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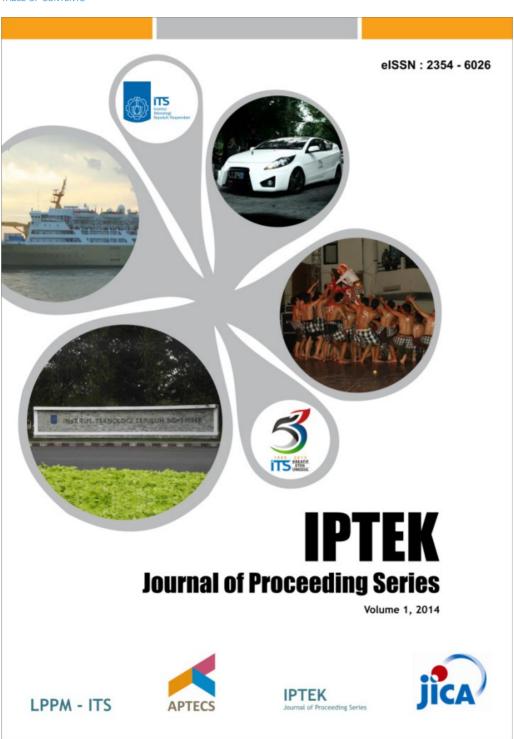
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ANNOUNCEMENTS EDITORIAL BOARD ONLINE SUBMISSION

Home > Archives > Vol 1, No 1 (2014)

Vol 1, No 1 (2014)

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TABLE OF CONTENTS



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Home > No 6 (2019)

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No 6 (2019): The 1st International Conference on Global Development (ICODEV)

TABLE OF CONTENTS







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The 1st International Conference on Global Development (ICODEV)

Surabaya, 19 November 2019

Editors:

Hermanto Arfan Fahmi Lienggar Rahardiantino

DEPARTMENT OF DEVELOPMENT STUDIES
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Home > Archives > Vol 1, No 1 (2014)

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Table of Contents

Articles

Development of a 'Fish Tail' Rudder to Improve a Ship's Maneuverability in Seaway

Aries Sulisetvono

Maximum Peak-Gain Margin 2DOF-IMC Tuning for a 2DOF-PID Filter Set Point

Controller Under Parametric Uncertainty

Nur Hidayah, Juwari Juwari, Renanto Handogo <u>Design of Curriculum Matrix for Robotics Education Derived from Bloom's Taxonomy</u> and Educational Curriculum of 2013

Eko Henfri Binugroho, Endah Suryawati Ningrum, Dwi Kurnia Basuki, Adnan Rachmat Anom Besari

Viscous-Resistance Calculation and Verification of Remotely Operated Inspection

Submarine

Ardi Nugroho Yulianto, I Ketut Suastika, Aries Sulisetyono Bioremoval of Chromium, Copper and Cadmium by Bacillus cereus in Simulated

Electroplating Wastewater

Yulinah Trihadiningrum

An Investigation into Hybrid Catamaran Fishing Vessel: Combination of Diesel Engine, Sails and Solar Panels

Pramudya Imawan Santosa, I Ketut Arya Priya Utama Synthesis of UiO-66 Using Solvothermal Method at High Temperature

Ika Diah Rahmawati, Ratna Ediati, Didik Prasetyoko

On-line State Estimator for Three Phase Distribution Networks Displayed on

Geographic Information System

Indri Suryawati, Ontoseno Penangsang, Adi Suprijanto, Dimas Fajar U. P., Mat

Syai'in

Impacts of River Groove Propagation on Irrigation Infrastructural Failures

Kuntjoro Kuntjoro, Choirul Anwar, Didik Harijanto

Steering System Kinematic and Steady-State Cornering Analyses of the ITS Electric Car

Unggul Wasiwitono, Indra Sidarta, Agus Sigit Pramono, Sutikno Sutikno, Alief

The Rate of Turbulent Kinetic Energy Dissipation in a Turbulent Boundary layer on a

Flat Plate

Sutardi Sutardi

The Influence of Burning Temperature of MIRHA On Mechanical Properties of Foamed

Ridho Bayuaji

System Application of Genetic Algorithm for Scheduling Optimization Study Using

Java (Case Study: Department of Computer System UNTAN)

Fatma Agus Setyaningsih

Simulation of Direct Digital Synthesizer with LabView

Elan Djaelani Suardja

Technology Development of Kupang Processing with Dryer Machine Base on Expert System

Dedin F Rosida, Dedid Cahya Happyanto

Kinetics of Hg and Pb Removal in Aqueous Solution Using Coal Fly Ash Adsorbent

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PDF

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PDF

PDF

PDF

PDF

PDF

1/5

Eko Prasetyo Kuncoro, Mochammad Zakki Fahmi	
Microstructures Behavior in Biomedical Co-Cr-Mo-C-Si-Mn Alloys Containing Nitrogen	PDF
Alfirano Alfirano New Concept of PQR-Coordinate on Power Transformation for Harmonics Reducing	PDF
Indriarto Yuniantoro, Rudy Setiabudy, Ridwan Gunawan	
The Effect of High Switching Frequency on Inverter Against Measurements of kWh-	PDF
Meter Isdawimah Isdawimah, Rudy Setiabudy, Ridwan Gunawan	
Analysis of Buttering Method on Mechanical Properties Welded Material Low Carbon	PDF
<u>Steel</u>	
Gathot Dwi Winarto, Rivai Wardhani, Irsyadus Syarif Vibration and Cylinder Chamber Pressure Characteristics of an Air Reciprocating	PDF
Compressor	<u>FDF</u>
Bambang Daryanto Wonoyudo, David A. Siregar	
The Effect of Chemical Substance and Immersion Time to Tensile Strength of	PDF
Bamboo Betung (Dendrocalamus asper) as Chemical Preservation Treatment Afif Rizqi Fattah, Ken Ninez Nurpramesti Prinindya, Hosta Ardhyananta	
Optimal Location of a Capacitor Bank in a Primary Feeder to Load up Linearly	PDF
Hermagasantos Zein, Erwin Dermawan	
Power of Regenerative Braking with Kinetic Energy Recovery System	PDF
Mochamad Edoward Ramadhan, Harus Laksana Guntur The Art and Sustainable Aspects of Natural Dyeing in KANAWIDA Hand Drawn Batik	PDF
(Green Batik)	PDF
July Hidayat, Fatmahwaty Fatmahwaty	
Novel Hybrid Orthogonal Large Set Code Sequence for High Density Wireless	PDF
Networks Nyoman Pramaita, P. I. Johnson	
Nyoman Pramaita, P. L. Johnson Novel Centralized Control Battery Operation, on Microgrids to Supply The Load	PDF
Priority	_
Hartono Budi Santoso, Wike Handini, Rudy Setiabudy, Budiyanto Budiyanto	
Learning based on Virtual Class using combination of Second Life and Learning	PDF
Managment System Firli Irhamni, Indah Agustien Siradjuddin, Arik Kurniawati, Ari Kusumaningsih,	
Rima Triwahyuningrum	
Physic Simulation Experiments using Augmented Reality	PDF
Surya Sumpeno, Christyowidiasmoro Christyowidiasmoro Performance of Anaerobic Reactor in Attached and Suspended Growth Mechanisms	PDF
for the Removal of Tofu Wastewater	<u></u>
Rosvita Tri Jullyanti, H. Purnobasuki, T. Nurhariyati, F. N. Rachman, N. I. Oktavitri	
Analysis Characteristic of Province Travelling Intercity of AKDP Makassar-Parepare	PDF
Route, South Sulawesi	_
St. Maryam Hafram, Herman Parung, Tri Harianto, Muh. Isran Ramli	
<u>Investigation of Flow and Heat Transfer Characteristics in Two-Phase Flow Through</u> <u>Evaporator of Diffusion Absorption Refrigerator (DAR)</u>	PDF
Siti Masita Rakhmawati	
Detection of Critical Situation of Longitudinal Vehicle Dynamics	PDF
Widya Nila Velayati, Sebastien Varrier	
A Robust Frequency Control Approach in PV-Diesel Hybrid Power System	PDF
Heri Suryoatmojo, Adi Kurniawan, Feby Agung Pamuji, Nursalim ST MT, IGP Suta Wijaya, Herbert Innah	
Assessment the Role of Sediment in the Sumber Brantas Rivers Watershed Affected	PDF
<u>by Agroforestry Activities</u>	
Barlah Rumhayati, Hari Siswoyo, MH. Chusnuddin Nurochman	DDE
Study at the Bolted Joint Assembly with Slotted Hole Ramadhan Prasetya Utama, Alain Daidié	PDF
Augmented Reality Prototype to Introduce Gamelan Based on Web	PDF
Avriyasendy Ramadiyan, Sari Wijayanti	
Power Flow Control of Power Systems Using UPFC Based on Adaptive Neuro Fuzzy	PDF
Agus Jamal, Ramadoni Syahputra Distribution Network Efficiency Improvement Based on Fuzzy Multi-objective Method	PDF
Ramadoni Syahputra, Imam Robandi, Mochamad Ashari	<u>FDI.</u>
Crystal Growth of IRMOF-3 (Isoreticular Metal-Organic Frameworks-3) Synthesized	PDF
<u>using Solvothermal Method</u>	
Pemta Tia Deka, Ratna Ediati, Didik Prasetyoko	DDE
<u>Utilization of Fertilizers for Improving The Process Rate of Organic Material</u> <u>Degradation in Anaerobic Reactor</u>	<u>PDF</u>
Adelia Anju Asmara, Agoes Soegianto, Trisnadi Widyaleksono Catur Putranto,	
Hairul Amin, Nur Indradewi Oktavitri Database Integration Model for Automatic Identification System and Shipping	PDF
Database In Real Time Traffic Monitoring	<u>. Dr.</u>
Akhmad Maulidi, Trika Pitana, Ketut Buda Artana, A.A. Bagus Dinariyana,	
Muhammad Badrus Zaman, Agoes A. Masroeri, Ricky Randall Sembiring Energy Efficiency Analysis of Trawlers (Case Study: Indonesian's Trawler)	PDF
Energy Efficiency Analysis of Trawlers (Case Study: Indonesian's Trawler) Alyuan Dasira, Jean Marc Laurens	<u>FDF</u>
Analysis of The Effect of Linear and Non-Linear Loads against The Quality of Power at	PDF
, <u> </u>	

The Power Converter	
Anicetus Damar Aji, Kusnadi Kusnadi	DD.
Interactive Whiteboard and Instant Quiz for Multimedia Classroom Activity Ary Mazharuddin Shiddiqi, Royyana M. Ijtihadie, Emerson Eridiansyah Z, Baskoro Adi Pratomo, Tohari Ahmad, Waskhito Wibisono	<u>PD</u>
Chemical Bonds Visualization using Particle Effect and Augmented Reality Christyowidiasmoro Christyowidiasmoro, Surya Sumpeno	PD
The Alternative Use Of Water Hyacinth and Reed As Attached Growth Of Microbial In Waste Water Treatment	PD
Hery Purnobasuki, Nur Indradewi Oktavitri, Tri Nurhariyati, B. Saadah, Santini Ika Rafsanjani	
Modelling and Analysis of the Dynamic Behavior of a Double Cylinder Inline 650cc Gasoline Engine with Crank Angle 0o for Rubber Mount	<u>PD</u>
Harus Laksana Guntur, Karina Yulia The Erythemato-Squamous Dermatology Diseases Severity Determination using Self-	PD
<u>Organizing Map</u> Haryanto Haryanto, Miftahul Ulum, Diana Rahmawati Rahmawati, Koko Joni, Ahmad Ubaidillah, Riza Alfita, Lilik Anifah, Bain Khusnul Khotimah	
Location Time of Power Quality Disturbances and Noise Using Morphology Gradient and Skeletonization in 3D	PD
I Gusti Ngurah Agung Dwijaya Saputra, Henry Wu, Wenhu Tang Pattern of Biogas Production and Removal of Chemical Oxygen Demand in Semi Continues Hybrid Anaerobic Reactor	PD
Nur Indradewi Oktavitri, Agoes Soegianto, T. Rahman, Bidayatus Saadah, Deavy Trianingtyas	
Three-Dimensional (3D) Reconstruction for Detecting Shape and Volume of Lung	PD
Cancer Nodules Rodiah Rodiah, Sarifuddin Madenda, Fitrianingsih Fitrianingsih	
Thermal Performance of Apartment (High-Rise) in Surabaya with Precast Concrete for the Building Envelope	<u>PD</u>
Romanria Violina Mahardhika, Ima Defiana, V. Totok Noerwasito	
Pretreatment of Sludge Milk Waste as Source Of Composting using Microbes S. R. Juliastuti, Rian Setya Budi, Taufiqurrusydi Taufiqurrusydi	<u>PD</u>
Further Investigation on Building and Benchmarking A Low Power Embedded Cluster for Education	PD
Sritrusta Sukaridhoto, Achmad Subhan Khalilullah, Dadet Pramadihanto	
Real Time Train Monitoring using Google Maps Supeno Djanali, Ary M. Shiddiqi, Hudan Studiawan, Henning T. Ciptaningtyas	<u>PD</u>
Structural Damage Detection of A Steel-Truss Railway Bridge Using its Dynamic Characteristics	<u>PD</u>
T. Susanto, A. Budipriyanto	
Android App for Information of Food Additives Contained in Packaged Foods Umi Sa'adah, Tita Karlita, Muchammad Arfian	<u>PD</u>
The Application of Neural Network for Predicting Corrotion Rate in Metal Pipe Installation	<u>PD</u>
Zulkifli Abdullah, Detak Yan Pratama, Dyah Sawitri, Doty Dewi Risanti Aquaculture Water Quality Prediction using Smooth SVM	PD
Wijayanti Nurul Khotimah	<u></u>
The Effect of Furnace Temperature and Precursor Concentration Ratio to The	PD
Characteristics of Nanocomposite ZnO-Silica Iva Maula, Widiyastuti Widiyastuti, Tantular Nurtono, Fadlilatul Taufany, Siti Machanydah, Sugana Wijasrdi	
Machmudah, Sugeng Winardi An Efficient Method for the Production of Biodiesel from Rice Bran	<u>PD</u>
Siti Zullaikah, Yulia T. Rakhadima, M. Rachimoellah, Tri Widjaja, Sumarno Sumarno	
Miniature and Low-Power Wireless Sensor Node Platform: State of the Art and	<u>PD</u>
Current Trends C. Bambang Dwi Kuncoro	
Holistic Model for SMEs Growth Studies: A Conceptual Overview Johan K. Runtuk, Budisantoso Wirjodirdjo, Iwan Vanany	PD
Interior Batik Gallery Using Normal Mapping For Virtual Reality	<u>PD</u>
Ayung Candra Padmasari, Christyowidiasmoro Christyowidiasmoro, Mochamad Hariadi	
Strength and Ductility of RC Columns Retrofitted by FRP under Cyclic Loading	PD
Tavio Tavio, Karmila Achmad, Parmo Parmo, Agus Sulistiawan Characterization of A Directional Coupler Waveguides Five Layers based on A	<u>PD</u>
Variation of The Electric Field in Two Border Area Farida Wati, Ali Yunus Rohedi, Yono Hadi Pramono	
An Optimization Model for Container Capacity in New Sorong-Papua Port	PD
Iwan Vanany, Duha Awaluddin Kurniatullah, Edi Jadmiko, Dody Hartanto,	
zulfiyah Musfiroh	
Design of the Exterior & Interior Car Body for Soekarno-Hatta Airport Railing Service	PDi
Design of the Exterior & Interior Car Body for Soekarno-Hatta Airport Railing Service Agus Windharto	
Design of the Exterior & Interior Car Body for Soekarno-Hatta Airport Railing Service	<u>PDI</u> PDI

ACIIIIau Noityaiii, Filiiippe Sciiiiiic, Ciiiistiile Laitorgue Biofuel Produced from Nyamplung Oil Using Catalytic Cracking Process with Zn-PDF **HZSM-5 Catalyst** Agus Budianto, Danawati Hari Prajitno, Kusno Budhikarjono, Achmad Roesyadi, Ratna Ediati The Exterior and Driver Cabs of Locomotive CC300 Based on Integrated Digital PDF Design Agus Windharto Design Development of Food & Beverage Product Packaging for SMES co in Java and PDF West Nusa Tenggara, Economic Corridor. Agus Windharto <u>PDF</u> Agro-industrial Development in Lamongan District, East Java, Indonesia Ajeng Nugrahaning Dewanti, Jayant Kumar Routray, Rima Dewi Modelling Back-to-Back Converter with Average Model in Wind Power System Using PDF Anisa Harumwidiah, Mochamad Ashari, Dedet Candra Riawan Design and Implementation of Photovoltaic Single Stage Inverter Connected to Grid PDF Bayu Prasetyo, Mochamad Ashari, Dedet Candra Riawan Development of Supply Chain Risks Interrelationships Model using Interpretive PDF Structural Modeling and Analytical Network Process Chatarina Dian Indrawati, Putu Dana Karningsih, Iwan Vanany Scratch Area Effect in Coating to Protection Current Needing in ICCP System for AISI PDF 1045 Steel in Sea Water Environment Dewanti Dewanti, Tubagus Noor Rohmannudin, Sigit Tri Wicaksono The Design Process for Entrapping Limonin and Naringin in Siam Juice by PDF Cyclodextrin Dian Fajarika, Erliza Noor Mp Tuning for Internal Model Control 2x2 Multi Input Multi Output (MIMO) System PDF Dinny Winda Astuti, Juwari Juwari, Renanto Handogo Implementation of Social Networking Application as a Tool for English Language **PDF** Learning in Vocational and Boarding School Eka Widhi Yunarso, Pikir Wisnu Wijayanto, Ismail Ismail Vulnerability Assessment of Reinforced Concrete Building Subjected to Seismic Loads **PDF** Using Pushover Analysis Endah Wahyuni, Data Iranata, Heppy Kristijanto Energy Estimation on BLDC Electric Vehicle using MATLAB **PDF** Hasnira Hasnira, Didi Istardi Design and development of Web-Based Information System for The Batik Industry PDF An Electronic Traceability System for an Indonesian Fresh Fruit Supply Chain **PDF** Iwan Vanany, Kuntoro Boga Andri, Ronny Mardiyanto, Niniek Fajar Puspita, Wiwik Heny Winarsih Design and Implementation of Axial Flux Induction Motor Single Stator - Single Rotor PDF for Electric Vehicle Application Mochamad Ashari, Heri Suryoatmojo, Dedet Candra Riawan, Ronny Mardiyanto, D Fahmi, S Hidayat, K B Adam Shear Behavior of Joint The Partial Prestressed Concrete Beam-Column Reinforced PDF Concrete of Ductile Frame Structure Building In a Scure Residents and for Settlement **Environment** Made D Astawa, IGP Raka, Tavio Tavio Maximum Crack Width Prediction in Deck Slab Concrete Structure PDF Nawir Rasidi Construction the Statistics Distributions for Characterizing the Transfer Factors of **PDF** Metals from Soil to Plant (TFsp) Using Bayesian Method Pratnya Paramitha Oktaviana, Marie-Pierre Etienne Stability Phenotypic Characters and The Scent of Gama Melon Parfum Cultivar PDF Sigit Dwi Maryanto, Rini Etika Ranis, Budi Setiadi Daryono Test Of Bio-Activator Model with Natural Zeolite from South Sulawesi As Filter Media **PDF** and Adsorbents Heavy Metal Pb, Ni, and Cu Sukmasari Sukma Antaria, Mary Selintung, Muh Saleh Pallu, Mukhsan Putra Go East Java: A Mobile Application For Tourist Guides In East Java **PDF** Umi Sa'adah, Imam Mustafa Kamal, Fawaati Tsabita, Vickrilia Yulia Ekaputri Power Management of A Microgrid with A Distributed Energy Storage in Grid PDF Connected and Islanded Modes Wike Handini, Hartono Budi Santoso, Rudy Setiabudy, Eko Adhi Setiawan Utilization of Rice Straw Waste in The Production of Crude Cellulase by Aspergillus PDF Niger for Biodeingking Processes Yanty Maryanty, Kristina Widjayanti, Sri Rulianah, Meiliefiana Meiliefiana, Windra Putri Juwita Phenotypic Characters Stability of Melodi Gama-3 Melon (Cucumis melo L.) Cultivar **PDF** in Rainy Season Based on Multilocation Test Budi Setiadi Daryono, Rizko Hadi, Yasir Sidiq, Sigit Dwi Maryanto The Use of Matrix Heat Pipe as Photovoltaic Cooler **PDF** Rahmat Subarkah, Tatun Hayatun Nufus, Rahmat A Setiawan, Achmad Subakri, Wisnu Tri Nur Kahfi, Try Laksono Prasetyo Experimental Characterization of A Load Sensor Based on Singlemode - Multimode -PDF

Singlemode (SMS) Fiber Structure with A Variation of The Number of Bends	
Rionda Bramanta Kuntaraco, Agus Muhamad Hatta, Sekartedjo Koentjoro, Catur	
Arif Prastyanto, Insrasurya Budisatria Mochtar	
Optimization of Reaction Conditions for Synthesis of MOF-5 using Solvothermal	PD
<u>Method</u>	
Tri Ana Mulyati, Ratna Ediati, Muhammad Nadjib	
Subcritical Water Extraction of Phenolic Compounds from Moringa Oleifera Leaf	PD
Siti Zullaikah, Irfan Saputra, Ghuzrina Prihandini, M. Rachimoellah	
Medical Breakthrough of Anthropometric Methods as Basis for 3D Digital Modelling of	PD
<u>Indonesian Female Facial Type (Descriptive-analytic-applied Study)</u>	
Ulfah Elfiah, M. R. Hutagalung, D. S. Perdanakusuma, Phill T Koesbandriati, Agus	
Windharto	
Development of Wireless Sensor Network for Monitoring Air Temperature and Soil	PD
Moisture on Crops	
Wulandari Wulandari	
<u>Design and Implementation of Inventory Domain for Enterprise Resource Planning</u>	PD
<u>Using SOA and Workflow Approach</u>	
Dwi Sunaryono, Adhatus Solichah Ahmadiyah, Riyanarto Sarno, Riswandy	
Setiaputra G	
Analysis of Ergonomics Risk Factors and Manual Material Handling Assessing	PD
Alternative Using Cost Benefit Methods	
Eko Nurmianto, Udisubakti Ciptomulyono	
Javanese-Indonesian Female Facial Expression (JIFFE-3D) Based on 3D Marker	PD
Muhtadin Muhtadin, Surya Sumpeno, Hosyi'ah Rusdiana	
Female Body Anthropometric Variation and Breakthrough in Anthropology for Digital	PD
<u>Modelling</u>	
Badriatut Dini, M. R. Hutagalung, D. S. Perdanakusuma, Phill T Koesbandriati,	
Agus Windharto	

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Biofuel Produced from Nyamplung Oil Using Catalytic Cracking Process with Zn-HZSM-5 Catalyst

Agus Budianto¹, Danawati Hari Parjitno¹, Kusno Budhikarjono¹, Achmad Roesyadi¹, and Ratna Ediati²

Abstract—Indonesia Presidential Regulation No. 5/2006 on National Energy Policy suggests that the government should speed up the implementation of the use of alternative energy or fuel substitution. Biofuel synthesis is one way to overcome the shortage of energy and reduce global warming due to the use of fossil fuel. Biofuel can be produced from a variety of vegetable oil. Beside palm oil, nyamplung oil can be used to produce biofuel. The technically main obstacle in producing biofuel is the availability of the catalyst. The availability catalyst are only imported and expensive. Researchers have tried to engineer a new type of catalyst that complete the weakness of zeolite based catalyst. The study was conducted through experimental approaches, testing and observations and conducted the correlation of experiment variables with the quality of the resulted catalyst. The experiment was done by synthesizing catalyst and testing it to produce biofuel from nyamplung oil. The focus of the research is directed to the effect of operating variables on the composition of the resulted biofuel and obtain catalyst performance condition and optimum condition to produce biofuel in the fixed bed reactor. The resulted catalyst can change the nyamplung oil into biofuel. Biofuel from nyamplung oil cracking process showed that the composition is biogasoline, biokerosene and biodiesel. Biodiesel fraction is the highest fraction of the biofuel produced. The highest percentage of biodiesel at a temperature of 400°C was 60%, while the lowest percentage of biodiesel at a temperature of 300°C was 48%. Products density was in the range of 0.81 to 0.86 g/ml. The highest density occured at a reactor temperature of 300 °C was 0.86g/ml. The higher the nitrogen gas flow rate the more the biodiesel formed. At a temperature of 300°C and a nitrogen flow rate of 100 ml/min, the composition solar achieved was 60%.

Keywords—Biofuel, Cracking, Catalyst, Nyamplung Oil, and Zeolites.

I. INTRODUCTION

In 2006, the Indonesian government has issued a policy that mandates a policy of national energy diversification. Presidential Decree No..5/2006 on National Energy Policy. This regulation states that the government should speed up the implementation of the use of alternative energy or fuel substitution in order to reduce fuel subsidy. Fuel used by the transport sector is the biggest consumption of the energy mix, so as to improve fuel security of supply in order to achieve a balance of energy mix by 2025. On the other hand depletion of world oil reserves and increasing environmental concerns pose a great demand to find a replacement for alternative sources of petroleum based fuel, including diesel and gasoline fuel (Leung et al, 2010).

Many scientists began researching to find a new kind of energy that is cheap, easy and environmentally friendly to replace energy sources that are available now, namely the use of vegetable oil. Vegetable oil which is converted into biodiesel by esterification and transesterification processes (Leung et al, 2010; Bhale, 2009, Demirbas, 2003; Budianto, et al, 2003, Kansedo et al, 2008) produces alkyl ester biodiesel. The completion of the characteristic of alkyl ester combustion was done by forming ozonida biodiesel (Bismo, 2005; Budianto, 2009). Vegetable oil can also be converted to biofuel by

cracking to produce the broader product, i.e., biofuel (Dandik et al., 1980; Chang, 1983; Bhatia, et al., 1999; Farouq et al., 2004; Charusiri, & Vitidsant, 2005; Masuda et al., 2001; Thiam, 2009; Nurjannah et al, 2009a). Biofuel has advantages compared with alkyl ester biodiesel. The advantages of biofuel is that the product is liquid fuel with chemical components similar to those in conventional diesel oil (Dykstra. et al, 1988; Adjaye, et al, 1995).

Biofuel can be produced from a variety of plant and animal oil, but it is very interesting if the oil is an Indonesia's distinctive plant such as nyamplung. Nyamplung advantage as a raw material of biofuel is the seed has high yield, up to 74 %, and in the interests of its utilization, it does not compete with food. Several advantages in terms of the prospect of nyamplung development and other uses are nyamplung plant grows and spread naturally evenly in Indonesia; it easily regenerates and fruits throughout the year showed a highly survival power of the environment; a crop plant which is relatively easily cultivated either type (monoculture) or mixed forest (mixed-forest); it is suitable in drier climates; almost all parts of the the plant are useful and it produces a variety of products that has economic value; stand of nyamplung forest serves as a wind breaker for agricultural crops and conservation of coastal border, and the utilization of nyamplung biofuel reduces the use of forest trees for firewood; seed productivity is higher than that of other types of plant. Jatropha, palm and nyamplung produce seed of 5 tonnes/ha, 6 tonnes/ha and 20 tonnes/ha respectively. (PRESS RELEASE Number: S. 428/II/PIK-1/2008, Ministry of Forestry Republic of Indonesia)

Nyamplung oil is one of the alternatively potential raw materials to produce biofuel as a substitution of gasoline, kerosene and diesel because it is potential for the

¹Agus Budianto, Danawati Hari Parjitno, Kusno Budhikarjono, and Achmad Roesyadi are with Departement of Chemical Engineering, Faculty of Industrial Technology, Institut Teknologi Sepuluh Nopember, Surabaya, 60111, Indonesia. E-mail: budi_2050@yahoo.com.

²Ratna Ediati is with Departement of Chemistry, Faculty of Mathematics and Science, Institut Teknologi Sepuluh Nopember, Surabaya, 60111, Indonesia.

development of nyamplung oil. This is supported by the fact that nyamplung cultivation does not require high investment. Potential availability of land for the development of nyamplung plants spread throughout Indonesia. When all of the needs of the biofuel were supplied by nyamplung, it will be needed 720,000 kilo liters of biodiesel, equivalent to 5.1 million tonnes of nyamplung seed, assuming that 2.5 kg of the seeds will produce 1 liter nyamplung oil. Thus it would need a minimum area of nyamplung crops of 254,000 acres in 2025. With the same pattern with the economic analysis study on the construction of Public Plantation Forest (HTR) which states that it takes 1 worker for 1 ha. Thus nyamplung cultivation area of 254 thousand hectares will be able to absorb 254 thousand workers. (ibid)

Production of biofuel from plant oil such as palm oil using zeolite as catalyst in the cracking process has been widely studied (Nurjannah, 2009a; Twaig et al 1999; Twaig et al., 2003; Twaig et al., 2004; Ooi et al., 2003, Ooi et al.2004a; Ooi et al. 2004b; Nurjannah et al., 2009). HZSM-5 zeolite and its modification using a steering metal such as Cu, Ni, Zn, Pd, Au and Pt have properties and good performance in hydrocracking process (Budianto et al 2010, Budianto et al 2011, Budianto et al 2012, Nurjannah et al, 2009a). Zn-HZSM-5 is highly recommended for palm oil cracking because it has the highest yield for gasoline (Roesyadi et al, 2012). The modified catalysts are very good for palm oil cracking process, but the catalysts have not been tested for catalytic cracking of nyamplung oil to produce biofuel. The research team estimated that Zn-HZSM-5 catalyst is very interesting and it can be used for catalytic cracking process of nyamplung oil to produce biofuel, where the operating condition of the process still needs to be studied further. Although it is triglyceride oil but the composition of fatty acids is different, so it is interesting to be studied.

A. Research Objectives

The purpose of this research is to study the operating condition of catalyst preparation of Zn-HZSM-5 and to determine the characteristics of the resulted catalyst and cracking reaction operating condition of nyamplung oil converted into biofuel with the yield of biogasoline, biokerosene and biodiesel. Additionally, the purpose of this research is to study the effect of feed flow rate, temperature, catalyst composition of nyamplung oil cracking reaction on the selectivity of biofuel produced.

B. Literature Study

Biofuel is liquid or gaseous fuel that can be produced from biomass or substrate of bioresource (Stocker, 2008). Biofuel offers a number of advantages over fossil fuel in terms of (a) the availability of renewable resources, (b) represents the CO₂ cycle in the combustion, (c) green (d) biodegradable and sustainable (Puppan, 2002). In recent years, research on biofuel has been done a lot, such as the use of various types of catalyst for biofuel products (Farouq et al, 2004, Chewand Bhatia, 2008), studies of the optimization of the reactor used (Tamunaidu and Bhatia, 2007). Kinetics of cracking oleic acid to produce biofuel (Nurjannah et al, 2010), the use of composite catalyst and deactivation studies on palm oil cracking process to produce biofuel

(Bhatia et al, 2009). Hydrocracking of used oil can also be processed into biofuel (Bezergianni and Kalogianni, 2009).

The increasing of interest in producing biofuel is caused by the depletion of fossil fuel. Environmental pollution is also a major factor to look for alternative energy sources. Today, 86% of world energy consumption and nearly100% of the energy needed in the transport sector is fulfilled by fossil fuel (Dorian et al, 2006). Since the world's oil reserve has begun gradually depleted, it is important to develop a suitable long-term strategy and based on the utilization of renewable fuel, it will replace fossil fuel gradually (Westermann et al, 2007). Palm oil has attracted the attention of researchers to develop environmentally friendly products and high-quality fuel, free of nitrogen and sulfur (Tamunaidu, 2006). Thus, nyamplung oil is expected to give higher quality than palm oil.

Nyamplung oil is vegetable oil. Oil is a general term for all organic liquid that are not soluble/mixed in water. Vegetable oil has long hydrocarbon chains that are similar to petroleum. Because of the similarity of the hydrocarbon chain, nyamplung oil can be used as biofuel. Therefore, petroleum cracking process can be applied to the manufacture of biofuel from vegetable oil (Nurjannah et al, 2010).

II. METHOD

Materials used for this study are divided into two groups. The first group is materials for making synthetic Zn-HZSM-5 catalyst: water glass, alum/Al₂(SO₄)₃.18H₂O, H₂SO₄, HCl, NaOH, NH₄Cl, Na SiO₄, H₂ gas and Zn Chloride. The second group is materials for cracking N₂ gas, nyamplung oil and seed.

The research equipments used to produce catalyst are autoclave and furnace equipped with thermocouple and temperature indicator, stainless steel hose, thermometer, hotplate, stirrer, micro bed reactor with high pressure, stative and clamp. While the characteristics of the catalyst test were conducted by Brunauer Emmett Teller (BET), Atomic Absorption Spectroscopy (AAS), and Scanning Electron Microscope (SEM). Equipment used in nyamplung oil cracking process establish biofuel is presented in Figure 1.

This study include:

The preparation stage of basic catalyst of synthetic Zn-HZSM-5 was the process of making synthetic zeolite catalyst which was done in two steps, i.e. synthesis and characterization of catalyst. In synthesis step, the process of making synthetic zeolite catalyst in an autoclave reactor and the results are expected to form NaZSM-5 and continued with the process of change into Zn-HZSM-5. In the second step, the catalyst was characterized to obtain the ratio of Si/Al, surface area and pore volume (porosity). Catalyst activation process was then conducted with impregnation of zinc metal.

The stage of nyamplung oil catalytic cracking process in the fixed bed micro-reactor gas phase was conducted at equilibrium temperature at a pressure of 1 atm. Cracking products were analyzed by gas chromatography FID column carbowax types of 20 meter. Analysis of the density was done by pycnometer.

III. RESULT AND DISCUSSION

A. Determination of the composition of biofuel

The resulted catalyst was tested in a fixed bed reactor at various temperatures to convert nyamplung oil into biofuel. The resulted products, biofuel, were analyzed using gas chromatography equipment (GC). By comparing the chromatogram of biofuel against the chromatogram of gasoline, diesel and kerosene, it will be obtained commercially produced biofuel composition. Figure 2, Figure 3 and Figure 4, are chromatograms using GC for commercially gasoline, kerosene and diesel fuel.

Figure 2, 3 and 4 were used to calculate the composition of biofuel based on its rentention time. Rentention time is between 0-17, 17-35 and 35-60 for gasoline, kerosene and diesel respectively. The chromatograms using GC for biofuels from nyamplung oil can be seen in Figure 5, 6 and 7.

By comparing the biofuel retention time data with the rentention time of commercial gasoline, kerosene and diesel, it was obtained biofuel product composition as shown in Figure 8. Figure 8 shows that the highest liquid biofuel composition is biodiesel and the lowest composition is biogasoline. In the temperature range of 300-400°C, it shows that the higher the temperature the higher the composition of biodiesel, but the higher the temperature the lower the composition of biokerosene.

B. Effect of nitrogen flowrate

Nitrogen gas is used to push the vapor of nyamplung oil feed to the reactor. The nitrogen flow rate affects the composition of the resulted product. In Figure 9, it can be seen that the higher the nitrogen gas flow rate the higher the composition of biodiesel, but the higher the nitrogen gas flow rate the lower the compositions of both biokerosene and biogasoline.

IV. CONCLUSION

From the research that has been done a few conclusions can be drawn: Biofuel from nyamplung oil cracking process showed that the composition was biogasoline, biokerosene and biodiesel. The Results of the chromatogram of biofuel from nyamplung oil using GC equipment showed that the highest percentage of biodiesel at a temperature of 400°C was 60%, while the lowest percentage of diesel fuel at a temperature of 300°C was 48%. Products density was 0.81 to 0.86 g/ml. The highest density occured at a reactor temperature of 300°C was 0.86 g/ml. The higher the nitrogen gas flow rate the more the biodiesel formed. At a temperature of 300°C and a nitrogen flow rate of 100 ml/min, the composition biodiesel achieved was 60%.

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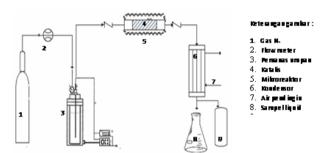


Figure 1. Equipment of vegetable oil cracking process

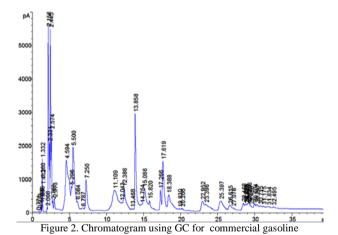
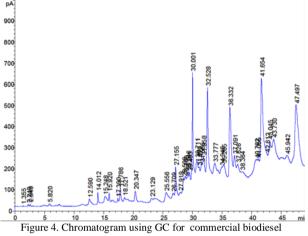


Figure 3. Chromatogram using GC for commercial kerosene

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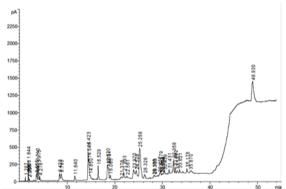


Figure 5. Chromatogram using GC for biofuel from nyamplung oil at a temperature of 300°C

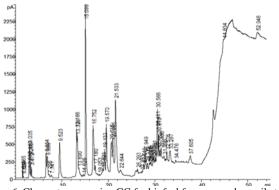
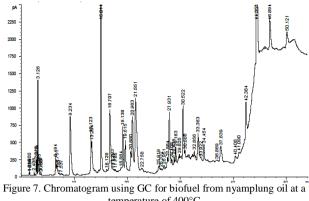
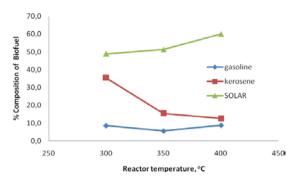


Figure 6. Chromatogram using GC for biofuel from nyamplung oil at a temperature of 350°C





temperature of 400°C

Figure 8. The effect of reactor temperature on the composition of the resulted biofuel

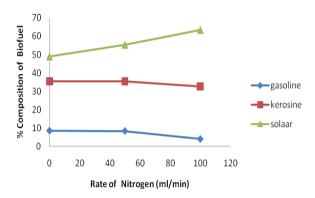


Figure 9. The effect of nitrogen gas flow rate on the composition of biofuel at 300°C