Thai Environmental Engineering Journal Vol. 35 No. 3 (2021) : 59-68
www.eeat.or.th

Heat Management Options to Reduce Carbon Footprint of Green Zeolite Faujasite Synthesis from Rice Husk Ash

Patchaya Tobarameekul[1] **and Patcharin Worathanakul**[1,2*]
[1]Department of Chemical Engineering, Faculty of Engineering,
King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand
[2]Center of Eco-Materials and Cleaner Technology (CECT), Science and Technology Research Institute,
King Mongkut's University of Technology North Bangkok, Bangkok 10800, Thailand
[*]E-mail : patcharin.w@eng.kmutnb.ac.th

Abstract

Zeolite could be derived from agricultural waste for green zeolite synthesis technology. It is one of the adsorbents that could reduce carbon dioxide emissions. This research was to develop a novel zeolite faujasite synthesis from rice husk ash as a silica source and to determine the carbon footprint with different heat management options including of crystallization temperature and time. The carbon footprint of zeolite faujasite was evaluated by quantifying the greenhouse gas emissions in terms of carbon dioxide equivalent using the framework of life cycle assessment methodology. The zeolite with 0.3 g was selected as the functional unit to evaluate the impacts of each condition from rice husk ash compared to conventional methods. The utilization of rice husk ash as a silica source instead of a chemical source could reduce the carbon footprint to 0.48 kgCO_2eq, which is 8% less than the conventional one. The highest carbon footprint value was associated with the production stage under high temperature and longtime crystallization. The carbon footprint for successfully synthesized zeolite faujasite derived from rice husk ash with crystallization at room temperature and short time could be reduced by 48%.

Keywords : Rice husk ash; Faujasite; Heat management; Carbon footprint; Green zeolite

Introduction

Global warming and climate change caused by the accumulation of greenhouse gases (GHGs) in the atmosphere are serious environmental problems. Consequently, the aim to decrease carbon dioxide (CO_2) has been an important driver. In recent years, the use of more sustainable, lower cost and high-efficiency adsorbents have received attention because of its global warming impacts. The adsorbent production for CO_2 adsorption, such as amine groups, metal organic frameworks and cryogenics, has a serious environmental impact with high energy consumption, corrosion problems, high production cost and complicated operation [1]. Zeolite is one of the sustainable adsorbents that could reduce CO_2 emissions. Several studies have been performed to develop zeolite for CO_2 adsorption such as zeolite faujasite (FAU) including of linde type A (LTA) and low silica X (LSX) because of its good adsorption [2-4]. However, the zeolite FAU synthesis also has an impact on the environment from the synthesis. Zeolite FAU synthesis has produced around 0.52 kgCO_2eq, accounting for more than 79% of usage step compared to total CO_2 emission [5]. Therefore, it is interesting to find a sustainable technique to reduce CO_2 emission for green zeolite synthesis. The commonly used precursor of conventional zeolite synthesis is produced from the chemical source [6]. To reduce the CO_2 emissions, zeolite synthesis from different sources as a precursor is to replace the chemical source with agricultural waste such as bagasse, palm oil and straw wastes [7-9].

Rice husk ash has a high silica source produced as a by-product from biomass power plants. It is a residual waste and abundantly available in large amounts in Thailand, with a total output of approximately 20% of the weight of rice husk after use for electricity production [10]. As a result, rice husk ash is an excellent green precursor for zeolite synthesis. However, the synthesis technologies are still limited. The good physical properties of zeolite can be achieved with the optimum condition [11]. According to the last review of zeolite FAU synthesis, heat management options from the use of rice husk ash for zeolite synthesis has not been investigated yet. Thus, there is a need for development on synthesis technologies and green precursors.

For the challenge to reduce CO_2 emissions, the evaluation of carbon footprint has become an important tool and widely used to describe the total amount of GHG emissions from a process or product [12]. The carbon footprint is carried out following the life cycle assessment method (LCA) of the product from the extraction of the raw materials to disposal at the end of life in terms of CO_2 equivalents (CO_2eq) to a functional unit of the product [13-15]. It can be used for process improvement to get more environmentally friendly alternatives [16].

The objectives of this research were to develop a novel zeolite FAU synthesis by seeding method from rice husk ash as a silica source and to determine the carbon footprint of zeolite FAU synthesis with different heat management options including crystallization temperature and time. The results could help to reduce energy consumption in the process for environmental impact minimization.

Material and Methods

1. Materials

The rice husk ash with 97.73 %wt. SiO_2 used in this study was from U-Thong biomass Co, LTD., Thailand. The chemicals used for silica extraction and zeolite FAU synthesis included of hydrochloric acid (HCl; 37 wt.% HCl, Merck), sodium aluminate ($NaAlO_2$; Al_2O_3 50-56 wt.% Fe_2O_3 0.05 wt.% and Na_2O 40-45 wt.%, Sigma Aldrich), sodium hydroxide (NaOH; 99 wt.% NaOH, Merck), Sodium silicate (Na_2SiO_3; Na_2O 18 wt.% and SiO_2 63 wt.%, Sigma Aldrich), distilled water and deionized water.

2. Zeolite FAU synthesis from rice husk ash

The extracted silica derived from rice husk ash was used by a solvent extraction method based on obarameekul et al. [4]. Initially, the rice husk ash 5 g was added with 2 molar of NaOH solution and stirred. Silica is extracted in the form of Na_2SiO_3 during this procedure. The solution was filtered and titrated with HCl solution to form a gel compound. After that, the gel compound was aged overnight at 25 °C. The obtained gel was filtered and washed with DI water. Then gel was dried overnight at 110 °C to obtain Na_2SiO_3 powder for zeolite FAU synthesis.

The methodology used for zeolite FAU synthesis at a Si/Al ratio of 0.75 was based on the research of Worathanakul and Rakpasert [6]. Initially, the $NaAlO_2$ and Na_2SiO_3 solutions were prepared by mixing with NaOH and distilled water while stirring. After that, the Na_2SiO_3 solution was mixed into the $NaAlO_2$ solution. The solution was stirred and aged overnight at 25 °C to obtain seed gel. The $NaAlO_2$ and Na_2SiO_3 solutions were prepared in the same way to obtain feedstock gel. The seed gel solution was added to the feedstock gel solution under stirring to obtain the overall gel. The overall gel was aged overnight at 25 °C to form a gel compound. Then the gel compound was heated by oven under different crystallization temperatures (25, 50 and 70 °C) and times (1 and 3 h). The obtained gel was filtered and washed with DI water. Then gel was dried overnight at 110 °C to obtain zeolite FAU. The above methods were repeated with chemicals as a silica source for zeolite FAU synthesis.

3. Carbon footprint of zeolite FAU synthesis

The carbon footprint assessment of products is generally based on life cycle assessment (LCA) following the guidelines in ISO14067:2018 which consists of four main steps:

3.1 Goal and scope definition

The goal of this study was to assess the carbon footprint of zeolite FAU synthesis with different heat management options including crystallization temperature and time. Based on the conventional method of zeolite FAU synthesis, the crystallization temperature and time of 70 °C for 3 h were used [6]. The crystallization temperature and time of zeolite FAU synthesis range from 70 °C to 25 °C for 3 h to 1 h, including using rice husk ash as a silica source instead of chemicals.

The functional unit was 0.3 g zeolite. Since the purpose of this research was to develop a novel zeolite FAU which still maintains the same good physical properties of zeolite FAU, therefore it is the same amount of zeolite for CO_2 adsorption in the laboratory. In addition, the results have been compared to the previous carbon footprint of conventional zeolite synthesis [5]. The system boundaries of this research were evaluated on the impacts of each condition of zeolite FAU synthesis compared to conventional methods from the raw material extraction, zeolite FAU synthesis, distribution, and disposal. The consumption of zeolite FAU was not considered.

Input and output were mainly collected from laboratory data. It was assumed that there is no emission at the distribution stage because of usage only in the laboratory. The electrical equipment used in the process consumed constant power throughout the lifetime of the device. At the disposal stage, waste was assumed to be assigned to landfills.

3.2 Life cycle inventory analysis

The different precursors or heat management options of zeolite FAU synthesis were based on the experimental data measured in the laboratory. All materials were transported to the laboratory area. The collection of raw data can be obtained by direct (activity level data or raw data in experiments) and indirect (emissions factor from literature and software). The carbon footprint of product can be calculated using the following Eq. (1):

$$Carbon\ Footprint = Activity\ level\ data\ x\ Emission\ factor \qquad (1)$$

In this analysis, the following processes including of silica extraction, zeolite FAU synthesis and disposal were considered as shown in Figure 1. Zeolite FAU was synthesized according to the research of Worathanakul and Rakpasert [6] with different crystallization temperatures and times. Initially, the Na_2SiO_3 and $NaAlO_2$ solutions were prepared. Na_2SiO_3 and $NaAlO_2$ solutions were then mixed and aged. Then, the solutions were heated in the crystallization step. The obtained gel was filtered and washed with distilled water. Then, the final product of zeolite FAU was dried. All the waste materials were assumed to be transported to a disposal site. Rice husk ash, filter paper and pH paper were landfilled.

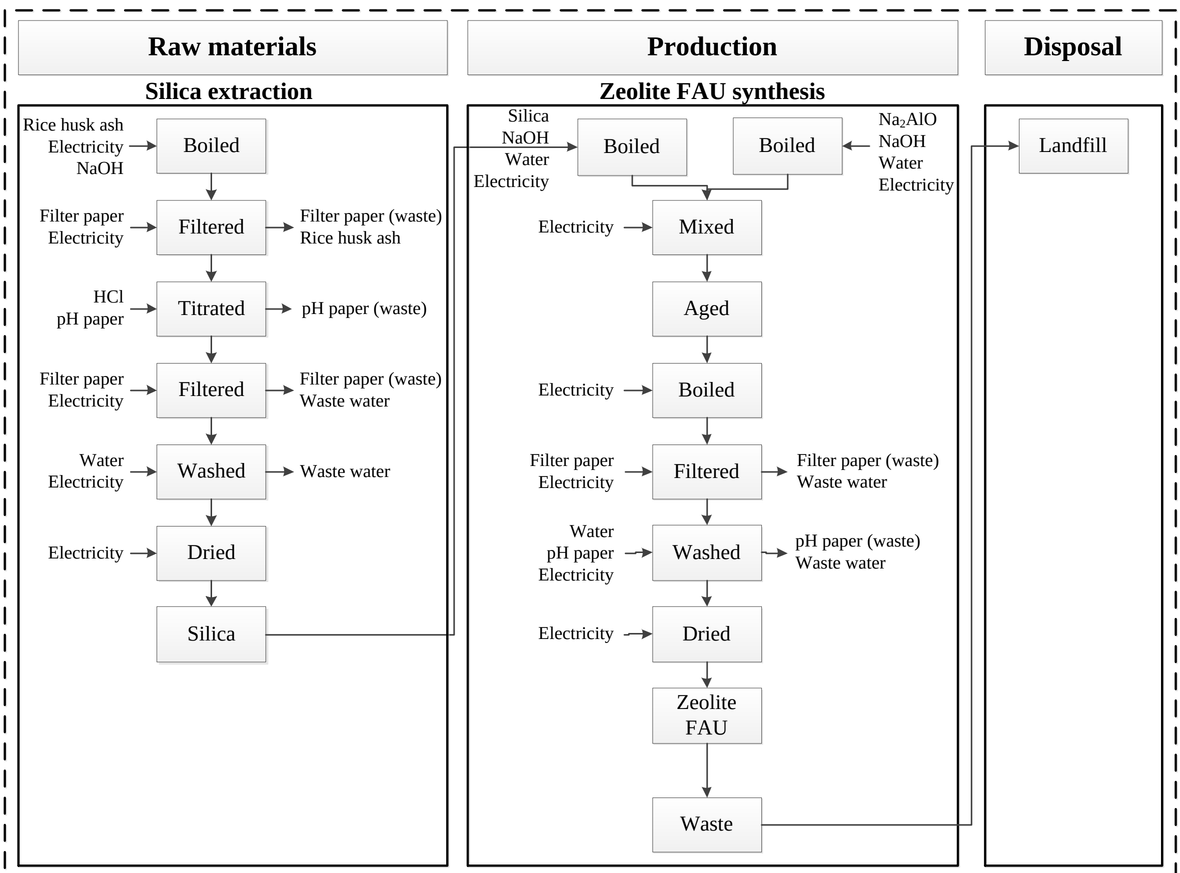

Figure 1 Inventory analysis in each procedure of the zeolite FAU synthesis

3.3 Impact assessment

The database concerning the embodied environment aspects of materials, transport use and other processes in this study is mostly used the emission factor of product carbon footprint based on the Thailand Greenhouse Gas Management Organization (TGO) [17] and according to the Intergovernmental Panel on Climate Change (IPCC) version 1.03 [18]. The results of the impact analysis were converted to the amount of GHG emissions for zeolite FAU synthesis with kgCO_2eq unit.

3.4 Interpretation

The interpretation was used to analyze carbon footprint based on conventional and green FAU synthesis accounting results and draw conclusions, allowing to identify environmental concerns and the hot spot with the highest emissions to provide project recommendations. This step can help to develop environmentally friendly products for finding the best environmental practices as well as improving the appropriate processes in the future.

Results and Discussion

1. Physical properties

Figure 2 (a) showed the effect of precursors (chemical and rice husk ash), crystallization temperatures (25 °C and 70 °C) and time (1 h and 3 h) on zeolite FAU synthesis. The main peaks of all samples were observed in line to the standard peak of zeolite FAU. It was clearly shown that the complete formation of pure zeolite FAU was successfully synthesized by rice husk ash as the silica source.

Figure 2 (b) and (c) showed the SEM images of zeolite FAU crystallized with different precursors, crystallization temperatures and time. At low temperature and short time (25 °C and 1 h) showed spherical crystal shaped and uniform distribution like from a chemical source.

In addition, the BET analysis of zeolite FAU synthesized from chemical at 70 °C and 1 h showed that BET surface area, micropore volume and total pore volume were about 15.56 m^2/g, 0.001 cm^3/g and 0.032 cm^3/g, respectively. While those BET analysis results of zeolite FAU synthesized from rice husk at 25 °C and 1 h were 18.27 m^2/g, 0.001 cm^3/g and 0.059 cm^3/g, respectively. It can be concluded that the precursors, temperature and time of crystallization were related to the crystal quality of zeolite FAU corresponds to the research of Rakpasert [19]. Furthermore, the temperature and time used in our zeolite FAU synthesis were lower than in the previous study, but still the zeolite FAU remained performed well.

2. Carbon footprint of zeolite FAU synthesis

The zeolite FAU synthesis under different precursors and heat management options was focused on the carbon footprint assessment. The carbon footprint value of zeolite FAU synthesized from rice husk ash at 70 °C and 3 h crystallization was calculated as shown in Table 1 to Table 2. The results showed that the amount of carbon footprint values from the silica extraction from rice husk ash step was 0.10 kg CO_2-eq. The amount of carbon footprint values from the zeolite FAU synthesis and disposal step was calculated as 0.37 and 0.01 kg CO_2-eq, respectively.

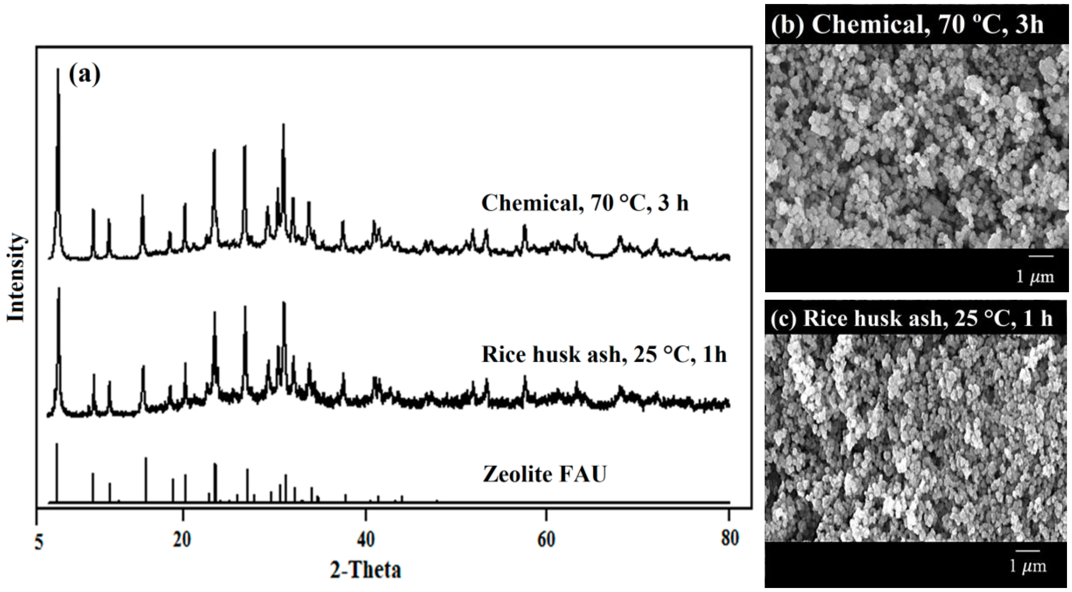

Figure 2 XRD patterns (a) and SEM images (b) of zeolite FAU synthesized with different precursors, crystallization temperatures and time

Table 1 Carbon footprint in the silica extraction from rice husk ash for zeolite FAU synthesis

Step	Amount	Unit	Emission Factor (kg CO_2eq/unit)	Carbon footprint (kg CO_2eq)
Rice husk ash	6.70×10^{-4}	kg	7.22	4.84×10^{-3}
pH meter paper	5.51×10^{-4}	kg	4.9×10^{-5}	2.70×10^{-9}
NaOH	8.43×10^{-3}	kg	1.04	8.75×10^{-3}
HCl	7.08×10^{-2}	kg	1.12	7.94×10^{-2}
Filter paper	2.64×10^{-3}	kg	0.74	1.94×10^{-3}
DI water	4.00×10^{-5}	m^3	2.58×10^{-5}	1.03×10^{-9}
Electricity of oven	3.37×10^{-4}	kWh	0.58	1.96×10^{-4}
Electricity of magnetic stirrer	8.43×10^{-5}	kWh	0.58	4.91×10^{-5}
Electricity of pump	1.05×10^{-5}	kWh	0.58	6.11×10^{-6}
Total carbon footprint in silica extraction step				0.10

Table 2 Carbon footprint in the zeolite FAU synthesis from rice husk ash

Step	Amount	Unit	Emission Factor ($kgCO_2eq$/unit)	Carbon footprint (kg CO_2eq)
Silica	$5.91x10^{-4}$	kg	$9.52x10^{-2}$	$5.63x10^{-5}$
pH meter paper	$8.24x10^{-1}$	kg	$4.90x10^{-5}$	$4.04x10^{-5}$
NaOH	$5.61x10^{-4}$	kg	1.04	$5.82x10^{-4}$
$NaAlO_2$	$1.58x10^{-4}$	kg	1.12	$1.77x10^{-4}$
Filter paper	$1.32x10^{-4}$	kg	0.74	$9.70x10^{-5}$
DI water	$4.07x10^{-5}$	m^3	$2.58x10^{-5}$	$1.05x10^{-4}$
Electricity of oven	$1.55x10^{-1}$	kWh	0.58	$9.00x10^{-2}$
Electricity of magnetic stirrer	$4.64x10^{-1}$	kWh	0.58	$2.70x10^{-1}$
Electricity of pump	$1.72x10^{-2}$	kWh	0.58	$1.00x10^{-2}$
Total carbon footprint in zeolites synthesis step				0.37
Waste	$3.10x10^{-3}$	kg	2.93	$9.08x10^{-3}$
Wastewater	4.070	kg	$1.20x10^{-3}$	$4.88x10^{-3}$
Total carbon footprint in disposal step				0.01

The highest carbon footprint of the conventional zeolite FAU synthesis was attributed in the production stage, accounting for 78% of the total carbon footprint. This is due to the chemical source input to zeolite FAU synthesis, including high electricity consumption to the oven under high temperature and long time for heat the crystallization (Table 3). Using rice husk ash instead of chemical as a silica source for zeolite FAU synthesis, the carbon footprint was reduced to 0.48 $kgCO_2eq$. Furthermore, several heat management options were considered through changes in the crystallization temperature and time. From Figure 3, the optimum condition for zeolite FAU synthesized was found at 25 °C and 1 h crystallization with 0.25 $kgCO_2eq$ because of low energy consumption. The carbon footprint for successfully synthesized zeolite FAU from rice husk ash at room temperature and short time (25 °C and 1 h) crystallization could reduce 48% carbon footprint compared to conventional methods. Therefore, the use of rice husk ash replaced for chemical precursors in zeolite FAU synthesis could help energy savings and emissions reductions. In addition, heat management options could significantly improve electricity consumption which is the key factor for the zeolite synthesis development.

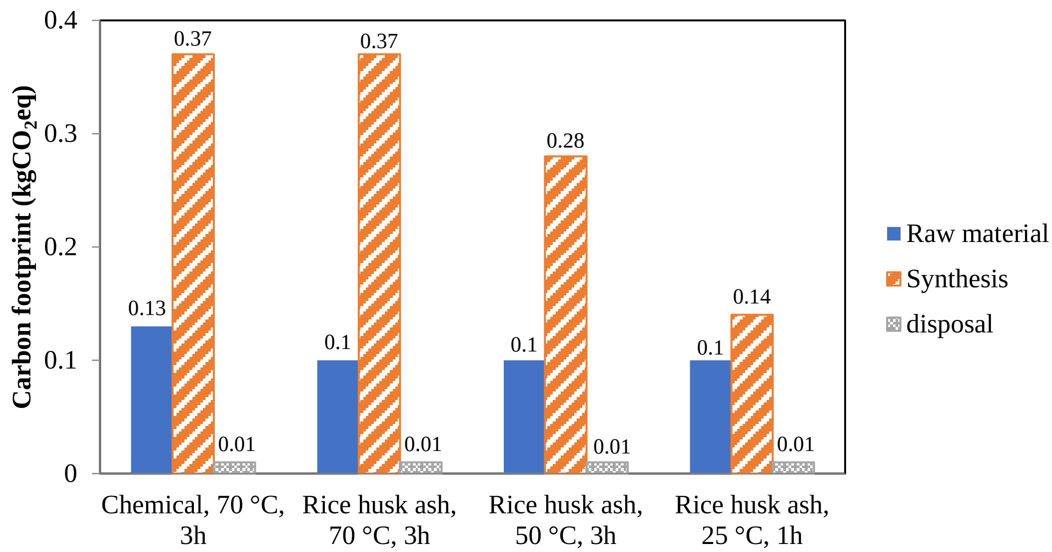

Figure 3 Carbon footprint in each stage for zeolite FAU synthesis

Table 3 Total carbon footprint and carbon dioxide adsorption capacity of zeolite FAU

Source	Crystallization		Total	Electricity of zeolite FAU synthesis step	CO_2 adsorption capacity
	Temperature	Time	kg CO_2eq	kW	mmol/g
Chemical	70 °C	3 h	0.52	0.64	4.56
Rice husk ash	70 °C	3 h	0.48	0.64	4.28
Rice husk ash	50 °C	3 h	0.39	0.55	5.17
Rice husk ash	25 °C	1 h	0.25	0.27	6.51

Conclusions

Silica source from rice husk ash was successfully extracted to synthesize green zeolite FAU. The utilization of rice husk ash as a silica source instead of a chemical source for zeolite FAU synthesis could reduce the carbon footprint compared to conventional methods. The lowest carbon footprint of zeolite FAU was derived from rice husk ash at room temperature and 1 h crystallization. These conditions represented a feasible option to reduce carbon footprint value and costs for zeolite FAU synthesis without affecting the physical properties of zeolite FAU. The great obtained results could be used to be an appropriate guideline for zeolite synthesis technologies and green precursors for further CO_2 adsorption.

Acknowledgement

This research was financially supported by the Thailand Research Fund (TRF), contract no. PHD59I0018 and U-Thong biomass Company Limited. We also acknowledge the Graduate school of King Mongkut's University of Technology North Bangkok for Patchaya Tobarameekul.

References

[1] Lee, S.Y. and Park, S.J. 2015. A review on solid adsorbents for carbon dioxide capture. Journal of Industrial and Engineering Chemistry. 23: 1-11.

[2] Chen, Y., Wang, B., Zhao, L., Dutta, P. and Winston Ho, W.S. 2015. New Pebax®/zeolite Y composite membranes for CO_2 capture from flue gas. Journal of Membrane Science. 495: 415-423.

[3] Kodasma, R., Fermoso, J. and Sanna, A. 2019. Li-LSX-zeolite evaluation for post-combustion CO_2 capture. Chemical Engineering Journal. 358: 1351-1362.

[4] Tobarameekul, P., Sangsuradet, S., Na Chat, N. and Worathanakul, P. 2020. Enhancement of CO_2 Adsorption containing zinc-ion-exchanged zeolite NaA synthesized from rice husk ash [Online]. Available at: http://ojs.kmutnb.ac.th/index.php/ijst/article/view/3640 [Accessed on 29 July 2021].

[5] Na chat, N., Maksiri, R., Vichaikham, P. and Worathanakul, P. Carbon footprint of zeolite A and zeolite Y derived from bagasse ash for CO_2 Adsorption. At the 7th International Conference on Green and Sustainable Innovation, Bangkok, Thailand on October 17-19, 2018.

[6] Worathanakul, P. and Rakpasert, N. 2016. Influence of different preparation methods of copper loading on Na-Y zeolite for green gas emission. International Journal of Environmental Science and Development. 7: 885-888.

[7] Kongnoo, A., Tontisirin, S., Worathanakul, P. and Phalakornkule, C. 2017. Surface characteristics and CO_2 adsorption capacities of acid-activated zeolite 13X prepared from palm oil mill fly ash. Fuel. 193: 385-394.

[8] Worathanakul, P. and Saisuwansiri, P. 2017. Effect of copper and iron loading on zeolite Y for carbon dioxide adsorption. The Journal of King Mongkut's University of Technology North Bangkok. 28: 373-381.

[9] Mohammed Ali, M.M., Ahmed, M.J. and Hameed, B.H. 2018. NaY zeolite from wheat (*Triticum aestivum* L.) straw ash used for the adsorption of tetracycline. Journal of Cleaner Production. 172: 602-608.

[10] Prasara-A, J. and Gheewala, S.H. 2017. Sustainable utilization of rice husk ash from power plants: A review. Journal of Cleaner Production. 167: 1020-1028.

[11] Belviso, C. 2018. State-of-the-art applications of fly ash from coal and biomass: A focus on zeolite synthesis processes and issues. Progress in Energy and Combustion Science. 65: 109-135.

[12] Gui, F., Ren, S., Zhao, Y., Zhou, J., Xie, Z., Xu, C. and Zhu, F. 2019. Activity-based allocation and optimization for carbon footprint and cost in product lifecycle. Journal of Cleaner Production. 236: 117627.

[13] Jradi, S., Chameeva, T.B., Delhomme, B. and Jaegler, A. 2018. Tracking carbon footprint in French vineyards: A DEA performance assessment. Journal of Cleaner Production. 192: 43-54.

[14] Ledgard, S.F., Falconer, S.J., Abercrombie, R., Philip, G. and Hill, J.P. 2020. Temporal, spatial, and management variability in the carbon footprint of New Zealand milk. Journal of Dairy Science. 103: 1031-1046.

[15] Percic, M., Ancic, I. and Vladimir, N. 2020. Life-cycle cost assessments of different power system configurations to reduce the carbon footprint in the Croatian short-sea shipping sector. Renewable and Sustainable Energy Reviews. 131: 110028.

[16] Buslaev, G., Morenov, V., Konyaev, Y. and Kraslawski, A. 2021. Reduction of carbon footprint of the production and field transport of high-viscosity oils in the Arctic region. Chemical Engineering & Processing: Process Intensification. 159: 108189.

[17] Thailand Greenhouse Gas Management Organization (TGO). 2020. Emission factor of product carbon footprint [Online]. Available at: http://thaicarbonlabel.tgo.or.th/admin/uploadfiles/emission/ts_117a1351b6.pdf [Accessed on 29 July 2021].

[18] IPCC. 2013. Climate Change 2013: The Physical Science Basis. Cambridge University Press, Cambridge, United Kingdom.

[19] Rakpasert, N. Preparation of different metals loading on FAU zeolite for NO reduction. Masters Degree, King Mongkut's University of Technology North Bangkok, Bangkok, Thailand, 2012.