming
SRT	solids retention time
STP	standard temperature and pressure
STPs	sewage treatment plants
T _{fcell}	fuel cell temperature
TPB	triple-phase boundary
TREACT	reactor temperature
TVSMRE	total volatile solids mass removal efficiency
UASB	up-flow anaerobic sludge blanket
VOC	volatile organic compound
WGSR	water gas shift reaction

WWTP	wastewater treatment plant
XRD	x-ray diffraction
Y	solid production yield
YSZ	yttria-stabilized zirconia
η_{act}	activation overpotential
η_{ohmic}	ohmic overpotential
η_{conc}	concentration overpotential