International Symposium of Technologies and Innovation for Material Circulation

Osaka University ASEAN Campus SDGs Co-creation Forum on Plastic Solution

Date and time: May 29, 2024, 9:30 –14:50 (MYT), 10:30 – 15:50 (JST)
Location: Webex (online) and at Osaka University ASEAN Campus Office, Toray Building 3F, USM (on-site)

Webex link : https://usm-cmr.webex.com/usm-cmr/j.php?MTID=m18a47d4ba1559939a39f8b7b814246f9

Meeting number : 2514 026 2855
Password : USM2024

Subjects:

- Innovative Biomass Conversion Technologies
- Biodegradable and Bio-based Plastic Development
- Life Cycle Assessment of Biomass-based Materials
- Circular Economy and Waste Valorization Strategies
- Advanced Functional Materials from Biomass
- Policy and Socio-economic Analyses of Biomass Material Circulation

Program:

Opening Remarks

Start time			
Penang	Osaka		
9:30	10:30	(5 min)	Professor Dr. Zainovia Lockman, Director, Research Creativity & Management Office, USM

Keynote Presentation

Start time			
Penang	Osaka		
9:35	10:35	(20 min)	Professor Dr. Hiroshi Uyama, Division of Applied Chemistry, Graduate School of Engineering, OU *Plant-based bioplastics for sustainable development: Recent progress and advancement*
9:55	10:55	(20 min)	Professor Dr. K Sudesh Kumar, School of Biological Sciences, USM *Biosynthesis and biorecovery of microbial bioplastics*

Research Talks

Start time			
Penang	Osaka		
10:15	11:15	(10 min)	Professor Dr. Norikazu Nishiyama, Division of Chemical Engineering, Graduate School of Engineering Science, OU *Zeolite catalysts for polymer cracking*
10:25	11:25	(10 min)	Professor Ir. Dr. Mariatti Jaafar, School Of Materials And Mineral Resources Engineering, USM *Development of plastic composites using recycled plastic and natural fiber*

10:35	11:35	(10 min)	Assistant Professor Dr. Sho Yamaguchi, Department of Materials Engineering Science, Graduate School of Engineering Science, OU *Development of high performance heterogeneous catalysts toward valorization of renewable resources*
10:45	11:45	(10 min)	Professor Dr. Mohd Azmier Ahmad, School of Chemical Engineering, USM *Co-pyrolysis of biomass waste with plastic waste for bio-oil production*
10:55	11:55	(10 min)	Assistant Professor Dr. Yuki Nakaya (Division of Applied Chemistry, Graduate School of Engineering, OU) *Development of multifunctional alloy catalysts for carbon neutralization and recycle*
11:05	12:05	(10 min)	Ts. Dr. Ku Marsilla Ku Ishak, School Of Materials And Mineral Resources Engineering, USM *Fabrication of thermochromic films as smart temperature sensor*

Discussion

Start time

Penang	Osaka		
11:15	12:15	(15 min)	Discussion on further collaborations (all speakers)

Break **11:30– 12:30 (Penang), 12:30 - 13:30 (Osaka)**

Short talks by students and junior researchers 1

Start time

Penang	Osaka		
12:30	13:30	(64min)	Session 1 (**8** minutes each, 8 presenters total)

1. Nuraina Hanim Mohd Nizam, School Of Materials And Mineral Resources Engineering, USM
   Development of biodegradable plastic wrapping materials of pathological waste buried in soil environment
2. Specially Appointed Assistant Professor Dr. Li Duanxing (Division of Applied Chemistry, Graduate School of Engineering, OU)
   Coke-resistant Ce-based multi-element oxide supports for CO_2-propane dehydrogenation
3. Massara Samsul Baharin, School Of Materials And Mineral Resources Engineering, USM
   3D printing of biodegradable PLA/PBAT blends through the 3-glycidoxypropyltrimethoxysilane (GPTMS)
4. Katsumasa Sakoda, Division of Chemical Engineering, Graduate School of Engineering Science, OU
   Reductive amination of carboxylic acids using a Pt–Mo catalyst for sustainable alkylamine syntheses
5. Tan Kai Qi, School of Chemical Engineering, USM
   A sustainable route from waste to wealth: Upcycling disposable face masks into value-added hydrocarbons for carbon nanomaterials production via innovative pyrolysis setup
6. Shinya Kokuryo, Division of Chemical Engineering, Graduate School of Engineering Science, OU
   Coke deposition resistance by Cr^{6+} loading on zeolite defects: Reduced regeneration in low-density polyethylene cracking
7. Amirah Ayu Kartika Asman, School of Chemical Engineering, USM
   Assessing environmental impacts of facemasks via pyrolysis process and its life cycle assessment (LCA)

8. May Myat Noe, Division of Applied Chemistry, Graduate School of Engineering, OU
   Tough host–guest composite hydrogels reinforced by dual supramolecular cross-linkers

Break 13:34 – 13:40 (Penang), 14:34 - 14:40 (Osaka)

Short talks by students and junior researchers 2

Start time			
Penang	Osaka		
13:40	14:40	(64min)	Session 2 (**8** minutes each, 8 presenters total)

9. Muhammad Shafizruddin Firdaus Fazli-Ku, School of Chemical Engineering, USM
   Catalytic cracking of waste cooking oil over activated carbon supported trimetallic oxide catalysts
10. Ying Yao, Division of Applied Chemistry, Graduate School of Engineering, OU
    Effect of poly(vinyl alcohol) on the physical properties of starch-based composite films
11. Muhammad Safiuddin Rosli, Center for Global Sustainability Studies, USM
    The utilisation of organic domestic waste to high grade smokeless fuel (HiGraS-Fuel) using a carbonizer
12. Soshi Tsubota, Division of Chemical Engineering, Graduate School of Engineering Science, OU
    Effect of Brønsted acidity of zeolite on catalytic cracking temperature of low density polyethylene
13. Janet Wan, Center for Global Sustainability Studies, USM
    Electrifying rural areas: The story of Sarawak
14. Taiki Kawakami, Division of Chemical Engineering, Graduate School of Engineering Science, OU
    Nickel carbide nanoparticle catalyst for direct hydrogenation of furfural to tetrahydrofurfuryl alcohol
15. Ganapaty Manickavasagam, School of Chemical Sciences, USM
    Transforming sawdust into high-performance Co-N-co-doped hydrochar catalyst for ciprofloxacin removal via peroxymonosulfate activation
16. Zhang Fang, Division of Applied Chemistry, Graduate School of Engineering, OU
    Study on multinary alloys and oxides as efficient catalysts for the oxidative dehydrogenation of propane using CO_2

Closing Remarks

Start time			
Penang	Osaka		
14:45	15:45	(5 min)	Professor Dr. Kiyoshi Fujita, Director of Campus Innovation Unit, Center for Global Initiatives and Regional Director of ASEAN Center for Academic Initiatives, OU

ABSTRACT

Plant-based bioplastics for sustainable development: Recent progress and advancement

Hiroshi Uyama
Division of Applied Chemistry, Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0861, Japan
Tel: +81-6-6879-7364; Email: uyama@chem.eng.osaka-u.ac.jp

Reducing the plastics amount and developing marine-biodegradable plastics are important social issues related to plastic waste. Using biomass as starting material for chemicals and plastics contributes to global sustainability without depletion of scarce resources, because of their large potential to substitute petrochemical derivatives to bio-based ones in industries. Poly(L-lactic acid) (PLA) is one of the most typical plant-based bioplastics (biomass plastics) showing good biodegradability. However, its demerits such as low physical properties and slow molding process have restricted the development of industrial applications of PLA. The present talk deals with recent progress and advancement of biomass plastics applications.

We have focused biomass products such as plant oils and cellulose for preparation of functional bio-based materials.[1] For example, natural plant oil sources are found in abundance in the world; and hence, are expected as an ideal alternative chemical feedstock. Inexpensive triglyceride natural oils have been utilized extensively for coatings, inks, plasticizers, lubricants, resins and agrochemicals in addition to their applications in food industry.

We have synthesized branched bio-based polyesters from lactic acid and a plant oil and examined their applications to improve properties of PLA. Castor oil was used as initiator for synthesis of a branched bio-based polymer by ring-opening polymerization of L-lactide or polycondensation of L-lactic acid. The addition of only a small amount of the branched polymer to PLA improved the strain at break, showing good plasticization effect of the branched polymer for PLA. The terminal hydroxyl group in this branched polymer was modified by phthalic anhydride. The product with potassium salt form greatly induced the crystallization of PLA to improve the thermal and mechanical properties of PLA.

A new thermoplastic starch (TPS) has been developed and TPS can blend a variety of common plastics such as polyethylene and polypropylene as well as biodegradable plastics such as PLA and poly(butylene succinate) (PBS) by a conventional extruder to produce novel bioplastics with tuned properties. Thermoplasticity of TPS enables the production of the plastics showing the enhanced biodegradability in soil and marine conditions by blending PLA and/or PBS. Mechanical properties of the TPS-containing biodegradable plastics are easily tuned by the selection of the biodegradable polyesters and the blending ratio with TPS.

Reference: 1) H. Uyama, *Polym. J.*, **50**, 1003-1011 (2018).

ABSTRACT

Biosynthesis and biorecovery of microbial bioplastics

Kumar Sudesh
School of Biological Sciences, Universiti Sains Malaysia, 11800 Pulau Pinang, Malaysia
Tel: +604-6534367; E-mail: ksudesh@usm.my

Plastics are essential material in our modern lifestyle, however, they are produced from petroleum, which is a non-renewable resource. The disposal of plastics materials is a major problem and this issue is made worst by the generation of microplastics particles and fibers which are persistent in the environment and bioaccumulate in living organisms. Therefore, there is an urgent need to develop and use bio-based and biodegradable plastics. Among the many types of bioplastics, polyhydroxyalkanoates (PHA) is attractive because of its thermoplastics properties and complete biodegradability in land and sea. The efficient production of PHA from waste oils [1,2,3] has been achieved by using an engineered PHA synthase [4,5,6]. An industrial symbiosis approach to recover and purify PHA from bacterial cells is proposed to minimize waste and reduce the cost of producing PHA. The use of mealworms in the purification of PHA can benefit both the production of insect protein and PHA. Recent progress in the commercialization of PHA bioplastics in Malaysia will be presented.

References

[1] A. Surendran, M. Lakshmanan, J.Y. Chee, A. M. Sulaiman, K. Sudesh, *Front. Bioeng. Biotech.* 8 (2020) 169.
[2] D.V. Thuoc, D.N. My, T.T. Loan, K. Sudesh, *Int. J. Biol. Macromol.* 141 (2019) 885.
[3] B. Gutschmann, B. Huang, L. Santolin, I. Thiele, P. Neubauer, S.L. Riedel, *Microbiol. Res.* 264 (2022) 127177.
[4] H.J. Tang, S.Z. Neoh, K. Sudesh, *Front. Bioeng. Biotech.* 10 (2022) 1057067.
[5] H.T. Tan, M.F. Chek, M. Lakshmanan, C. P. Foong, T. Hakoshima, K. Sudesh, *Int. J. Biol. Macromol.* 159 (2020) 250.
[6] H.T. Tan, M.F. Chek, Y. Miyahara, S.Y. Kim, T. Tsuge, T. Hakoshima, K. Sudesh, *J. Biosci. Bioeng.* 134 (2022) 288.

Acknowledgement

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ABSTRACT

Zeolite catalysts for polymer cracking

Norikazu Nishiyama
Division of Chemical Engineering , Graduate School of Engineering Science, Osaka University

n.nishiyama.es@osaka-u.ac.jp

Development of new catalysts with high activity and selectivity is a key technology for the future chemical and petrochemical industry to reduce energy consumption. In the future society, carbon dioxide and biomass as well as waste plastics will play an important role as energy and material sources. Chemical recycling of plastics has appeared as a promising alternative to deal with a massive amount of plastic waste. In chemical recycling, plastic waste can be converted into valuable products such as monomers or petrochemical feedstock.

Catalytic cracking has emerged as a solution to overcome the drawbacks of thermal cracking. The catalytic cracking has a great potential to improve the product distribution and lower energy consumption depending on catalyst design. Solid acid catalysts such as zeolites have been applied for the catalytic cracking of plastics.

(1) Acid-controlled zeolites for polymer cracking [1-3]
This study investigated the effect of the acidity (acid type, strength, and amount, *etc*.) in zeolites on the activity for the catalytic cracking of polyolefins.

Although it is well known that Brønsted acid sites (BAS) in zeolites catalyze polyolefin cacking, the detailed effects of BAS have remained unclear. We prepared MFI-type zeolites with controlled strength and amount of BAS by introducing various trivalent metals, and the low-density polyethylene (LDPE) cracking temperatures were compared. The cracking temperature decreased as the BAS amount of the catalyst increased but reached a plateau as the acid amount was further increased. The saturated cracking temperature depends on the acid strength of the zeolites, thus the acid strength has a stronger influence than that acid amount.

To date, most studies have focused on BAS, as the cracking reaction of the polymer has been assumed to proceed via BAS, and the importance of Lewis acid sites (LAS) has not been understood. However, LAS can also generate carbenium ions via the abstraction of hydride ions from polyolefins. Herein, we prepared LAS-rich Beta zeolites by introducing defect sites and confirmed the more outstanding activity on LDPE cracking than the conventional Beta zeolites. This enhancement was caused by the increase in LAS; extra-framework aluminum and silanol.

Moreover, we investigated the effects of the acid type in zeolites on the yields of light olefins during the catalytic cracking of polyolefins. We prepared Lewis acidic Sn-Beta zeolites without BAS via a dealumination procedure. The synthesized Sn-Beta zeolites exhibited higher performance on LDPE cracking with light olefin yields. The absence of BAS dramatically inhibited the conversion of light olefins, thereby increasing the light olefin yields.

(2) Coke-resistant zeolite catalysts for polymer cracking [4-5]

In most of these processes, zeolites suffer from deactivation inevitably. This deactivation is mainly due to the trapping of heavy organic compounds (called coke) within the micropores, which result from secondary reactions during various hydrocarbon conversions.

In this study, we have developed a high coke-resistant zeolite catalyst by Cr loading on zeolite defects. A Cr-loaded zeolite cannot be considerably deactivated by coke deposition and can be reused for LDPE cracking without regeneration. The Cr species interact with hydroxyl groups derived from structural defects in the zeolite framework to inhibit coking. This study provides useful information on coking inhibition in various chemical industries.

References

[1] S. Kokuryo, K. Miyake, Y. Uchida, A. Mizusawa, T. Kubo, N. Nishiyama, "Defect engineering to boost catalytic activity of Beta zeolite on low-density polyethylene cracking," Materials Today Sustainability, 17, 100098 (2022).
[2] S. Kokuryo, K. Tamura, K. Miyake, Y. Uchida, A. Mizusawa, T. Kubo, N. Nishiyama, "LDPE cracking over mono- and divalent metal-doped beta zeolites," Catal. Sci. Technol., 12, 4138-4144 (2022).
[3] S. Kokuryo, K. Miyake, Y. Uchida, S. Tanaka, M. Miyamoto, Y. Oumi, At. Mizusawa, T. Kubo, N. Nishiyama, "Design of Zr- and Al-Doped *BEA-Type Zeolite to Boost LDPE Cracking," ACS Omega, 7, 12971–12977 (2022).
[4] S. Kokuryo, K. Tamura, K. Miyake, Y. Uchida, M. Miyamoto, Y. Oumi, A. Mizusawa, Ta. Kubo, N. Nishiyama, "High coke deposition resistance by Cr loading on zeolite defects: reduced regeneration in cracking reactions," Catal. Sci. Technol., 12, 7270-7274 (2022).
[5] S. Kokuryo, K. Tamura, S. Tsubota, K. Miyake, Y. Uchida, A. Mizusawa, T. Kubo, N. Nishiyama, "Coking Reduction of Cr-loaded Beta Zeolite during Polymer Cracking: Hydrocracking of Aromatics by Synergistic Effect of Cr6+ and Zeolitic Acid Sites" ChemCatChem, 15, e202300461 (2023).

ABSTRACT

Development of plastic composites using recycled plastic and natural fiber

Mariatti Jaafar
School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia,
Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia

mariatti@usm.my

The global generation of wastes is in continuous growth over the last decades, due to the increasing population, industrial development and consumption model. Particularly, plastic consumption has grown at an enormous rate in the past two decades, and so has grown the quantity of plastic wastes, thus becoming a global emergency. Plastic waste is one of the major worldwide problems that cause pollution and are dangerous to human health. One method for reducing plastic waste is by recycling and produce new materials. Plastic composites with combination of natural fibers turns out to be an interesting alternative to incorporate plastic wastes in their formulations, producing environmentally friendly materials. Recently we have investigated the potential use of recycled polypropylene (PP) and polyvinyl chloride (PVC) in combination with natural fibres (oil palm empty fruit bunch (OPEFB), wood fiber and kenaf bast fiber), in the production of plastic composites-type materials of high sustainability. Fabrication and mechanical performance of the plastic composites, as well as fire properties and morphology will be presented.

ABSTRACT

Development of high performance heterogeneous catalysts toward valorization of renewable resources

Sho Yamaguchi
Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University

yamaguchi@cheng.es.osaka-u.ac.jp

The biomass-derived compounds have attracted significant attention as a carbon-neutral resource with the potential to minimize the CO_2 emissions. The biogenic platform chemicals such as polyols, sugars, furanic compounds are identified as the best suited alternatives for petroleum-derived chemicals. However, transformation of these oxygenated molecules to valuable chemicals often encountered low selectivity due to the high reactivity of the biogenic compounds. Recently, we have developed high performance nanostructured heterogeneous catalysts for highly selective transformation of biogenic oxygenates. Herein, our research on the catalyst design for the selective transformation of biogenic compounds will be presented.

ABSTRACT

Co-pyrolysis of biomass waste with plastic waste for bio-oil production

Mohd Azmier Ahmad
School of Chemical Engineering, Universiti Sains Malaysia

chazmier@usm.my

In Malaysia, oil palm solid wastes are an attractive biomass source for biofuels production due to the large scale of wastes generated annually. This study investigated the co-pyrolysis of oil palm empty fruit bunch (EFB) with low-density polyethylene (LDPE) in a fixed-bed reactor. The co-pyrolysis of EFB-LDPE blend were employed at optimum preparation conditions of 534 °C, 57.2 (w/w%) and 33 °C/min for pyrolysis temperature, EFB:LDPE blending ratio and heating rate, respectively which resulted in bio-oil yield of 70.8% and hydrocarbon content of 64.6%. Further upgrading of bio-oil was performed via co-pyrolysis of LDPE-EFB over clinoptilolite catalyst. The influence of pyrolysis temperature as well as catalyst:feedstock ratio on product yields and bio-oil chemical compositions were investigated. The optimum conditions of clinoptilolite catalyst:feedstock mass ratio of 0.2 and temperature of 500 °C resulted in bio-oil yield of 72.6% and hydrocarbons yield of 77.8% with high HHV value of 44.7 MJ/kg.

ABSTRACT

Development of multifunctional alloy catalysts for carbon neutralization and recycle

Yuki Nakaya
Division of Applied Chemistry, Graduate School of Engineering, Osaka University

nakaya@chem.eng.osaka-u.ac.jp

We focus on the development of heterogeneous solid catalysts based on functional multi-element inorganic materials that are highly efficient for carbon neutralization and recycle. For carbon neutralization, we developed Pt-based multinary alloys that are highly efficient for the oxidative dehydrogenation of propane using CO_2 ($C_3H_8 + CO_2 \rightarrow C_3H_6 + CO + H_2O$). This reaction is capable of light olefin production, effective and stoichiometric CO_2 utilization, carbon neutralization of the existing industrial petroleum processes. Ternary Pt-Co-In alloy catalyst supported on CeO_2 exhibits high C_3H_8 conversion, C_3H_6 selectivity, CO_2 utilization efficiency, and long-term stability, which are all the world-record performance. We improved the catalytic performance of this catalyst by further multi-metallization, from the Pt-Co-In ternary system to the PtCoNiSnInGa hexanary “high-entropy” system. Due to the entropy effect, in which the mixing entropy on alloying makes the formation free energy on alloying significantly negative, the thermodynamic stability of alloy phase was greatly enhanced. The $PtCoNiSnInGa/CeO_2$ catalyst showed much greater stability and reusability than $PtCoIn/CeO_2$. For the carbon recycle direction, we recently started the study on plastic decomposition using multinary alloy catalysts. It has been known that polyolefins such as polyethylene or polypropylene can be cracked into lighter alkanes using metallic catalysts under the presence of hydrogen. We are now trying to develop new catalysts that can produce alkanes corresponding to fuel fraction including gasoline and diesel oil in higher content compared with reported catalytic systems, by designing multinary alloy catalysts capable of this molecular transformation.

ABSTRACT

Fabrication of thermochromic films as smart temperature sensor

Chan Qi-Hua, K.I. Ku Marsilla[a]*
[a]School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia
14300 Nibong Tebal, Pulau Pinang, Malaysia.

*ku_marsilla@usm.my

This research focuses on the development of intelligent thermochromic sensors using polylactic acid (PLA) and commercial thermochromic black pigments (TCB) at various concentrations (0-3 wt.%) via melt blending. Additionally, polyethylene glycol (PEG) plasticizers (10 wt.%) with molecular weights of 1400, 2250, and 6000 were employed to investigate their impact on thermochromic behavior. Color transition and reversion dynamics were analyzed at different temperatures (-10, 10, 35, and 50 °C) through colorimetric assessment. The study also evaluated thermal aging and repetitive temperature cycling effects. Results indicate that higher TCB pigment content darkened the initially transparent PLA, enhancing color change visibility, particularly at 2 wt.% pigment concentration. Color alterations remained consistent post-thermal aging and over five heating (50 °C) and freezing (-10 °C) cycles. Incorporating PEG6000 improved pigment dispersion and distribution within the matrix, slightly intensifying color change. Notably, plasticizer addition did not compromise thermochromic properties' resilience to thermal aging or reproducibility. The resulting samples exhibited reversible thermochromism within a temperature range of 10-35 °C, suggesting suitability for smart packaging applications.

ABSTRACT

Development of biodegradable plastic wrapping materials of pathological waste buried in soil environment

[a]Nuraina Hanim Mohd Nizam, [a]*Ku Marsilla, [b]Sharifah Emilia Tuan Sarif, [c]Oh Wen Da
[a]School of Materials and Mineral Resources Engineering, Engineering Campus, Universiti Sains Malaysia, Penang, Malaysia.
[b]Department of Pathology, School of Medical Sciences, Health Campus, Universiti Sains Malaysia, Kubang Kerian, Kelantan, Malaysia.
[c]School of Chemical Sciences, Universiti Sains Malaysia, 11800 Penang, Malaysia.

*ku_marsilla@usm.my

This experimental investigation explores the efficacy of utilizing a formalin neutralizer and a low-cost biodegradable wrapper to mitigate the health and environmental risks associated with the deep burial disposal of pathology samples containing formalin. Despite incineration being the predominant method for disposing pathological waste due to its effectiveness, it is marred by its high cost and adverse environmental impacts. Moreover, there are religious considerations regarding the appropriate disposal of human waste, which adds complexity to waste management practices. Deep burial has emerged as a potential alternative, recognized across various religious beliefs, and perceived as a cost-effective solution. However, the ecological impact of burying formalin-fixed pathological waste remains uncertain. To address this gap, a stimulated burial approach model will be employed in this study. The compatibilizer agent poly(butylene adipate-co-terephthalate-grafted-succinic anhydride (PBAT-g-SA) will be utilized in the blown film processing of thermoplastic (TPS) and PBAT blends to fabricate the biodegradable wrapper. The mechanical properties, formalin permeability test and soil burial properties of the blown film biodegradable wrapper will be thoroughly characterized. The goal is to establish a groundbreaking pathological waste management protocol that is both safe and cost-effective, while also adhering to Shariah principles. The implications of this research extend beyond Malaysian hospitals, potentially influencing waste management practices. Furthermore, the findings could contribute to the development of new guidelines for healthcare management systems in Malaysia.

ABSTRACT

Coke-resistant Ce-based multi-element oxide supports for CO_2-propane dehydrogenation

Li Duanxing and Furukawa Shinya*
Division of Applied Chemistry, Graduate School of Engineering, Osaka University.
TEL: 06-6879-7844, E-mail: furukawa@chem.eng.osaka-u.ac.jp

Achieving net-zero CO_2 emission in the chemical industry by the year of 2050 presents a challenging task. Using CO_2 as the oxidant for propane dehydrogenation not only increases value-added ethylene yield but also effectively utilizes CO_2, contributing to the carbon neutrality of the chemical industry. Coke deposition decreases catalyst lifetime and limits the industrial application of this reaction. We previously reported Pt-Co-In/CeO_2 catalyst with world-leading performance for CO_2-C_3H8 dehydrogenation (*Nat. Catal.*, **2022**, *5*, 55). In this catalyst system, CeO_2 support removed coke via combustion with lattice oxygen, leading to outstanding catalyst lifetime.

In the current study, to further enhance the coke combustion ability of Ce-based oxides, we tried to systematically improve its lattice oxygen-releasing ability. We introduced cations with various ionic radii of groups 3 and 4 to the Ce^{4+} site of CeO_2 to induce lattice distortion. The multi-element oxides were synthesized with a sol-gel method. Typically, metal precursors and PVP were mixed in H_2O and dried to form gel. The obtained gel was calcined in flowing air to form multi-element oxides. XRD and H_2-TPR were performed to evaluate the crystal structure and oxygen-releasing ability of prepared samples.

We found that multi-element oxides with fluorite structure showed improved O-releasing properties, which could potentially serve as catalyst supports with enhanced coke removal ability, improving catalyst lifetime in the future.

ABSTRACT

3D printing of biodegradable PLA/PBAT blends through the 3-glycidoxypropyltrimethoxysilane (GPTMS)

Massara Samsul Baharin, Nurul Fazita Binti Mohammad Rawi[a], K.I. Ku Marsilla[b]*

[a]School of Industrial Technology, Universiti Sains Malaysia, 11800 Pulau Pinang, Malaysia
[b]School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia
14300 Nibong Tebal, Pulau Pinang, Malaysia
*ku_marsilla@usm.my

Blends of poly(lactic acid) (PLA) and poly(butylene adipate terephthalate) (PBAT) are frequently utilized for producing filaments in fused deposition modelling (FDM) 3D printing. However, due to their differing molecular structures and chemical compositions, these materials often encounter compatibility issues. To address this challenge, 3-Glycidoxypropyltrimethoxysilane (GPTMS) is introduced as a compatibilizer, enhancing the mechanical performance and compatibility of PLA/PBAT blends. This study focuses on compounding polymers in ratios of 80PLA/20PBAT and 80PBAT/20PLA, with a consistent addition of 2phr GPTMS, using twin-screw extrusion to manufacture the filament. Subsequently, the properties of both the blend and printed specimens were characterized through various analyses, including Fourier transform infrared spectroscopy (FTIR), tensile testing, melt flow index measurement, thermal analysis, and morphology observation. The objective is to attain an optimal blend that ensures excellent printability and delivers high-quality outcomes in 3D printing applications. This approach represents a significant advancement in addressing the compatibility challenges of PLA/PBAT blends, offering promising prospects for the production of 3D printing filaments.

ABSTRACT

Reductive amination of carboxylic acids using a Pt–Mo Catalyst for sustainable alkylamine syntheses

Katsumasa Sakoda[1], Sho Yamaguchi[1,2], Takato Mitsudome[1,2], Tomoo Mizugaki[1,2,3]*
[1] Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan.
[2] Innovative Catalysis Science Division, Institute for Open and Transdisciplinary Research Initiatives (ICS-OTRI), Osaka University, Suita, Osaka 565-0871, Japan
[3] Research Center for Solar Energy Chemistry, Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan

*mizugaki.tomoo.es@osaka-u.ac.jp

Reductive amination of carboxylic acids under mild conditions is achieved by using a Pt–Mo catalyst. Notably, the Pt–Mo catalyst operates well even under atmospheric pressure of H_2. Various carboxylic acids, including biomass-derived fatty acids, are successfully transformed into the corresponding alkylamines in excellent yields. The unique catalysis of Pt–Mo will make a significant contribution to establish a future sustainable process for alkylamine synthesis.

ABSTRACT

A sustainable route from waste to wealth: Upcycling disposable face masks into value-added hydrocarbons for carbon nanomaterials production via innovative pyrolysis setup

Kai Qi Tan[a], Siew Chun Low[b]
School of Chemical Engineering, Engineering Campus, USM

[a]kaiqi98@gmail.com
[b]chsclow@usm.my

Pyrolysis of plastic or polymer waste, specifically disposable face masks, often overlooks the potential of the gas product and its role as a valuable precursor for carbon nanotube (CNT) production. To address this issue, this study focuses on the pyrolysis of disposable face masks into non-condensable hydrocarbon gases. Typical studies increase the operating temperature to enhance the output gas yield. However, as pyrolysis process is energy-intensive, increasing the operating temperature requires more energy for the reaction. In this study, the gas yield was enhanced by modifying the reactor configuration without increasing the operating temperature. Results showed that adding a layer of cold zone in the reactor produced a 1.6-fold higher gas yield due to the retainment of volatiles for end chain scission mechanism. The liquid product analysis also showed that the retention of the cold zone promotes further cracking of volatiles into lighter liquids. Using the same reactor configuration, the output gas composition was obtained from the pyrolysis of disposable face masks without any component segregation. The comparison of the output gas composition with the reported CNT precursor composition showed that it is feasible to upgrade the output gas into CNT. Hence, no segregation is required before sending the disposable face masks into pyrolysis process, creating a more sustainable production with a significant economic return.

ABSTRACT

Coke deposition resistance by Cr^{6+} loading on zeolite defects: Reduced regeneration in low-density polyethylene cracking

Shinya Kokuryo[a]*, Koji Miyake[a], Yoshiaki Uchida[a], Norikazu Nishiyama[a]

[a] Division of Chemical Engineering, Graduate School of Engineering Science, Osaka University, Japan

*e-mail: skokuryo@cheng.es.osaka-u.ac.jp

The increase in plastic waste has induced critical environmental problems, thereby plastic waste should be managed better than ever. Chemical recycling (CR) has recently been considered as an attractive candidate to solve this problem[1]. In CR, plastic wastes can be converted into valuable products (e.g., monomers or petrochemical feedstocks). Recently, there has been much research on the catalytic cracking of polymers using zeolites. However, the deactivation of zeolites due to coke deposition is a serious problem. Although significant efforts have been devoted toward coking inhibition, the previous methods are not suitable for polymer cracking. Therefore, new approaches are needed to inhibit coke deposition without degrading catalytic performance.

Recently, it has been reported that Cr^{6+} connected to silanol groups in zeolites is stable and promotes the dehydrogenation of light alkanes[2]. Since many alkanes are produced in the catalytic cracking of polyolefins, these alkanes are considered dehydrogenated during the reaction. Moreover, aromatics, which are coke precursors, are decomposed via hydrogenation on acid sites of the zeolite. Therefore, hydrogen is generated from the alkanes in the product at the Cr^{6+} sites, and the hydrogenation and decomposition of the coke precursor at the zeolite acid sites are thought to have inhibited coke deposition. In this study, Beta zeolite was doped with Cr^{6+}, and the effect of the synthesized catalysts on coke inhibition was examined by the catalytic cracking of low-density polyethylene (LDPE).

We prepared Cr/Beta(x) samples at a mass ratio of Cr/Beta = x/100 (x = 0.1, 0.5, 1, 5, and 10) by the impregnation method. UV-Vis spectroscopy was conducted to investigate the valency of Cr species in zeolites. The absorption band derived from Cr^{3+} was only detected for Cr/Beta(5) and Cr/Beta(10), indicating the presence of Cr_2O_3. Moreover, the peaks derived from Cr^{6+} were detected for all Cr-loaded samples. We conducted FT-IR spectroscopy to investigate the detailed chemical states of the Cr^{6+} species. The Cr/Beta samples showed peaks attributed to the Si-O-Cr^{6+} vibration. Moreover, the weak band assigned to the Cr-O or Cr=O vibration in the Cr^{6+} species was observed. Thermogravimetric (TG) curves for the catalytic cracking of LDPE under N_2 flow at a heating rate of 5 K min^{-1} were measured to investigate the catalytic performance of Cr/Beta. The decomposition temperature increased slightly when the Cr loading was higher than 5 wt.%, but the Cr loading did not significantly decrease the catalytic activity of Beta zeolite.

In the reaction, Cr^{6+} species connected to silanol groups performed as a catalyst for the dehydrogenation of light alkanes in the products, and aromatics were decomposed via hydrogenation at the zeolite acid sites. In the ethane dehydrogenation reaction, Cr/Beta generated H_2 at a lower temperature than pristine Beta. And it was confirmed that coke precursor was decomposed at zeolite acid sites by using toluene and cyclohexane. Therefore, the synergetic effect of the H_2 generation by Cr^{6+} and the decomposition of coke precursor by zeolite acid sites led to coking inhibition. As a result, it was possible to recycle the obtained catalysts several times without regeneration during the catalytic cracking of LDPE.

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ABSTRACT

Assessing environmental impacts of facemasks via pyrolysis process and its life cycle assessment (LCA)

Amirah Ayu Kartika binti Asman
amirahasman7@gmail.com

A key hurdle in navigating the aftermath of COVID-19 pandemic is effectively handling the plastic waste generated by personal equipment, while also grappling with the broader issue of plastic pollution. The widespread adoption of face masks as a primary preventive measure during the pandemic has led to a significant surge in demand in recent years. This study employs pyrolysis process of facemasks and its environmental impact through life cycle assessment. Throughout the pyrolysis process, several face masks types, including 3ply (polypropylene-based), KF94 (polypropylene-based) and N95 (polyester-based) were used as individual feedstocks using prescribed reactor configuration. The pyrolysis of 3ply and KF94 produce 62.9% and 65.42% propylene gas respectively since total feedstocks were predominantly consisted of PP as the major plastic component. The N95 mask produces the most CO_2 gas with 49.65%, due to the presence of polyester in the face mask shell layer, thus causing a decarboxylation process at temperatures ranging from (400-480℃). The results of product distribution from pyrolysis process (char, bio-oil, gas) demonstrate that different types of face masks used, have a significant effect on product distribution, resulting in various impacts on the environment. Marine aquatic ecotoxicity has the greatest environmental impact, followed by global warming and human toxicity in third place.

ABSTRACT

Tough host–guest composite hydrogels reinforced by dual supramolecular cross-linkers

May Myat Noe[1], Akihide Sugawara[1], Takashi Konishi[2], Yoshinori Takashima[3,4,5,6], and
Hiroshi Uyama[1]

[1] Grad. Sch. of Eng. Osaka Univ., [2]GSHES, Kyoto Univ., [3] Grad. Sch. of Sci. Osaka Univ., [4] FRC, Osaka Univ.,
[5] OTRI, Osaka Univ., [6] Inst. for Advanced Co-Creation Studies, Osaka Univ.
may@chem.eng.osaka-u.ac.jp

Conventional chemically cross-linked hydrogels are mechanically fragile because stress concentration occurs due to the randomly arranged cross-links, leading to permanent fracture of the network. This drawback limits the practical applications of hydrogels.[1] There are promising approaches, in which the network is constructed by reversible bonding as cross-linker such as covalent bonding, based on enamines, disulfides, and Diels-Alder reactions, and physical bonding, like ionic interactions, hydrophobic interaction, and hydrogen bonding.[2] These bonding and interactions can be dissociated sacrificially and reformed reversibly upon the deformation of materials, contributing to high mechanical performance of hydrogels.

In this work, tough composite hydrogels were prepared by designing the physically cross-linked network through host–guest (HG) interaction utilizing dual supramolecular cross-linking. β-Cyclodextrin (β-CD) and adamantane (Ad) were selected as host and guest molecules, respectively. The first cross-linker for the hydrogel network is HG cross-linker which was incorporated between the polymer matrices to form supramolecular hydrogels. The second cross-linking was incorporated between the matrix and the filler. Ad-modified cellulose nanocrystal (Ad-CNC) was introduced as the supramolecular filler and enabled the HG complex formation with β-CD on the matrix. The synergistic effects of these supramolecular cross-linking upon the mechanical properties were evaluated.

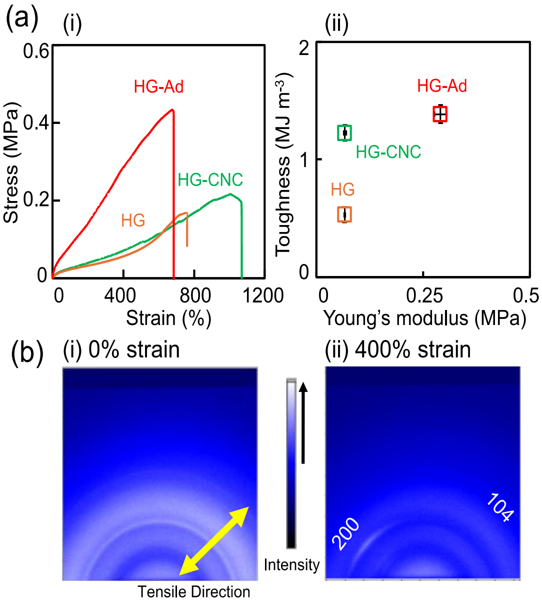

Figure 1. (a) Stress-strain curves of (i) HG-Ad, HG-CNC and HG gels and (ii) Relationship between their Young's modulus and toughness. (b) 2D WAXS images of HG-Ad gel at (i) 0% and (ii) 400% strain.

The HG-Ad gel with dual supramolecular cross-linking was prepared by free-radical copolymerization of acrylamide and acrylamide derivatives of β-CD and Ad in the presence of Ad-CNC. The HG-CNC and HG gels were prepared in the presence of unmodified CNC, and in the absence of filler, respectively. The dual cross-linked HG-Ad gel exhibited significant enhancement in the mechanical properties. The increasing values of both toughness and Young's modulus were obtained in the HG-Ad gel whereas the single cross-linked hydrogels, HG-CNC and HG gels showed about 5 times smaller in the values of Young's modulus (**Fig. 1a**). Therefore, this is attributed to the cooperation of dual cross-linking in the network. Wide-angle X-ray scattering (WAXS) revealed the preferred orientation of CNC along with polymer network upon stretching.[3] In **Fig. 1b**, at the 400% strain, the intensity of the 200 reflection of CNC increased in the vertical direction and that of the 104 reflection in the tensile direction. It showed that CNC was oriented along with polymer extension. This research will be the versatile approach for preparing tough hydrogels by incorporation of dual supramolecular cross-linkers in networks.

Keywords: Cellulose Nanocrystal, Composite, Hydrogel, Host–Guest Interaction, Cyclodextrin

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ABSTRACT

Catalytic cracking of waste cooking oil over activated carbon supported trimetallic oxide catalysts

Muhammad Shafizruddin Firdaus Bin Fazli-Ku & Ching Thian Tye*
School of Chemical Engineering, Engineering Campus, Universiti Sains Malaysia,
14300 Nibong Tebal, Pulau Pinang, Malaysia

*Email: chcttye@usm.my

The energy generation and transportation industries can benefit from the conversion of waste cooking oil into biofuels, which are renewable and sustainable fuels. This study investigated the catalytic cracking of used cooking oil over a series of activated carbon supported metal oxide catalysts for the production of renewable fuels. The reactions were carried out in a batch reactor at 400 °C for 1 hour. Metals such as Ni, Fe, Cu, Co, Zn, Ce, La, Mg and Ca were incorporated to activated carbon to prepare single metal oxide, bimetal oxides or trimetal oxides with a metal loading of 10 wt%. The results of this study revealed that trimetal oxide performed better than bimetal and monometal oxide catalysts, especially in terms of the liquid yield and coke yield. It is believed that the synergistic effect of three metals improve deoxygenation. NiO-Fe_3O_4-ZnO/AC was identified as the best catalyst in this study which has achieved 92.42 wt% liquid yield with only 0.05 wt% coke formation. Besides, the liquid product was found to have 49 vol% diesel range with 52.44% alkane (hydrocarbons).

Keywords: waste cooking oil; catalytic cracking; renewable energy; trimetal oxides; renewable fuel

ABSTRACT

Effect of poly(vinyl alcohol) on the physical properties of starch-based composite films

Ying Yao, Yu-I Hsu[1,*] and Hiroshi Uyama[1]

[1] Department of Applied Chemistry, Graduate School of Engineering, Osaka University, Suita, Osaka 565-0871, Japan

*Corresponding author: yuihsu@chem.eng.osaka-u.ac.jp

In recent years, the research and development of biomaterials based on various natural polymers (such as polysaccharides, proteoglycans and proteins) for use in biocompatible wound dressings and drug delivery systems have gained widespread attention. Among them, starch-based films have emerged as one of the most attractive and practical biodegradable biomaterials due to their advantages of low cost, biocompatibility, and renewability. Poly(vinyl alcohol) (PVA) is highly water-soluble, non-carcinogenic, and flexible, and widely used in chemical and medical industries. Therefore, PVA is considered a suitable candidate polymer for blending with starch to enhance the physicochemical properties of the resulting blend films. However, most studies have utilized single grades of PVA blended with starch, limiting the potential applications of PVA/Starch blend films. To address this limitation, this study aimed to prepare starch-based composite films via solution casting method using PVA with different degree of saponification (DS) and degree of polymerization (DP) and two types of modified starches: sodium octenyl succinate starch (OctS) and hydroxypropyl starch (HPS). The results indicated that the strength and flexibility of the blended films were improved by the addition of PVA. PVA/Starch/Gly-7/3/3 exhibited enhanced transparency, mechanical properties, thermal stability, and water absorption. The starch-based blend films with a higher DP of PVA had better tensile properties, as indicated by the larger tensile strength and elongation at break. In addition, the presence of carboxyl substituents resulted in better stability of the OctS-based films in neutral and acidic solutions. In conclusion, incorporating different PVA grades into starch-based films offers an exciting avenue for improving their properties, thereby expanding their potential applications as biocompatible and versatile biomaterials in the medical field.

Keywords: blend films, hydroxypropyl starch, sodium octenyl succinate starch, PVA, biomedical applications

ABSTRACT

The utilisation of organic domestic waste to high grade smokeless fuel (HiGraS-Fuel) using a carbonizer

Muhammad Safiuddin Rosli
Center for Global Sustainability Studies, Universiti Sains Malaysia

msafiuddinr@student.usm.my

The proliferation of organic domestic waste poses a significant challenge to sustainable waste management systems. Despite advancements in waste management practices, the current approach fails to efficiently address the growing volume of organic waste. Although waste segregation is practiced in certain households, the overall infrastructure remains highly dependent on landfill areas as dumping sites. The research aims to utilize thermochemical decomposition of organic materials for producing a valuable carbon-based product. The thermochemical technique will yield a product named High Grade Smokeless Fuel (HiGraS-Fuel), suitable for cooking, domestic heating, and plant fertilization. The test involved removing moisture and volatile matter, leading to the conversion of the remaining material into char. The mass yield factor of HiGras-Fuel after the processing procedure has yielded a noteworthy outcome surpassing 60%. On average, the operational duration ranging from 3 to 5 hours incurs a yield cost of RM0.27. Consequently, the resultant product can be marketed at 30% of its estimated sale value, which amounts to RM0.351. The contrast in market valuation between HiGras-Fuel and conventional fuel cells, exemplified by coal priced at RM0.52, underscores the emergence of a substantially more economical and environmentally benign alternative. This discrepancy manifests as a notable reduction of 67.5% in market value, thereby positioning HiGras-Fuel as an attractive proposition in terms of both cost-effectiveness and ecological sustainability when juxtaposed with traditional fuel sources like coal. The conversion of waste into a valuable resource, as exemplified by HiGras-Fuel, epitomizes the concept of a circular economy. This approach not only underscores the tenets of sustainable development goals but also addresses the imperative of reducing greenhouse gas emissions, particularly stemming from methane emissions at landfill sites. In effect, the utilization of HiGras-Fuel represents a tangible strategy for mitigating environmental impact while concurrently advancing economic and environmental sustainability objectives.

ABSTRACT

Effect of Brønsted acidity of zeolite on catalytic cracking temperature of low density polyethylene

S. Tsubota[1]*, K. Miyake[1], Y. Uchida[1], N. Nishiyama[1]
[1]Nishiyama lab., Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka, 560-8531, Japan

*s.tsubota@cheng.es.osaka-u.ac.jp

Introduction

The global production and consumption of plastics have increased at an amazing rate over the last few decades, and plastics that are disposed of without proper treatment cause various environmental problems.[1] Therefore, immediate and decisive action is needed to resolve this. Chemical recycling (CR) is a process in which waste plastics are chemically decomposed and reused as raw materials for chemical products, and it has been attracting attention in recent years as a beneficial recycling process in the chemical industry.[2] In particular, catalytic cracking using zeolites has been the subject of much research in recent years because it can lower the cracking temperature compared to thermal cracking and selectively recover C1-C6+ gases.[3]

Zeolite is an inorganic compound with a characteristic pore structure that also functions as a solid acid catalyst. Developing the best zeolite for polymer cracking requires both Lewis acidity and Brønsted acidity approaches. Our group has successfully lowered the cracking temperature of low-density polyethylene by increasing the Lewis acidity of zeolites.[4] However, the effect of zeolite Brønsted acidity on polymer cracking is still unclear, which hinders the development of suitable catalysts for catalytic cracking. In this study, we prepared MFI-type zeolites with precisely controlled Brønsted acidity and morphology based on our previous works and quantitatively elucidated the effect of Brønsted acidity on polymer cracking. In particular, we investigated the cracking temperature of polymer over these zeolites using a thermogravimetric analysis (TGA).

Materials and Methods

MFI-type zeolite T-x was synthesized by hydrothermal synthesis (T=Al, Ga, Fe and x=Si/T=50, 80, 100). In addition, zeolite with controlled outer surface acid sites was synthesized by coating Al-50 with silicalite-1. Al-80 with a smaller particle size was also synthesized by dry gel conversion. The activity of zeolites in LDPE cracking was evaluated by thermogravimetric (TG) analysis at a heating rate of 5 °C /min to 600 °C under N2 and air atmosphere, respectively.

Results and Discussion

The NH_3-TPD measurement allows us to measure the heat of adsorption of ammonia, which is an indication of acid strength, in addition to the acid amount of the catalyst. The acid strength of the catalysts was quantified by the Niwa et al. plot method, and it was confirmed that the acid strength became weaker in the order of Al-x, Ga-x, and Fe-x.[5]

Figure 1(a) shows the LDPE cracking temperature T_{half} for each sample. It was confirmed that the T_{half} decreased with increasing the amount of any metal introduced into the zeolite. However, the lowest cracking temperature depends on the type of metal introduced, i.e., the acid strength of the zeolite. Specifically, it was observed that zeolites with higher acid strength resulted in lower cracking temperatures.

Al-80 with different particle sizes was also synthesized and used for catalytic cracking of LDPE to compare their T_{half}. The larger the outer surface area, the lower the temperature. This result indicates that acid sites on the outer surface also have an effect on contact LDPE cracking.

Figure 1(b) compares the T_{half} of core-shell zeolite with that of uncoated zeolite. The cracking temperatures were higher than those obtained with the uncoated zeolite. This indicates that the acid sites on the external surface of the zeolite have a significant effect on the catalytic cracking of LDPE. Meanwhile, the T_{half} values of the core-shell zeolites were lower than that of silicalite-1, indicating that Brønsted acid sites inside the zeolite crystals also contributed to lowering the catalytic cracking temperature of LDPE. From the above, it was found that the catalytic cracking of plastics using zeolites takes place in the following two steps. (1) The polymer is cracked at the acid sites on the external surface until it is large enough to enter the zeolite pore. (2) The polymer decomposed in step (1) is further decomposed at the acid site inside the pore to obtain C1-C6+ gas.

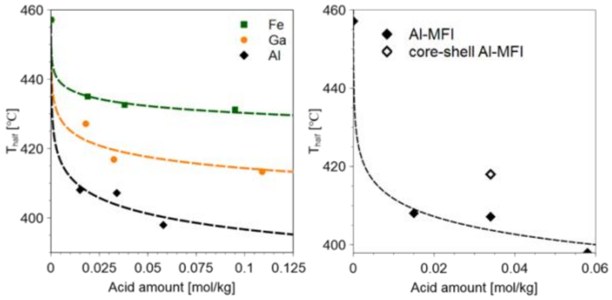

Figure 1. (a) Relationship between the amount of acid in zeolite and T_{half} value and (b) Comparison of T_{half} between uncoated and core-shell zeolites (Al-MFI).

Significance

This study will provide insight into the Brønsted acidic properties of zeolite catalysts on LDPE cracking, which will serve as the basis for future catalyst design guidelines.

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ABSTRACT

Electrifying rural areas: The story of Sarawak

Janet Wan
Center for Global Sustainability Studies, Universiti Sains Malaysia

janetwan@student.usm.my

In 2009, only 56% of the rural population in Sarawak had access to reliable 24/7 electricity supply, though overall domestic coverage is already almost 100%. Dispersed areas in rural areas bring the challenges of remoteness and limited accessibility issues in the effort to provide more reliable 24-hour electricity and to achieve a prosperous rural vision. In the spirit of Sustainable Development Goal 7, to ensure access to affordable, reliable and renewable energy for all, Sarawak uses renewable energy like hydro and solar energy to provide reliable, renewable, clean and affordable power across remote and inaccessible areas in the region and not all are feasible for grid solutions. Information used in this paper is collected from articles or publications by other authors, annual reports, news reported in newspapers and policy documents to analyse and understand the initiatives that have been put into place by Sarawak government to achieve 100% electrification by 2025. This paper will show how the initiatives implemented by both the Federal and State Governments with the collaboration of the private sector and local communities to provide 24-hour electricity access to places that are deemed to be remote and inaccessible can be done using green renewable energy available abundantly in the area.

Keywords: Sarawak, Rural Areas, Sustainable Development Goal 7, Electricity Access

ABSTRACT

Nickel carbide nanoparticle catalyst for direct hydrogenation of furfural to tetrahydrofurfuryl alcohol

Taiki Kawakami[1], Sho Yamaguchi[1,2], Takato Mitsudome[1,2], Tomoo Mizugaki[1,2,3]*
[1] Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan.
[2] Innovative Catalysis Science Division, Institute for Open and Transdisciplinary Research Initiatives (ICS-OTRI), Osaka University, Suita, Osaka 565-0871, Japan
[3] Research Center for Solar Energy Chemistry, Graduate School of Engineering Science, Osaka University, 1-3 Machikaneyama, Toyonaka, Osaka 560-8531, Japan

*mizugaki.tomoo.es@osaka-u.ac.jp

The hydrogenation of furfural (FUR) to tetrahydrofurfuryl alcohol (THFA) is an attractive reaction to produce valuable THFA directly from biomass-derived feedstock. Non-precious metal-based catalysts have been developed for this reaction, but harsh reaction conditions are usually required to achieve sufficient THFA yield. Herein, we report that nickel carbide nanoparticles (Ni_3C NPs) act as an efficient heterogeneous catalyst for the direct hydrogenation of FUR into THFA in water. The Al_2O_3-supported Ni_3C NPs (Ni_3C NPs/Al_2O_3) is the first non-precious metal catalyst providing THFA in excellent yield under just 1 bar of H_2. In this presentation, we discuss the origin of the high activity of Ni_3C NPs/Al_2O_3 based on control experiments, kinetic studies, and various spectroscopic methods.

ABSTRACT

Transforming sawdust into high-performance Co-N-co-doped hydrochar catalyst for ciprofloxacin removal via peroxymonosulfate activation

Ganapaty Manickavasagam, Wen-Da Oh*
School of Chemical Sciences, Universiti Sains Malaysia, 11800, Penang, Malaysia

*Corresponding author: ohwenda@usm.my

A total of 9.83 million m^3 of sawdust is generated as carbon-rich biomass waste annually in Malaysia. With the aim of transforming biomass waste into valuable materials for environmental application, sawdust was employed to prepare a series of Co-N-co-doped hydrochar (Co-N-HC) with varying Co content from 0.5 to 3.0 wt.% via a one-step acid-assisted hydrothermal method. Characterization study using X-ray diffraction (XRD) technique indicated that the Co-N-HC consists of amorphous carbon with short-range order, while the field emission scanning electron microscope (FESEM) study illustrated that the surface of Co-N-HC is relatively smooth with low porosity. The Co-N-HC was employed as peroxymonosulfate (PMS) activator to remove ciplofloxacin (CIP) from contaminated water. The Co-N-HC with 0.95 wt.% of Co exhibited the best performance with 87% CIP removed in 60 min (pseudo first-order rate constant, k_{app} = 0.034 min^{-1}). Apparently, the k_{app} was positively correlated with increasing the catalyst loading and PMS dosage, while pH 7 was found to be the optimum pH for CIP removal. The presence of humic acid reduced the effectiveness of the Co-N-HC in removing CIP, whereas interfering ions (i.e., Mg^{2+}, Ca^{2+}, NO_3^-) slightly increased the removal rate of CIP. The Co-N-HC can be applied as PMS activator in secondary effluent and river water, albeit with reduced k_{app} values of 0.021 and 0.027 min^{-1}, respectively. Based on the scavenger study, the contributions of $^1O_2$, $^\bullet OH$, $SO_4^{\bullet -}$, and other pathways was estimated at 18%, 62%, 6%, and 14%, respectively. The dominant pathway of $^\bullet OH$ could be associated with the redox cycle of Co^{2+}/Co^{3+}, whereas the minor pathways of nonradical electron-transfer, $^1O_2$, and $SO_4^{\bullet -}$ could be related to pyridinic N, graphitic N, and redox cycle of Co^{2+}/Co^{3+}, respectively. The Co leaching was found to be about <2% of the total Co-content in the co-doped hydrochar and mineralization extent was found to be 70%. Overall, this work provided a promising approach to develop high performance hydrochar as carbocatalyst for CIP removal.

Keywords: Hydrochar; Metal-heteroatom-co-doping; Peroxymonosulfate; Hydroxyl radical; Ciprofloxacin

ABSTRACT

Study on multinary alloys and oxides as efficient catalysts for the oxidative dehydrogenation of propane using CO_2

Zhang Fang

Division of Applied Chemistry, Graduate School of Engineering, Osaka University

zhangfang@chem.eng.osaka-u.ac.jp

Introduction: With the requirement of zero-emission, how to utilization CO_2 become more important. Using CO_2 to oxidize the propane for dehydrogenation is a promising technology, which can not only consume CO_2 to achieve carbon neutrality, but also bridge the gap for demand for propene. In our previous work, the Pt-Co-Sn ternary catalyst, which has achieved the world's highest level of catalytic activity, propylene selectivity, durability, and carbon dioxide utilization efficiency, was reported.[1] This system utilized the metal alloy: Pt can promote the activation of propane, Co can activate CO_2, and In can inhibit unnecessary side reactions and improve the selectivity of propylene, thus reaching the world's highest level.

Due to the outstanding performance of the ternary alloy, and to improve the performance, using five or more metals, which could be formed high entropy intermetallics (HEI) catalyst, maybe predictable excellent performance. By increasing the number of elements, the thermodynamic stability (more negative ΔG of alloy formation) can be improved by enhanced "mixing entropy (ΔS_{mix})", which results in much greater stability of catalyst hence more suitability to practical application.

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