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Pages 1-1292 (15 September 2018)

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Export citations
Show all article previews

Contents

Regular Papers
VSI:ICAE 2017
VSI:DEM 2018
VSI:Energy Justice
VSI:REM 2017

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☐ ● Full text access
Editorial Board
Page ii
Download PDF

Regular Papers

☐ Research article ○ Abstract only
The recovery mechanism of proton exchange membrane fuel cell in micro-current operation
Pucheng Pei, Xiaoning Jia, Huachi Xu, Pengcheng Li, ... Shangwei Huang
Pages 1-9
Article preview

☐ Research article ○ Abstract only
Natural gas hydrate exploitation by CO_2/H_2 continuous Injection-Production mode
Yi-Fei Sun, Jin-Rong Zhong, Rui Li, Tao Zhu, ... Chang-Yu Sun
Pages 10-21
Article preview

☐ Research article ○ Abstract only
A novel catalytic alkylation process of syngas with benzene over the cerium modified platinum supported on HZSM-5 zeolite
Fan Yang, Jie Zhong, Xiaohui Liu, Xuedong Zhu
Pages 22-30
Article preview

☐ Research article ○ Abstract only
Assessing future water resource constraints on thermally based renewable energy resources in California
Brian Tarroja, Felicia Chiang, Amir AghaKouchak, Scott Samuelsen
Pages 49-60
Article preview

☐ Research article ○ Abstract only
Experimental results of a small-scale organic Rankine cycle: Steady state identification and application to off-design model validation
Steven Lecompte, Sergei Gusev, Bruno Vanslambrouck, Michel De Paepe
Pages 82-106
Article preview

☐ Research article ○ Abstract only
High-temperature stability of nitrate/nitrite molten salt mixtures under different atmospheres
Carolina Villada, Alexander Bonk, Thomas Bauer, Francisco Bolívar
Pages 107-115
Article preview

☐ Research article ○ Abstract only
Optimal design of a district energy system including supply for fuel cell electric vehicles
Christoph Wilke, Astrid Bensmann, Stefan Martin, Annika Utz, Richard Hanke-Rauschenbach
Pages 129-144
Article preview

☐ Research article ○ Abstract only
An integrated diesel fuel processing system with thermal start-up for fuel cells
Remzi Can Samsun, Matthias Prawitz, Andreas Tschauder, Joachim Pasel, ... Detlef Stolten
Pages 145-159
Article preview

☐ Research article ○ Abstract only
Enhancement of the cooling capability of a high concentration photovoltaic system using microchannels with forward triangular ribs on sidewalls
Mario Di Capua H, Rodrigo Escobar, A.J. Diaz, Amador M. Guzmán
Pages 160-180
Article preview

☐ Research article ○ Abstract only
Assessing the potential of a hybrid battery system to reduce battery aging in an electric vehicle by studying the cycle life of a graphite|NCA high energy and a LTO|metal oxide high power battery cell considering realistic test profiles
Raphael Wegmann, Volker Döge, Dirk Uwe Sauer
Pages 197-212
Article preview

☐ Research article ○ Abstract only
Appliance labeling and consumer heterogeneity: A discrete choice experiment in India
Manisha Jain, Anand B. Rao, Anand Patwardhan
Pages 213-224
Article preview

☐ Research article ○ Abstract only
System-level performance optimization of molten-salt packed-bed thermal energy storage for concentrating solar power
Bing-chen Zhao, Mao-song Cheng, Chang Liu, Zhi-min Dai
Pages 225-239
Article preview

☐ Research article ○ Abstract only
Categorising virtual water transfers through China's electric power sector
Xiawei Liao, Xu Zhao, Jim W. Hall, Dabo Guan
Pages 252-260
Article preview

☐ Research article ○ Abstract only
Cost-effective supply chain for electric vehicle battery remanufacturing
Lin Li, Fadwa Dababneh, Jing Zhao
Pages 277-286
Article preview

☐ Research article ○ Abstract only
Assessment of energy efficiency and solvent retention inside steam chamber of steam- and solvent-assisted gravity drainage process
Hao Liu, Linsong Cheng, Keliu Wu, Shijun Huang, Brij B. Maini
Pages 287-299
Article preview

☐ Research article ○ Abstract only
Design and optimization of a Tesla turbine for ORC applications
L. Talluri, D. Fiaschi, G. Neri, L. Ciappi
Pages 300-319
Article preview

☐ Research article ○ Abstract only
Optimal design method for concentrating photovoltaic-thermoelectric hybrid system
Ershuai Yin, Qiang Li, Yimin Xuan
Pages 320-329
Article preview

☐ Research article ○ Abstract only
Experimental validation of additively manufactured optimized shapes for passive cooling
Boyan S. Lazarov, Ole Sigmund, Knud E. Meyer, Joe Alexandersen
Pages 330-339
Article preview

☐ Research article ○ Abstract only
Engine-out emissions from a modern high speed diesel engine – The importance of Nozzle Tip Protrusion
Felix Leach, Riyaz Ismail, Martin Davy
Pages 340-352
Article preview

☐ Research article ○ Abstract only
Study on a high-performance photocatalytic-Trombe wall system for space heating and air purification
Bendong Yu, Jingxin Hou, Wei He, Shanshan Liu, ... Gang Xu
Pages 365-380
Article preview

☐ Research article ○ Abstract only
Cylindrical thermoelectric generator with water heating system for high solar energy conversion efficiency
Hirofumi Hazama, Yumi Masuoka, Akitoshi Suzumura, Masato Matsubara, ... Ryoji Asahi
Pages 381-388
Article preview

☐ Research article ○ Abstract only
Effects of water content on evaporation and combustion characteristics of water emulsified diesel spray
Zhaowen Wang, Shuguo Shi, Sheng Huang, Jie Tang, ... Jyh-Yuan Chen
Pages 397-407
Article preview

☐ Research article ○ Abstract only
Multiple learning backtracking search algorithm for estimating parameters of photovoltaic models
Kunjie Yu, J.J. Liang, B.Y. Qu, Zhiping Cheng, Heshan Wang
Pages 408-422
Article preview

☐ Research article ○ Abstract only
Optimal design of a hybrid electric propulsive system for an anchor handling tug supply vessel
Jianyun Zhu, Li Chen, Bin Wang, Lijuan Xia
Pages 423-436
Article preview

☐ Research article ● *Open access*
Cost-optimal electricity systems with increasing renewable energy penetration for islands across the globe
Dean Marcus Gioutsos, Kornelis Blok, Leonore van Velzen, Sjoerd Moorman
Pages 437-449
Download PDF Article preview

☐ Research article ○ Abstract only
Development of a vision-based soft sensor for estimating equivalence ratio and major species concentration in entrained flow biomass gasification reactors
Yngve Ögren, Pál Tóth, Attila Garami, Alexey Sepman, Henrik Wiinikka
Pages 450-460
Article preview

☐ Research article ○ Abstract only
Effect of water content on [Bmim][HSO_4] assisted in-situ transesterification of wet *Nannochloropsis oceanica*
Yingqiang Sun, Chunyan Xu, Thomas Igou, Peilu Liu, ... Yongsheng Chen
Pages 461-468
Article preview

☐ Research article ○ Abstract only
Bargaining-based cooperative energy trading for distribution company and demand response
Songli Fan, Qian Ai, Longjian Piao
Pages 469-482
Article preview

☐ Research article ○ Abstract only
Development of robust suboptimal real-time power sharing strategy for modern fuel cell based hybrid tramways considering operational uncertainties and performance degradation
Fei Peng, Yuanzhe Zhao, Ting Chen, Xuexia Zhang, ... Qi Li
Pages 503-521
Article preview

☐ Research article ○ Abstract only
Highly efficient conversion of methane using microsecond and nanosecond pulsed spark discharges
Yuan Gao, Shuai Zhang, Hao Sun, Ruixue Wang, ... Tao Shao
Pages 534-545
Article preview

☐ Research article ○ Abstract only
Numerical stiffness study of multi-physical solid oxide fuel cell model for real-time simulation applications
Rui Ma, Chen Liu, Elena Breaz, Pascal Briois, Fei Gao
Pages 570-581
Article preview

☐ Research article ○ Abstract only
Dynamic modeling of a particle/supercritical CO_2 heat exchanger for transient analysis and control
M. Fernández-Torrijos, K.J. Albrecht, C.K. Ho
Pages 595-606
Article preview

☐ Research article ○ Abstract only
Drastic bandwidth enhancement of bistable energy harvesters: Study of subharmonic behaviors and their stability robustness
Thomas Huguet, Adrien Badel, Olivier Druet, Mickaël Lallart
Pages 607-617
Article preview

☐ Research article ○ Abstract only
A new approach to electricity market clearing with uniform purchase price and curtailable block orders
Iacopo Savelli, Bertrand Cornélusse, Antonio Giannitrapani, Simone Paoletti, Antonio Vicino
Pages 618-630
Article preview

☐ Research article ○ Abstract only
Risk-averse stochastic model predictive control-based real-time operation method for a wind energy generation system supported by a pumped hydro storage unit
Uğur Yıldıran, İsmail Kayahan
Pages 631-643
Article preview

☐ Research article ○ Abstract only
Balancing marine ecosystem impact and freshwater consumption with water-use fees in California's power markets: An evaluation of possibilities and trade-offs
Jose Bolorinos, Yang Yu, Newsha K. Ajami, Ram Rajagopal
Pages 644-654
Article preview

☐ Research article ○ Abstract only
A high-fidelity wave-to-wire simulation platform for wave energy converters: Coupled numerical wave tank and power take-off models
Markel Penalba, Josh Davidson, Christian Windt, John V. Ringwood
Pages 655-669
Article preview

☐ Research article ○ Abstract only
Experimental investigation on the efficiency of circular cylinder-based wind energy harvester with different rod-shaped attachments
Gang Hu, K.T. Tse, Minghai Wei, R. Naseer, ... K.C.S. Kwok
Pages 682-689
Article preview

☐ Research article ○ Abstract only
Thermal analysis of a thermoelectric generator for light-duty diesel engines
Pablo Fernández-Yañez, Octavio Armas, Azael Capetillo, Simón Martínez-Martínez
Pages 690-702
Article preview

☐ Research article ○ Abstract only
Optimal scheduling of a microgrid in a volatile electricity market environment: Portfolio optimization approach
Y. Chen, M. Trifkovic
Pages 703-712
Article preview

☐ Review article ○ Abstract only
A Review of Transparent Insulation Material (TIM) for building energy saving and daylight comfort
Yanyi Sun, Robin Wilson, Yupeng Wu
Pages 713-729
Article preview

☐ Research article ○ Abstract only
Effects of hydrolysis and oxidative hydrolysis pretreatments on upgrading of the water-soluble fraction of bio-oil via decarboxylation
Yuan Zhang, Yong Wang, Hongyou Cui, Pingping Zhao, ... Lihong Wang
Pages 730-742
Article preview

☐ Research article ○ Abstract only
A hybrid system for short-term wind speed forecasting
Qingqing He, Jianzhou Wang, Haiyan Lu
Pages 756-771
Article preview

☐ Research article ● *Open access*
The development and validation of a vehicle simulator for the introduction of Worldwide Harmonized test protocol in the European light duty vehicle CO_2 certification process
Georgios Fontaras, Víctor Valverde, Vincenzo Arcidiacono, Stefanos Tsiakmakis, ... Biagio Ciuffo
Pages 784-796
Download PDF Article preview

☐ Research article ○ Abstract only
The impact from the direct injection and multi-port fuel injection technologies for gasoline vehicles on solid particle number and black carbon emissions
Liqiang He, Jingnan Hu, Shaojun Zhang, Ye Wu, ... Sheng Su
Pages 819-826
Article preview

☐ Research article ○ Abstract only
High catalytic performance of Fe-Ni/Palygorskite in the steam reforming of toluene for hydrogen production
Xuehua Zou, Tianhu Chen, Ping Zhang, Dong Chen, ... Steven L. Suib
Pages 827-837
Article preview

☐ Research article ● *Open access*
Microbial community adaptability to altered temperature conditions determines the potential for process optimisation in biogas production
M. Westerholm, S. Isaksson, O. Karlsson Lindsjö, A. Schnürer
Pages 838-848
Download PDF Article preview

☐ Research article ○ Abstract only
Influence of geometrical parameters on the flow characteristics of multi-pipe earth-to-air heat exchangers – experimental and CFD investigations
Łukasz Amanowicz
Pages 849-861
Article preview

☐ Research article ○ Abstract only
Potential of energy savings and CO_2 emission reduction in China's iron and steel industry
Runying An, Biying Yu, Ru Li, Yi-Ming Wei
Pages 862-880
Article preview

☐ Research article ○ Abstract only
A feasible way to handle the heat management of direct carbon solid oxide fuel cells
Haoran Xu, Bin Chen, Peng Tan, Weizi Cai, ... Meng Ni
Pages 881-890
Article preview

☐ Research article ○ Abstract only
Trade-offs between economic and environmental performance of an autonomous hybrid energy system using micro hydro
W. Apichonnabutr, A. Tiwary
Pages 891-904
Article preview

☐ Research article ○ Abstract only
Cooperative game theory and last addition method in the allocation of firm energy rights
Victor. A.D. Faria, Anderson Rodrigo de Queiroz, Luana M.M. Lima, José W.M. Lima
Pages 905-915
Article preview

☐ Research article ○ Abstract only
System-level energy consumption modeling and optimization for cellulosic biofuel production
Yuntian Ge, Lin Li
Pages 935-946
Article preview

☐ Research article ○ Abstract only
Experimental and analytical study on capture spray liquid penetration and combustion characteristics simultaneously with Hydrogenated Catalytic Biodiesel/Diesel blended fuel
Weiwei Shang, Zhixia He, Qian Wang, Jiawei Cao, ... Da Li
Pages 947-956
Article preview

☐ Research article ○ Abstract only
Risk constrained stochastic economic dispatch considering dependence of multiple wind farms using pair-copula
M.S. Li, Z.J. Lin, T.Y. Ji, Q.H. Wu
Pages 967-978
Article preview

☐ Research article ○ Abstract only
Optimal design and operation of integrated solar combined cycles under emissions intensity constraints
Philip G. Brodrick, Adam R. Brandt, Louis J. Durlofsky
Pages 979-990
Article preview

☐ Research article ○ Abstract only
Intertemporal optimization of synthesis, design and operation of integrated energy systems of ships: General method and application on a system with Diesel main engines
George N. Sakalis, Christos A. Frangopoulos
Pages 991-1008
Article preview

☐ Research article ○ Abstract only
Novel performance curves to determine optimal operation of CCHP systems
Sayyed Faridoddin Afzali, Vladimir Mahalec
Pages 1009-1036
Article preview

☐ Research article ○ Abstract only
Techno-economic analysis of high-efficiency natural-gas generators for residential combined heat and power
Gokul Vishwanathan, Julian P. Sculley, Adam Fischer, Ji-Cheng Zhao
Pages 1064-1075
Article preview

☐ Research article ● *Open access*
Thermodynamic evaluation of bi-directional solid oxide cell systems including year-round cumulative exergy analysis
G. Botta, R. Mor, H. Patel, P.V. Aravind
Pages 1100-1118
Download PDF Article preview

☐ Research article ● *Open access*
A thermo-economic analysis and comparison of pumped-thermal and liquid-air electricity storage systems
Solomos Georgiou, Nilay Shah, Christos N. Markides
Pages 1119-1133
Download PDF Article preview

☐ Research article ● *Open access*
A physical supply-use table framework for energy analysis on the energy conversion chain
Matthew Kuperus Heun, Anne Owen, Paul E. Brockway
Pages 1134-1162
Download PDF Article preview

☐ Research article ○ Abstract only
Effects of injection pressure on ignition and combustion characteristics of impinging diesel spray
Wei Du, Qiankun Zhang, Zheng Zhang, Juejue Lou, Wenhua Bao
Pages 1163-1168
Article preview

☐ Research article ○ Abstract only
Using ducted fuel injection to attenuate or prevent soot formation in mixing-controlled combustion strategies for engine applications
R.K. Gehmlich, C.J. Mueller, D.J. Ruth, C.W. Nilsen, ... J. Manin
Pages 1169-1186
Article preview

☐ Review article ○ Abstract only
Review and evaluation of wake loss models for wind energy applications
Cristina L. Archer, Ahmadreza Vasel-Be-Hagh, Chi Yan, Sicheng Wu, ... A. Eoghan Maguire
Pages 1187-1207
Article preview

☐ Research article ○ Abstract only
Towards a comprehensive model for characterising and assessing thermoelectric modules by impedance spectroscopy
Ramy Mesalam, Hugo R. Williams, Richard M. Ambrosi, Jorge García-Cañadas, Keith Stephenson
Pages 1208-1218
Article preview

☐ Research article ○ Abstract only
Methane production through anaerobic digestion: Participation and digestion characteristics of cellulose, hemicellulose and lignin
Wanwu Li, Habiba Khalid, Zhe Zhu, Ruihong Zhang, ... Eva Thorin
Pages 1219-1228
Article preview

☐ Research article ● *Open access*
Thermodynamic optimisation of a high-electrical efficiency integrated internal combustion engine – Organic Rankine cycle combined heat and power system
Maria Anna Chatzopoulou, Christos N. Markides
Pages 1229-1251
Download PDF Article preview

VSI:ICAE 2017

☐ Research article ○ Abstract only
Highly energy-efficient combination of dehydrogenation of methylcyclohexane and hydrogen-based power generation
Firman Bagja Juangsa, Lukman Adi Prananto, Zahrul Mufrodi, Arief Budiman, ... Muhammad Aziz
Pages 31-38
Article preview

☐ Research article ○ Abstract only
Gasification process integration with existing combined heat and power plants for polygeneration of dimethyl ether or methanol: A detailed profitability analysis
Chaudhary Awais Salman, Muhammad Naqvi, Eva Thorin, Jinyue Yan
Pages 116-128
Article preview

☐ Research article ○ Abstract only
Distribution patterns of energy consumed in classified public buildings through the data mining process
Yibo Chen, Jianzhong Wu
Pages 240-251
Article preview

☐ Research article ○ Abstract only
Zero dimensional dynamic model of vanadium redox flow battery cell incorporating all modes of vanadium ions crossover
M. Pugach, M. Kondratenko, S. Briola, A. Bischi
Pages 560-569
Article preview

☐ Research article *Open access*
Design and characterization of hybrid III–V concentrator photovoltaic–thermoelectric receivers under primary and secondary optical elements
T.K.N. Sweet, M.H. Rolley, W. Li, M.C. Paul, ... A.R. Knox
Pages 772-783
Download PDF Article preview

☐ Research article ○ Abstract only
A solar methane reforming reactor design with enhanced efficiency
Jian Jin, Xin Wei, Mingkai Liu, Yuhang Yu, ... Yong Hao
Pages 797-807
Article preview

☐ Research article ○ Abstract only
Methane hydrate decomposition and sediment deformation in unconfined sediment with different types of concentrated hydrate accumulations by innovative experimental system
Yi Wang, Jing-Chun Feng, Xiao-Sen Li, Yu Zhang, Han Han
Pages 916-923
Article preview

☐ Research article ○ Abstract only
Multi-time period optimized configuration and scheduling of gas storage in gas-fired power plants
Zheng Qiao, Qinglai Guo, Hongbin Sun, Tongtian Sheng
Pages 924-934
Article preview

VSI:DEM 2018

☐ Research article ○ Abstract only
Two-stage energy management for networked microgrids with high renewable penetration
Dongxiao Wang, Jing Qiu, Luke Reedman, Ke Meng, Loi Lei Lai
Pages 39-48
Article preview

☐ Research article ○ Abstract only
Flexible operation of active distribution network using integrated smart buildings with heating, ventilation and air-conditioning systems
Tao Jiang, Zening Li, Xiaolong Jin, Houhe Chen, ... Yunfei Mu
Pages 181-196
Article preview

☐ Research article ○ Abstract only
A systems approach for management of microgrids considering multiple energy carriers, stochastic loads, forecasting and demand side response
Damian Giaouris, Athanasios I. Papadopoulos, Charalampos Patsios, Sara Walker, ... Panos Seferlis
Pages 546-559
Article preview

☐ Research article ○ Abstract only
Real-time renewable energy incentive system for electric vehicles using prioritization and cryptocurrency
Tianyang Zhang, Himanshu Pota, Chi-Cheng Chu, Rajit Gadh
Pages 582-594
Article preview

☐ Research article ○ Abstract only
Multi-objective distributed generation planning in distribution network considering correlations among uncertainties
Shenxi Zhang, Haozhong Cheng, Ke Li, Nengling Tai, ... Furong Li
Pages 743-755
Article preview

☐ Research article ○ Abstract only
Optimal placement, sizing, and daily charge/discharge of battery energy storage in low voltage distribution network with high photovoltaic penetration
Mohammad Rasol Jannesar, Alireza Sedighi, Mehdi Savaghebi, Josep M. Guerrero
Pages 957-966
Article preview

☐ Research article ○ Abstract only
A Generalized Droop Strategy for Interlinking Converter in a Standalone Hybrid Microgrid
Sarmad Majeed Malik, Yingyun Sun, Wen Huang, Xin Ai, Zhikang Shuai
Pages 1056-1063
Article preview

☐ Research article ○ Abstract only
Optimal planning of electric vehicle charging stations comprising multi-types of charging facilities
Lizi Luo, Wei Gu, Suyang Zhou, He Huang, ... Xiaobo Dou
Pages 1087-1099
Article preview

VSI:Energy Justice

☐ Research article ○ Abstract only
"They are grinding us into the ground" – The lived experience of (in)energy justice amongst low-income older households
Nicola Willand, Ralph Horne
Pages 61-70
Article preview

☐ Research article ○ Abstract only
Intermediating energy justice? The role of intermediaries in the civic energy sector in a time of austerity
M. Lacey-Barnacle, C.M. Bird
Pages 71-81
Article preview

☐ Research article *Open access*
The role of community acceptance in planning outcomes for onshore wind and solar farms: An energy justice analysis
Philippa Roddis, Stephen Carver, Martin Dallimer, Paul Norman, Guy Ziv
Pages 353-364
Download PDF Article preview

VSI:REM 2017

☐ Research article *Open access*
Peer-to-peer energy sharing through a two-stage aggregated battery control in a community Microgrid
Chao Long, Jianzhong Wu, Yue Zhou, Nick Jenkins
Pages 261-276
Download PDF Article preview

☐ Research article ○ Abstract only
Study on an innovative three-dimensional wind turbine wake model
Haiying Sun, Hongxing Yang
Pages 483-493
Article preview

☐ Research article ○ Abstract only
A new hybrid photovoltaic/thermal and liquid desiccant system for trigeneration application
Bosheng Su, Wei Han, Wanjun Qu, Changchun Liu, Hongguang Jin
Pages 808-818
Article preview

VSI:ECOS 2017

☐ Research article ○ Abstract only
A thermoacoustic Stirling electrical generator for cold exergy recovery of liquefied nature gas
Mingyu Hou, Zhanghua Wu, Guoyao Yu, Jianying Hu, Ercang Luo
Pages 389-396
Article preview

☐ Research article ○ Abstract only
Modeling and experimental investigation of a free-piston Stirling engine-based micro-combined heat and power system
Shunmin Zhu, Guoyao Yu, Jongmin O, Tao Xu, ... Ercang Luo
Pages 522-533
Article preview

☐ Research article ○ Abstract only
Modelling household spatial energy intensity consumption patterns for building envelopes, heating systems and temperature controls in cities
Javier Urquizo, Carlos Calderón, Philip James
Pages 670-681
Article preview

VSI:Data on Energy

☐ Research article ○ Abstract only
Rapid growth of petroleum coke consumption and its related emissions in China
Yuli Shan, Dabo Guan, Jing Meng, Zhu Liu, ... Zhifu Mi
Pages 494-502
Article preview

VSI:EFC 2017

☐ Research article ○ Abstract only
A numerical and experimental comparison of a single reversible molten carbonate cell operating in fuel cell mode and electrolysis mode
Juan Pedro Perez-Trujillo, Francisco Elizalde-Blancas, Massimiliano Della Pietra, Stephen J. McPhail
Pages 1037-1055
Article preview

VSI:City sustainability

☐ Research article ○ Abstract only
Carbon emissions and their drivers for a typical urban economy from multiple perspectives: A case analysis for Beijing city
Jia Shuo Li, H.W. Zhou, Jing Meng, Q. Yang, ... Y.Y. Zhang
Pages 1076-1086
Article preview

☐ Research article ○ Abstract only
Data-driven model predictive control using random forests for building energy optimization and climate control
Francesco Smarra, Achin Jain, Tullio de Rubeis, Dario Ambrosini, ... Rahul Mangharam
Pages 1252-1272
Article preview

☐ Research article ○ Abstract only
Brick : Metadata schema for portable smart building applications
Bharathan Balaji, Arka Bhattacharya, Gabriel Fierro, Jingkun Gao, ... Kamin Whitehouse
Pages 1273-1292
Article preview

Previous vol/issue
Next vol/issue

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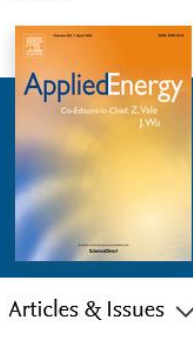

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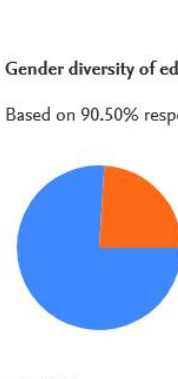

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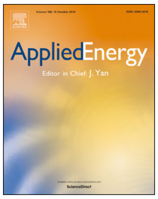

Highly energy-efficient combination of dehydrogenation of methylcyclohexane and hydrogen-based power generation

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HIGHLIGHTS

- Integrated system of MCH dehydrogenation and H_2 power generation is proposed.
- Enhanced process integration is adopted to realize high energy efficiency.
- The proposed system can realize very high energy efficiency of 58.9%.
- Compared to Graz cycle based system, the proposed system shows excellent efficiency.

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ABSTRACT

Hydrogen (H_2) has been well studied for its potential use in energy storage, which is particularly related with the intermittent characteristic of renewable energy sources. However, the gas form of H_2 at standard pressure and temperature (STP) poses a challenging problem in terms of storage, transportation, and low volumetric energy density. An effective and reversible method for H_2 storage is chemically bonded H_2 used in the toluene (C_7H_8)/methylcyclohexane (MCH, C_7H_{14}) cycle. This study investigates a power generation system from H_2 storage in MCH, involving the dehydrogenation process and the combined cycle as a power generation process. An adequate analysis of the heat circulation was performed through an enhanced process integration (EPI) to ensure the high energy-efficiency of the proposed system. A highly endothermic reaction of dehydrogenation was supplied by utilizing the energy/heat from air-fuel combustion to ensure the effective heat recovery of the system. The proposed system was analyzed through an adjustment of the main operating parameters, namely, the GT inlet pressure, GT inlet temperature, and the condenser pressure, to observe their effects on the efficiency of the system. It was found that these parameters have a significant influence on the system performance and provide the possibility of further improvement. Under optimum conditions, the proposed system can realize a very high system efficiency of 54.6%. Moreover, the proposed system is also compared to a Graz cycle-based system, which has been reported to achieve an excellent power generation cycle from H_2. This result implies that the proposed integrated system leads to a significantly higher power-generating efficiency. Numerically, the proposed system demonstrated a system efficiency of 53.7% under similar conditions as the Graz cycle based system, which achieved a system efficiency of 22.7%.

1. Introduction

Power generation from renewable energy sources has been studied for decades, and has been implemented at a large scale in many countries during recent years [1]. In addition, renewable energy is the fastest growing source of electricity generation, and it has been predicted that its share will increase to 39% by 2050 [2]. However, the power generation from renewable energy sources, particularly wind and solar, faces several challenges to a power grid operation, including an intermittent output and a mismatch between the power output and demand, resulting in grid instability and wasted energy during a period of oversupply [3].

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Electrical energy storage can be installed in the system to balance the energy between demand and supply, as well as store the surplus energy. Electrical energy storage in the form of chemical energy has been widely applied, such as through batteries, methane (CH_4), and hydrogen (H_2) [4]. Among such chemical storage types, H_2 has the best ratio of valence electrons to protons, and therefore, the energy gain per electron is quite high [5]. Energy storage achieved by converting excess electricity into H_2 through the water electrolysis (power-to-gas) process has been widely studied using well-established pilot plans [6,7]. In addition, the trends of decarbonization of fossil fuels and conversion of biomasses into H_2 have increased significantly owing to a high environmental concern and convenience [8,9]. The chemical energy per mass of H_2 (142 MJ kg^{-1}) is at least three-times higher than that of gasoline (44 MJ kg^{-1}) [5,10]. Because H_2 is one of the most abundant elements on Earth, H_2 has a high potential as an energy storage or energy carrier, and it is believed that the role of H_2 will increase in the future.

However, H_2, which is in gas form at standard pressure and temperature (STP), has been a challenging problem in terms of storage and transportation owing to its low volumetric energy density, which is only about 3 Wh L^{-1} [11]. Therefore, an effective storage method of H_2 will play a key role in H_2 technology development. There are many types of H_2 storage systems used with the general purpose of increasing the volumetric energy density of H_2. The compression of H_2 is one of the basic technologies applied to increase the molecule density, resulting in an increase in the volumetric energy density [12]. High-pressure tanks are required with a rated pressure of 200–450 bar. However, high-pressure containers have several significant disadvantages in terms of an additional pressure control required during depressurization and the safety risk of H_2 pressurization. Another method for H_2 storage is liquefaction, which is condensing the gas into a liquid, or even a solid, because both phases have significantly higher density than the gaseous phase [5,13,14]. However, a very low condensation temperature (-252 °C at 1 bar) is required, and cryogenic technology consumes a large amount of energy, and there are still many challenging problems with regard to liquefaction, including super-insulated low-temperature storage methods [5,13].

Many studies related to the effective chemical storage of H_2 have recently been conducted [15,16], including the use of organic materials and ammonia. Unfortunately, ammonia is poisonous and has a pungent odor [17]. The utilization of ammonia via combustion, such as gas turbine, has several problems including lower reactivity of ammonia and release of NO_x. Regarding the latter, the formation of NO_x increases significantly when the combustion temperature reaches about 1500 °C following Zeldovich mechanism (thermal NO_x). Therefore, as NO_x is a pollutant (GHG), direct utilization of ammonia via combustion is not environmental friendly. Although there are several technologies which can reduce the NO_x formation, however, they are still under development. In addition, in ammonia-based H_2 chemical storage system, N_2 is released during the dehydrogenation process, creating one-way transport from the site of H_2 production to the site of H_2 utilization. On the other hand, toluene in liquid phase as the result of dehydrogenation of MCH, will be returned to the hydrogenation plant and reused, allowing sustainable cycle of H_2 storage system.

Chemical H_2 storage can be applied by binding H_2 to produce H_2-rich molecules in a catalytic hydrogenation reaction [15]. To create a sustainable H_2 storage system, at least two processes, namely, H_2-rich molecule formation (hydrogenation) and H_2 release (dehydrogenation), are required. Numerous molecules can be utilized in chemical storage, which can be distinguished into two main categories: (1) natural H_2-lean molecules that can be extracted from an exhaust gas mixture such as CO_2 or N_2, and (2) a H_2-lean organic liquid, which allows a fully reversible cycle of hydrogenation/dehydrogenation. The latter is commonly referred to as liquid organic H_2 storage (LOHC) [16,15].

LOHC technology has been widely studied, with an option of different organic material pairs such as methylcyclohexane (MCH, C_7H_{14})-toluene(C_7H_8), cyclohexane-benzene, decalin-naphthalene, and dibenzyl-toluene. Among the first three pairs, MCH-toluene is preferable for easier storage and transportation owing to its wide temperature range under a liquid state [18,19]. Among the available LOHCs, MCH-toluene and dibenzyl-toluene have a relatively high H_2 content of 6.2% [20]. A dibenzyl-toluene pair has been recently reported with a focus on the dynamism of the system in supplying electricity [21]. However, this work remains at the laboratory scale, without a sufficient analysis at larger scales, and is designed for fuel cell application. In contrast, the MCH-toluene cycle has been evaluated at the pilot scale by a Japanese company, and has been demonstrated to be effective [18]. Gaseous H_2 is chemically bonded to toluene through hydrogenation forming liquid MCH [22]. Transportation and storage are the main features of MCH with a high boiling point, which make it a potentially safe medium for an H_2 carrier. This is also very promising because up to 6–8 wt% of H_2, or 60–62 kg m^{-3} (volume based under ambient conditions), can be stored [15,23]. Toluene as a raw material has been widely produced and utilized industrially, and provides a low-cost material for large-scale processes [23]. In addition, both toluene and MCH are in a liquid phase over a wide range of temperatures, which is favorable for long-term storage. At the industrial scale, in 2013, the Chiyoda Corporation began successfully operating a large-scale H_2 storage and delivery system by utilizing toluene-MCH as an H_2 carrier using a K-promoted Pt/Al_2O_3 catalyst [18]. Therefore, the toluene-MCH cycle is theoretically promising as an H_2 carrier, and is practically applicable at an industrial scale.

To gain energy from H_2 bonded in MCH, H_2 must be separated from toluene through a dehydrogenation process. The extracted H_2 can be converted into electrical energy through thermal energy (a combined cycle) or chemical energy (a fuel cell). Numerous studies have been carried out to develop an efficient dehydrogenation process and electricity generation from MCH. Scherer et al. developed a seasonal electricity generation from MCH by employing solid oxide fuel cells (SOFCs) [24]. However, despite exhibiting high energy efficiency, SOFCs have very fragile characteristics owing to the reformation-based H_2 used for the fuel [25]. Most studies on fuel cells have a common challenge in terms of the inability to provide a large power output [25,26]. It has been reported that, for power units with a capacity greater than 10 MW, steam-turbine-based units are preferable over fuel cell power units [27]. An H_2-fueled combustion turbine cycle (HFCTC) is expected to be a new energy source for the power sector, and certain countries, including Japan, have started its development [28]. To confirm its operability, a number of turbine manufacturers have reported studies on HFCTC, both numerically and experimentally, including H_2-fueled burners [28,29]. Milewski et al. investigated the utilization of H_2 as a fuel based on a combined cycle concept with various plant utility configurations, and successfully achieved 60% energy efficiency [28]. Among these cycles, the Graz cycle has been developed further with an improved net efficiency of greater than 65% [26]. However, most of the combined cycles, including the Graz cycle, employ pure O_2, resulting in additional utilities and energy required for O_2 separation from the air, leading to high-cost plants and a drop in efficiency of nearly 61% [26]. Moreover, the above studies have disregarded the hydrogenation process, and assumed that the H_2 feed is in a pure phase, which is very difficult to achieve in a real operation. Regarding the large-scale production of MCH, Aziz et al. developed novel integrated concepts of large-scale MCH production from both low-rank coal [11] and brown coal [30] by applying chemical looping and hydrogenation using toluene. These concepts have achieved high values of H_2 production while maintaining a clean technology for the environment. However, no evaluation regarding the dehydrogenation and utilization of H_2 from MCH has been conducted.

To the best of the authors' knowledge, few investigations have addressed the concept of energy-efficient electricity production from H_2 through MCH as a storage method. In this paper, we therefore propose the concept of an electricity generation plant, which is an integrated system consisting of dehydrogenation and combined cycle systems. The dehydrogenation of MCH is integrated with a combined hydro-fuel cycle, which employs ambient air for combustion. Therefore, the proposed system is more realistic as it is based on a comprehensive energy balance analysis of the cycle. An adequate analysis of the heat circulation has been conducted through enhanced process integration (EPI) to ensure the high energy-efficiency of the proposed system.

2. Process modeling

2.1. Conceptual model

The proposed integrated-system was designed and optimized based on the principles of EPI in order to significantly reduce the exergy loss throughout the integrated system. EPI focuses mainly on the heat circulation optimization throughout the systems by promoting an effective exergy recovery and process integration, leading to minimum exergy loss [31]. Exergy recovery is initially performed in each single process in order to achieve the optimum heat recovery through an exergy rate elevation and heat coupling [32]. In addition, the unrecoverable heat from any process is utilized further in other processes. Therefore, the unrecoverable heat can be minimized, resulting in minimum energy waste into the environment, and a high energy-efficiency of the system. Such technology has been applied to several processes, including drying [33], H_2 production from algae [34], and biomass gasification [35].

Fig. 1 shows a conceptual diagram of the proposed integrated system. The system consists of three combined modules: MCH dehydrogenation, H_2 combustion, and a combined cycle power generation. The solid, dotted, and dashed lines represent the material, heat, and electricity flows, respectively.

One of the main processes of the systems is the highly endothermic dehydrogenation reaction of MCH, which contains a 6.2 wt% capacity of H_2. MCH is supplied and transported from the hydrogenation plant using a large-sized vessel or pipeline. A packed bed reactor is employed in this module, and equipped with a catalyst to improve the reaction. Thermal energy for a dehydrogenation reaction is supplied mainly from recovered heat through pre-heating and combustion module exhaust before its remaining heat is converted into electrical energy through a combined cycle module.

As the main product of the dehydrogenation module, H_2 is further fed to the combustion module, whereas the separated toluene is returned to the hydrogenation plant for another cycle to be utilized as a carrier substance. In the combustion module, H_2 as a fuel is reacted with O_2 from the air, converting the chemical energy of H_2 into thermal energy. To obtain a reasonable combustion temperature, particularly the designated gas turbine inlet temperature, gas and steam discharged from the combined cycle are supplied back to the combustion chamber, cooling the burners and liners, and also controlling the fuel gas temperature. Exhaust gas, consisting of high-pressure and high-temperature steam and gas, leaves the combustion chamber under a pre-determined temperature condition.

The last module, the combined cycle module, converts the thermal energy of the exhaust gas into electricity by utilizing the combination of gas and steam turbines. The hot exhaust gas from combustion is utilized initially as a heat source for the packed bed reactor to conduct dehydrogenation through a heat exchanger immersed inside the reactor, and sequentially flows to the combined cycle module for expansion.

2.2. Detailed system

In this section, a detailed design of the system is explained based on the process flow diagram shown in Fig. 2. The proposed system was evaluated through a theoretical calculation and software modeling using Aspen HYSYS ver. 8.8 (Aspen Technology, Inc.). To establish the model of the system, the following additional assumptions were employed during the calculations:

(i) The rated flow rate of MCH is 100 t h^{-1}.
(ii) Heat exchangers, including HRSG, are a counter-flow type.
(iii) The ambient pressure and temperature are 101.33 kPa and 25 °C, respectively.
(iv) The kinetic and potential energy losses are negligible.
(v) Air consists of 79 mol% N_2 and 21 mol% O_2.
(vi) The adiabatic efficiency of the pump and compressor is 90%.
(vii) The pressure drop in the heat exchanger is 2%.
(viii) The minimum approach temperature in the heat exchanger is 10 °C.
(ix) There are no external heat losses.

2.2.1. MCH dehydrogenation

The dehydrogenation module employs a packed bed reactor, and a catalyst is used inside the reactor to improve the following reaction. In addition, the pressure drop across the reactor is approximated using the following Eq. (2) [36]:

$$C_7H_{14} \rightarrow C_7H_8 + 3H_2 \quad \Delta H = 204.6\ \mathrm{kJ\ mol^{-1}} \tag{1}$$

$$\Delta P = \frac{12.5(1-\varepsilon)^2}{\varepsilon^3} \times (29.32Re^{-1} + 1.56Re^{-n} + 0.1)\frac{\rho u^2}{2}\frac{1}{d_p} \tag{2}$$

Among the various hydrogenation catalysts, a Pt catalyst has been studied by a number of researchers [22,37,38]. A theoretical review on the reaction kinetics and experimental results have shown that Pt/Al_2O_3 is considered the best catalysts for MCH dehydrogenation in terms of activity, selectivity, and stability [22,38]. Dehydrogenation is an endothermic reaction, and therefore the required thermal energy is initially supplied through a self-heat exchange between the cold stream (process stream, F3) and hot stream (combustion flue gas, F9). Toluene, as an H_2 carrier, is returned to a hydrogenation plant (F17) for another cycle as an H_2 storage system.

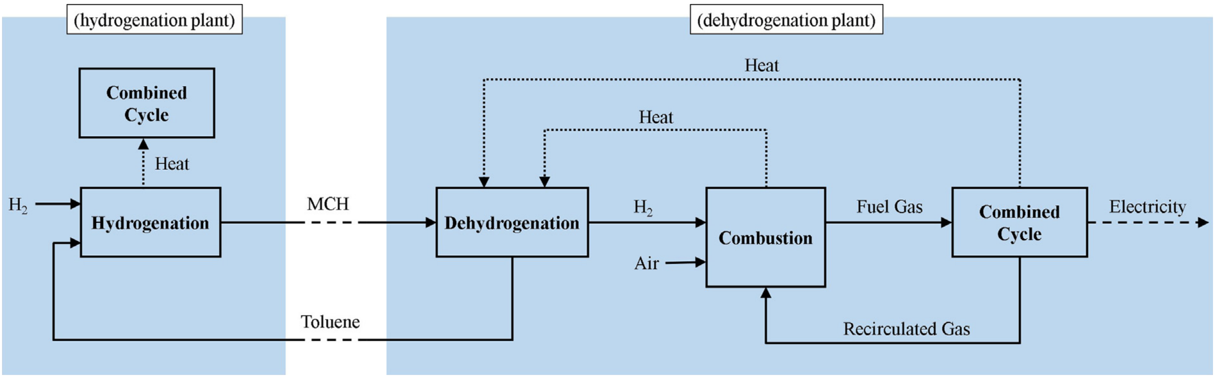

Fig. 1. Conceptual diagram of proposed integrated system.

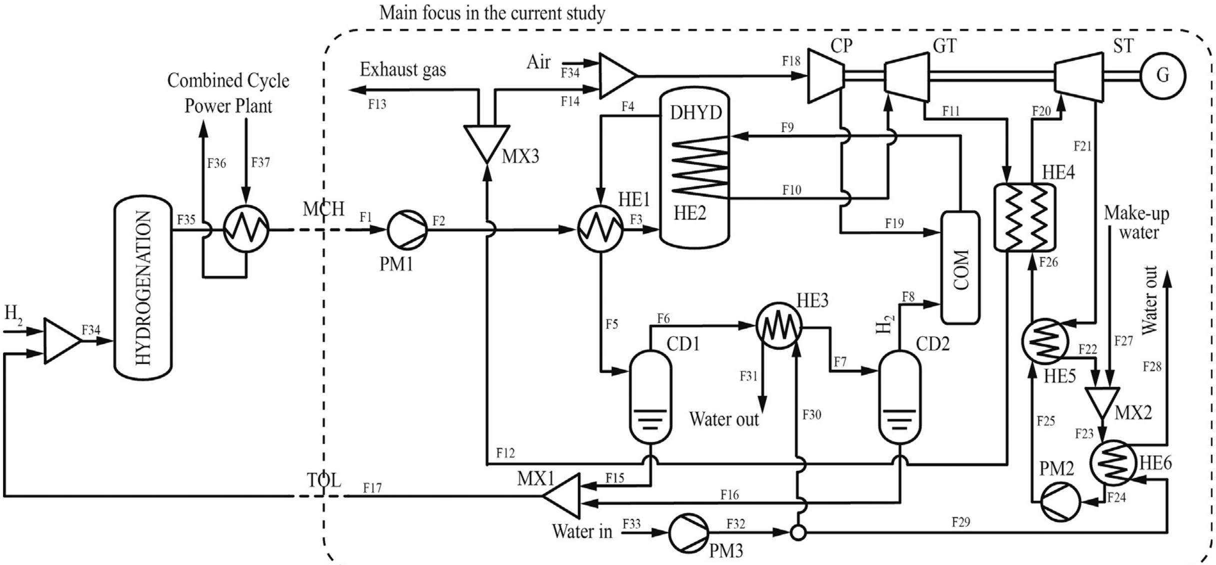

Fig. 2. Arrangement and process flow diagram of the proposed integrated system.

2.2.2. Combustion

H_2 produced from the dehydrogenation module is fed into the combustion chamber to be reacted with O_2 included in the air, converting the chemical energy of H_2 into thermal energy. Furthermore, thermal energy produced from a combustion reaction is utilized for producing the electricity in the combined cycle module, and the remaining heat is utilized to provide the thermal energy required for an endothermic reaction of dehydrogenation. Owing to the carbon-free combustion process of H_2, carbon dioxide (CO_2), which well known to be the main contributor of greenhouse gasses, is not emitted, as shown in reaction (3). In this study, after dehydrogenation, the gas and liquid phase substance is separated into separators (CD1, CD2) and cooled through water cooling (F30). Owing to the extremely different boiling temperature, H_2 is discharged in a gas phase, whereas toluene is discharged in a liquid phase. However, small portions of toluene in the gas phase remain and are mixed with H_2, flowing into the combustion chamber. Therefore, a very small amount of CO_2 production may occur, as shown in reaction (4).

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \quad \Delta H = -241.8\ \text{kJ mol}^{-1}(LHV) \tag{3}$$

$$C_7H_8 + 9O_2 \rightarrow 7CO_2 + 4H_2O \quad \Delta H = -3920\ \text{kJ mol}^{-1} \tag{4}$$

2.2.3. Combined cycle for electricity generation

The combined cycle consists of three main components: a gas turbine (GT), steam turbine (ST), and heat recovery steam generator (HRSG). EPI is applied throughout the system, including the heat circulation during the dehydrogenation, and the combined cycle to enhance the optimum heat circulation of the system and achieve a high energy-efficiency. Table 1 summarizes the conditions and assumptions for each module of the dehydrogenation and combined cycle.

In this study, the combined cycle is employed with a modification of the Graz cycle, whereas the exhaust gas from HRSG (F14) is partially returned to the combustion chamber for cooling, as well as to improve the energy density owing to the high heat capacity of water. Originally, the Graz cycle utilizes high-pressure steam from a high-pressure steam turbine to be fed into the combustion chamber for cooling [26,40]. However, in this study, the gas exhausted from the HRSG outlet is used instead of high-pressure steam, leaving the steam expansion process in the ST for optimum electrical generation. The HRSG outlet gas also has a lower temperature, providing an effective cooling process in the combustion chamber.

Table 1
Assumed conditions for each module of the dehydrogenation and combined cycle.

Parameter	Value	Symbol (Refer to Fig. 2)
Dehydrogenation (DHYD)[18,21,22,39]		
Reactor temperature (°C)	450	F4
Internal pressure (kPa)	120–150	DHYD
Catalyst	Pt/Al_2O_3	–
Fuel temperature before separation (°C)	70	F7
Minimum toluene discharge ratio	90%	F17
Power generation – Gas turbine (GT) [11,26]		
Isentropic efficiency (%)	90	–
Inlet temperature (°C)	1400–1600	F10
Inlet pressure (MPa)	2.8–4.0	F10
Power generation – Steam turbine (ST) [11,26]		
Isentropic efficiency (%)	90	–
Inlet pressure (MPa)	10	F20
Steam turbine inlet temperature (°C)	522–581	F20
Minimum vapor quality	0.9	–

3. Results and discussion

The performance of the proposed integrated system was evaluated through the optimization of several key operating parameters with regard to a detailed system performance, and the energy efficiency of the system was compared with other similar systems. The performance of the system was evaluated by determining the system efficiency (η_{power}) as follows:

$$\eta_{system} = \frac{W_{net}}{(\dot{m}_{MCH} \times LHV_{MCH} - \dot{m}_{Toluene} \times LHV_{Toluene})} \tag{5}$$

where $\dot{m}_{MCH}$, $\dot{m}_{Toluene}$, LHV_{MCH}, and $LHV_{Toluene}$ are the mass flow rate of MCH, mass flow rate of toluene, LHV of MCH, and LHV of toluene, respectively. The generated net power as the final result of this system is compared with the difference in calorific value between MCH, as an input material, and toluene, as an output material. The net generated power (W_{net}) is calculated based on the total power generated by the GT (W_{GT}) and ST (W_{ST}) with a consideration of the power consumed by auxiliaries (W_{Aux}), which is shown as follows:

$$W_{net} = W_{GT} + W_{ST} - W_{Aux} \tag{6}$$

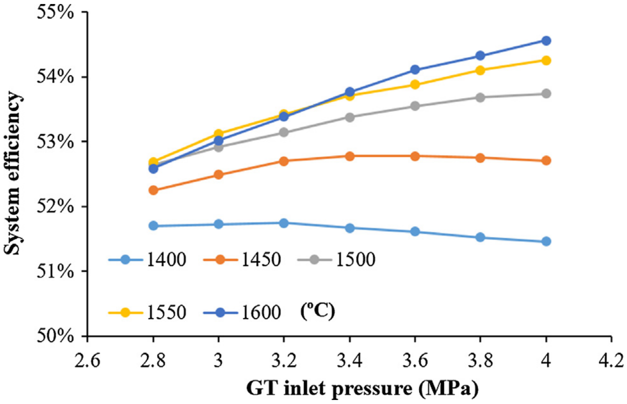

Fig. 3. System efficiency as a function of GT inlet pressure under different GT inlet temperatures.

3.1. Detailed system performance

The effects of various operating conditions, including the GT inlet pressure, GT inlet temperature, and ST inlet pressure, on the overall power generation and its efficiency were considered in this study. Calculations using Aspen HYSYS were utilized to investigate the required objectives.

3.1.1. GT inlet pressure

Fig. 3 shows the effects of the variations in GT inlet pressure on the system efficiency under different GT inlet temperatures. It has been shown that the system efficiency increases following an increase in GT inlet pressure. Numerically, the highest system efficiency, which is 54.6% (net generated power of 132 MW), is achieved under a GT inlet pressure and temperature of 4 MPa and 1600 °C, respectively. This system efficiency decreases to 52.6% (net generated power of 128 MW) when the GT inlet pressure is decreased to 2.8 MPa under the same GT inlet temperature.

In detail, power generated by GT and ST increases at a higher pressure of the fuel gas owing to higher enthalpy. However, the power consumed by the compressor also increases, resulting in a compensation relationship between the GT inlet pressure and the compressor power consumption. At high temperature, the amount of additional power generated by a pressure increase is relatively higher than that achieved through an additional compressor application. However, at a lower temperature, the compressor operation increasing the gas pressure is higher than the total generated power, resulting in a decrease in the system efficiency. To decrease the GT inlet temperature, more re-circulated gas (F14) is required, causing an increase in the gas flow rate, and leading to a higher compressor load. The GT inlet pressure, shown as F10 in Fig. 2, is determined based on the compression ratio of the compressor (CP). The influence of the compression ratio on the combined cycle performance has been reported well in other similar arrangements of the combined cycle [26,41]. The energy required for compression significantly affects the net power generated by the combined cycle module.

3.1.2. GT inlet temperature

Fig. 4 shows the influence of the GT inlet temperature on the system efficiency under different GT inlet pressures. In general, the GT inlet temperature strongly influences the system efficiency. At a GT inlet pressure of 4 MPa, the system efficiency increases from 52.7% (net generated power of 127 MW) to 54.6% (net generated power of 133 MW) when the GT inlet temperature is increased from 1400 °C to 1600 °C.

The GT inlet temperature is determined through a combustion reaction, which produces a very high-temperature flue gas, and by the temperature of the recycled gas from the HRSG outlet. The system efficiency increases with a higher GT inlet temperature. A higher gas temperature has a higher exergy rate leading to larger energy/heat, which can be recovered by GT. Moreover, a higher GT inlet temperature results in less re-circulated gas, leading to a lower energy required for compression. However, as mentioned previously, the system performance is significantly influenced by the GT inlet pressure, which determines the amount of gas flow. A high GT inlet temperature causes a lower gas flow, affecting the amount of power generated by ST. The ST power is the compensation between the flue gas temperature and flow, as can be observed at a GT inlet pressure of 2.8 MPa, where system efficiency decreases when the GT inlet temperature reaches above 1550 °C.

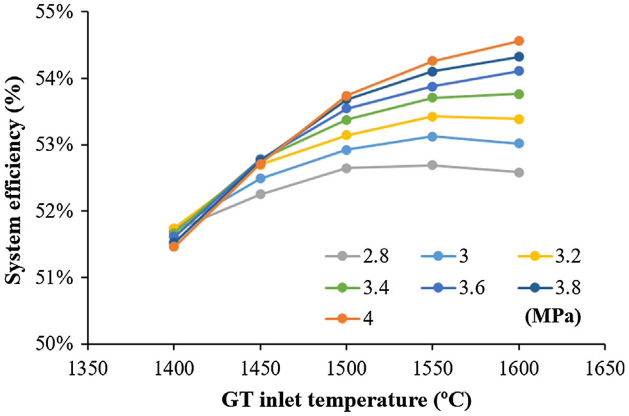

Fig. 4. System efficiency as a function of GT inlet temperature under different GT inlet pressures.

3.1.3. Condenser pressure

Another operating parameter that is believed to significantly influence the performance of the combined cycle is the condenser (HE6) pressure. Based on the thermodynamic curve, the condenser pressure, as the lowest pressure in the module, greatly affects the ST output. Fig. 5 shows the condenser pressure effect on the system efficiency at a GT inlet pressure and temperature of 4 MPa and 1500 °C, respectively. In general, the system efficiency increases following the lower pressure (more vacuum condition) of the condenser. The system efficiency increases from 53.7% to 52.9% when the condenser pressure is decreased from 85 to 35 kPa.

The condenser temperature is directly affected by the pressure owing to the saturated condition in the condenser. The lower pressure in the condenser can provide a lower temperature of ST expansion, which based on a Carnot cycle improves the system performance [42].

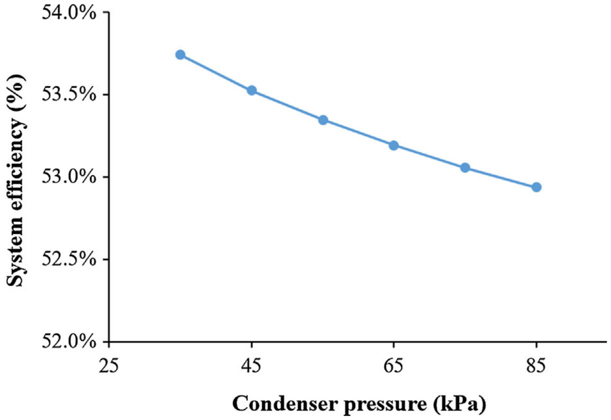

Fig. 5. System efficiency as a function of condenser pressure (GT inlet pressure and temperature of 4 MPa and 1500 °C, respectively).

Table 2
Conditions of major flows under optimum operation (GT inlet pressure of 4 MPa, GT inlet temperature of 1600 °C, and condenser pressure of 35 kPa).

Stream	Flow ($t\,h^{-1}$)	Pressure (kPa)	Temperature (°C)	Stream	Flow ($t\,h^{-1}$)	Pressure (kPa)	Temperature (°C)
F1	100	101.3	25	F11	597	150	710
F2	100	152	25	F12	597	147	80
F3	100	149	368	F17	85.3	140	37
F4	100	146	450	F19	597	4040	632
F5	100	143	70	F20	157	20,000	550
F7	78	140	25	F21	157	35	73
F8	14.7	140	25	F24	157	34	40
F9	597	4040	1845	F25	157	20,050	41
F10	597	4000	1600	F26	157	20,030	65

3.1.4. Optimum system condition

The optimum system condition is determined based on the main parameters, and was analyzed in the previous sections as resulting in the maximum system efficiency. Table 2 shows the mass flow rate, pressure, and temperature of the major flows, which represent the system performance under an optimal operation (GT inlet pressure of 4 MPa, GT inlet temperature of 1600 °C, and condenser pressure of 35 kPa).

3.2. Performance comparison with Graz cycle-based power generation

In terms of H_2 utilization when using steam turbine cycles, a number of studies have been reported under various configurations [28]. However, they have focused particularly on power generation from H_2. In this study, we compared our proposed system with another system, including the dehydrogenation process in each system. The power generation efficiency, described in Eq. (5), was compared for each configuration because it represents the overall performance, including the dehydrogenation process.

Table 3 shows the main parameters and system performance of the proposed integrated system compared with the Graz cycle, which is integrated with the dehydrogenation process (Graz cycle-based system), for a comprehensive analysis. The dehydrogenation process in the Graz cycle-based system is calculated through a stoichiometric reaction shown in Eq. (1). The amount of MCH and toluene are calculated based on the H_2 mass flow rate, with a toluene discharge ratio of 0.9. Although each system has a different rated power output, the system efficiency represents the overall system performance.

The Graz cycle employs oxy-fuel combustion, in which pure O_2 is supplied from an air separation unit (ASU) and through the O_2 compression process. Therefore, an additional power consumption of 24.3 MW is supplied from the generated power. Our proposed integrated system utilizes air-combustion, eliminating the requirement of ASU and an additional compression system. The power generation efficiency (η_{power}) can be calculated for each system based on the following equation:

$$\eta_{power} = \frac{W_{net}}{(\dot{m}_{H_2} \times LHV_{H_2})} \quad (7)$$

where W_{net}, $\dot{m}_{H_2}$, and LHV_{H_2} are the net power generation, mass flow rate of H_2, and LHV of H_2, respectively.

Compared to the Graz cycle-based system (with a power generation efficiency of 60.37%), as shown in Table 3, the integrated system proposed in this study shows a higher power generation efficiency of 63.7%. This may indicate that an air–fuel combustion cycle is more efficient than an oxy–fuel combustion cycle in terms of the total energy efficiency owing to the excessive power consumption of ASU. Furthermore, Sanz et al. proposed the use of the Graz cycle as an efficient power generation under the assumption that high purity O_2 is available as a by-product from the H_2 production through electrolysis [26]. Therefore, in an integrated H_2 storage and power generation system, in which O_2 is unavailable within the process, as proposed in the present study, air–fuel combustion is considered to be more effective. In addition, air–fuel combustion has been well applied in a typical combined cycle plant with fewer components, providing a low initial power plant cost.

In this study, an integrated power generation system achieved through H_2 storage (MCH) is proposed. The overall system performance was analyzed by comparing the system efficiency (η_{system}) for both a Graz cycle-based system and the proposed integrated system. The dehydrogenation process in the Graz cycle-based system was calculated stoichiometrically based on the reaction shown in Eq. (1). Moreover, the energy required for the endothermic reaction of dehydrogenation was calculated under the assumption that the dehydrogenation system was designed based on conventional heat recovery technology for minimum exergy loss.

The proposed integrated system was shown to have a significantly higher efficiency of 53.7% compared to the Graz cycle-based system, at 23%. The system efficiency of the Graz cycle-based system is lower due to an extensive energy demand following an endothermic reaction of dehydrogenation. In addition, in the proposed integrated-system, the hot flue gas after combustion (F9) has a higher temperature (1722 °C)

Table 3
Comparison of the main parameters and integrated system performance of Graz cycle based system [26] and the proposed integrated-system.

	Graz cycle-based system [26]	Proposed integrated system	Unit
Conditions			
Combustor outlet temperature	1500	1722	°C
GT inlet temperature	1500	1500	°C
GT inlet pressure	4	4	MPa
ST inlet pressure	17	20	MPa
Condenser pressure	0.0025	0.035	MPa
Minimum steam quality	0.893	0.9	
Power Generation			
GT power output	231.8	235.4	MW
ST power output	52.2	41.7	MW
Auxiliary power (pumps, etc.)	78.6	146.4	MW
ASU + O_2 compression	24.3	–[a]	MW
Net power	205.4	130.7	MW
H_2 LHV	120	120	MJ/kg
H_2 mass flow	2.5	1.71	kg/s
Power generation efficiency	**60.4%**	**63.7%**	
Dehydrogenation			
MCH mass flow	147,000	100,000	kg/h
MCH LHV	43.4	43.4	MJ/kg
Toluene mass flow	124150.5	85,298	kg/h
Toluene LHV	40.6	40.6	MJ/kg
Energy required for dehydrogenation	99.9[b]	–[b]	MW
System Efficiency	**22.7%**[b]	**53.7%**	

[a] ASU and O_2 are not required owing to the air-fuel combustion type in the proposed model

[b] The dehydrogenation process for the Graz cycle was calculated based on a stoichiometric reaction, using a conservative heat recovery system.

Table 4
Typical condition of hydrogenation plant.

Parameter	Value	Symbol (Refer to Fig. 2)
Pressure (kPa)	130	–
Temperature (°C)	200	F20
Catalyst	Ni-Mo/Al_2O_3	F20
Catalyst particle size (mm)	0.3	–
Sphericity (–)	0.5	

than that of the Graz cycle-based system (1500 °C). High-temperature gas is used as the thermal energy source for a reaction, reducing the exergy loss from the system, and thereby increasing the overall system efficiency.

In addition, further improvement can be achieved in the proposed system by decreasing the condenser pressure. As shown in Fig. 5, the system efficiency increases at a lower condenser pressure. The condenser pressure of the proposed integrated system is set to 35 kPa, although it can be decreased further to as low as that of the Graz cycle-based system (2.5 kPa). With a lower condenser pressure, it is expected that the proposed integrated system can achieve higher power generation efficiency, and thus higher system efficiency.

3.3. Overall performance including MCH production (hydrogenation)

MCH production is carried out in hydrogenation plant, where H_2 is bonded with toluene, resulting in the MCH formation. Hydrogenation is an exothermic reaction, producing heat as one of results of reaction. As mentioned above, theoretically, MCH has volumetric and gravimetric H_2 contents of 47% and 6.2%, respectively. Hydrogenation process including the catalyst used in the reaction is considered a well-established technology with the reaction as follow,

$$C_7H_8 + 3H_2 \rightarrow C_7H_{14} \quad \Delta H = -204.6 \text{ kJ mol}^{-1} \tag{8}$$

The produced heat from hydrogenation reaction can be recovered by other systems that are coupled with hydrogenation plant. Aziz and his co-workers have developed and reported several integrated H_2 production and power generation systems from microalgae [34], low rank coal with syngas chemical looping [11], and brown coal with direct chemical looping [30]. In their developed system, H_2 which is produced from the system, was bonded with toluene in hydrogenation plant. The remaining syngas is utilized as fuel gas in the combined cycle power plant to generate electricity. The excess heat produced from hydrogenation reaction is recovered for preheating the cold stream flow of combined cycle, reducing the exergy loss of the system. Therefore, highly energy-efficient system can be achieved.

Typical condition of hydrogenation process is presented in Table 4. Hydrogenator consists of a mixer, reactor, and heat exchanger. Fixed bed reactor is employed with catalyst loaded inside the bed.

In case of coal as the primary resource for H_2 production, based on [11,30], the total energy efficiencies, including produced H_2 which is hydrogenated in MCH and generated power, for each syngas and direct chemical looping are 84% and 91%, respectively. Combining those studies and the obtained result in the current study, the overall energy efficiency from primary energy source (coal) to the generated power at the utilization site after dehydrogenation is about 46–49%. This is considered very high and can be achieved due to effective heat circulation through EPI.

4. Conclusion

An integrated system using dehydrogenation and a power generation process was proposed to gain energy from chemically bonded H_2 in MCH. MCH-toluene has been well reported as a promising H_2 storage system, and has been practically applied at an industrial scale. A highly endothermic dehydrogenation reaction is covered by the heat generated from the air-fuel combustion of H_2. Moreover, the remaining heat is supplied to the combined cycle plant, and optimized using enhanced process integration (EPI) to reduce the exergy loss, leading to a highly efficient system.

A comparison between the proposed integrated-system and a Graz cycle-based system was carried out to evaluate the overall system performance. The results show that the proposed integrated system can provide energy for the dehydrogenation process while maintaining a highly efficient combined cycle as the power generation system. The power generation efficiency of the proposed integrated system is higher than that of the Graz cycle-based system, mainly owing to the air-fuel combustion, which requires less energy and fewer components. With a higher combustion temperature and heat recovery optimization, the proposed integrated system has a significantly high system efficiency of 53.7%, compared to that of the Graz cycle based-system, at 23%.

Acknowledgment

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