International Journal of Applied Pharmaceutics

ISSN- 0975-7058 Vol 13, Special Issue 2, 2021

Original Article

ETHANOL RECOVERY FROM PROPOLIS PRODUCTION WASTE USING ADSORPTION DISTILLATION METHOD

MUHAMAD SAHLAN[1,2*], LINDATIANA YULISTIONO[3], ASHADI SASONGKO[3], APRILIANA CAHYA KHAYRANI[1], KENNY LISCHER[1,2], DIAH KARTIKA PRATAMI[4]

[1]Department of Chemical Engineering, Faculty of Engineering, Universitas Indonesia, 16424, Depok, West Java, Indonesia, [2]Research Center for Biomedical Engineering, Faculty of Engineering, Universitas Indonesia, 16424, Depok, West Java, Indonesia, [3]Department of Chemical Engineering, Institut Teknologi Kalimantan, 76127, Balikpapan, East Kalimantan, Indonesia, [4]Lab of Pharmacognosy and Phytochemistry, Faculty of Pharmacy, Pancasila University, 12640, Jakarta, Indonesia
*Email: sahlan@che.ui.ac.id

Received: 27 Aug 2020, Revised and Accepted: 09 Oct 2020

ABSTRACT

Objective: This study objective to compare two adsorbents in the ability to recover ethanol in the propolis production by-product.

Methods: Research methods include initial identification of the compound content in ethanol by-products of propolis production with GC/MS and determination of ethanol levels by GC-FID. The process of increasing ethanol content is carried out by the adsorption distillation method; the adsorbent used are synthetic zeolite 3A and white silica gel which has activated with the furnace.

Results: The result obtained from the study is the initial ethanol content 27.5%. After the ethanol recycle process using synthetic zeolite 3A 30, 60, and 90 grams, it was obtained content 82.3%, 87.1%, and 90.9%. By using 30, 60, and 90 grams silica gel, it was obtained content 72.0%, 79.2%, and 82.2%.

Conclusion: Based on these results it can be concluded that the ethanol content of the by-products of propolis production has been successfully increased by the distillation adsorption method with synthetic zeolite 3A 90 grams as the best result. The ethanol which recycling with this formulation has the characteristic odor of ethanol, which is very strong and does not has the characteristic odor of propolis.

Keywords: Adsorption distillation, Ethanol, Silica gel, and zeolite

DOI: http://dx.doi.org/10.22159/ijap.2021.v13s2.03. Journal homepage: https://innovareacademics.in/journals/index.php/ijap

INTRODUCTION

Propolis is a primary product produced by bees, besides honey, which is widely used in the health sector [1]. Propolis is a mixture of resin and bee saliva collected by honeybees from plant buds, leaves, and stems with various physical properties [2]. It acts as the primary protection for bees nests [3]. The chemical composition of propolis depends on the geography and plant-dependent variables [4]. Previous research has discovered the existence of various components contained in propolis, such as phenolic acid, caffeic acid phenethyl ester (CAPE), flavonoids, terpenoids, fatty acids, steroids, aromatic aldehydes, and alcohols [5–10]. Indonesian propolis generally has a number of pharmacological properties as the antioxidant, anti-inflammatory, antibacterial, anticancer, anti-angiogenic, xanthine oxidase inhibitory and antifungal [7, 10–13]. Propolis is found in the market with many different forms such as capsule, tablets, liquid drop, and mouthwash, toothpaste, mixing with honey, face creams, and ointments. The medical applications of propolis led to an increased interest in its chemical composition as well as its origin [14, 15].

Propolis can be obtained by extracting raw propolis. The solvent used for the raw propolis extraction process is ethanol, while the extract is called ethanol propolis extract (EEP) [16]. The Propolis production process has developed using the bubbling vacuum evaporator method as a substitute for a rotary vacuum evaporator because in the ethanol regeneration process with a rotary vacuum evaporator unit, ethanol cannot be regenerated optimally because water and other volatile compounds are evaporated with ethanol. Besides, the small evaporator capacity and the type of evaporator that can only be used for the distillation process, resulting in the inability of the rotary vacuum evaporator unit to regenerate the ethanol by-product of propolis production. By using the bubbling vacuum evaporator method, it can provide more ethanol regeneration results than the rotary vacuum evaporator method. The bubbling vacuum evaporator method has proven can regenerate ethanol up to 5 liters from the initial use of ethanol in the extraction process of 10 liters [17]. Seeing these conditions, the greater the propolis capacity produced is directly proportional to a large amount of regenerated ethanol.

Ethanol produced from the regeneration process has a distinctive odor of propolis and does not has the distinctive odor of ethanol, then the value of ethanol content and composition are not known, it is necessary to carry out the initial testing and recycling process so ethanol can be reused as a raw material for hand sanitizer, soap, or other product. The ethanol recycle process can be done by extractive distillation, pressure swing distillation, membrane technology, and adsorption distillation, but the adsorption distillation method is the most effective in the ethanol recycle process [18–21]. Adsorption distillation is a separation method in which process distillation and adsorption are carried out simultaneously. The ethanol that wants to be recycled is heated with a heating mantle; then the steam will pass through the adsorbent in the adsorption column. The adsorbent will adsorb both volatile and non-volatile impurity components that have a smaller size than the adsorbent pore, so the ethanol purity content will increase, then the ethanol vapor enters the distillation column and changes the phase to liquid [22]. The adsorbents used are called desiccants. These include molecular sieves, chloride salts, silica gel, and biobased desiccants. Type molecular sieves are the most used in the application of ethanol-water azeotrope separation [23]. In addition, the adsorption distillation process with silica gel has also been carried out and gives a good value [24]. The aims of this study to compare two adsorbents in the ability to remove ethanol impurities in the propolis production by-product.

MATERIALS AND METHODS

Materials

Ethanol to be recycled is obtained from the by-product of the production of propolis in the vacuum evaporator bubbling unit from PT. RIN Biotek Indonesia, Tangerang, Indonesia. Adsorbent synthetic zeolite used has a specification pore size 3A; the synthetic zeolite is directly imported from Guangdong Xintao Technology Co., Ltd, Guangzhou, China. Adsorbent white silica gel used has a size specification of 2-5 mm; the white silica gel is directly imported PT. Damases Sejahtera from Klaten, Indonesia.

Determination of ethanol characteristics

Determination of ethanol characteristics was carried out at the beginning of the process, the content of compounds in the ethanol by-products of propolis production using the GC/MS instrument, the ethanol content by-products of propolis production using the GC-FID instrument. The instruments of GC/MS and GC-FID are manufactured by Perkin Elmer, Waltham, USA. Ethanol was analyzed for its compound content using GC/MS Clarus 680 instrument. 1 μl sample was injected into the GC/MS instrument, which was operated using a column with dimensions of 30 m long and 250 μm in diameter with a helium carrier gas and elite wax as a stationary phase. The furnace temperature is programmed between 50-350 °C with a temperature rise rate of 10 °C/min. Ethanol was analyzed for its content using the GC-FID Clarus 680 instrument. 1 μL sample was injected into the GC-FID instrument, which was operated using a column with dimensions of 30 m long and 250 μm in diameter with hydrogen-carrier gas flowing at a flow rate of 45 ml/min and elite 1 as the stationary phase. The furnace temperature is programmed between 180-220 °C with a temperature rise rate of 50 °C/min [25, 26].

Adsorbent activation

In this study, the adsorbents used were synthetic zeolite 3A and white silica gel. Activation is carried out on the adsorbent using physical methods by heating it in a furnace. The heat generated by the furnace will evaporate water molecules and other impurities in the adsorbent, so the adsorbent absorption increases. The physical activation process aims to maintain the structure of the adsorbent. Synthetic zeolite was heated at 300 °C for 3 h, white silica gel was heated at 150 °C for 40 min [27].

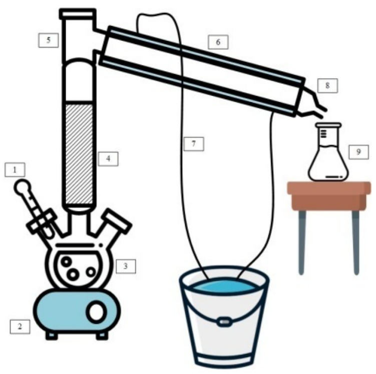

Fig. 1: Adsorption distillation process

Equipment

1. Thermometer
2. Heating Mantle
3. Three-Neck Flask
4. Adsorption column
5. Three-Way Adapter
6. Condenser
7. Hose
8. Distillation Adapter Along
9. Erlenmeyer

Adsorption distillation with adsorbent

Before carrying out the adsorption distillation process, chain the tools as presented in fig. 1. Put the adsorbent that has been weighed and activated according to the variables in the column of stuffing material. Pour 150 ml of the by-product of propolis into a three-neck flask and heat the ethanol using a heating mantle to a temperature of 87 °C. Furthermore, the ethanol that has evaporated will enter the adsorption column which has been filled with adsorbent according to the variables.

The adsorbent will absorb impurity components, both volatile and non-volatile components, which have a smaller size than the adsorbent pore; furthermore, the ethanol vapor which passes through the adsorbent pile will enter the distillation column and change the phase to liquid [22]. Pure ethanol from the adsorption distillation process will be accommodated in the Erlenmeyer. Carry out the adsorption distillation process until no ethanol is dripping. Furthermore, analysis of the final characteristics of ethanol includes parameters of compound content and ethanol content values using GC/MS and GC-FID instruments.

RESULTS AND DISCUSSION

Ethanol recovery from propolis production waste characteristic

The value of ethanol content and composition of ethanol produced from the regeneration propolis production never known before. Ethanol from the by-product of propolis production obtained from the bubbling unit vacuum evaporator was analyzed for ethanol content quantitatively with the GC-FID instrument, the percentage of ethanol content produced can be seen in table 1. The solute content was analyzed by GC/MS instrument. Based on this test, 50 components were detected as dissolved components in ethanol from the by-products of propolis production, but the main components that dominate as a by-product of propolis ethanol production presented in table 1.

Table 1: Results of initial ethanol component analysis

Retention time	% Relative area	Component	Concentration ethanol
0.96	6.21	1,2–propanediol	27.5 %
1.51–1.59	21.84	Acetaldehyde	
1.61	3.29	Methylamine	
1.69	58.24	Ethanol	
2.33	0.54	Ethyl acetate	
8.38	1.49	1,3,3-trimethyltricyclo[2.2.1.0(2,6)] heptane	
9.96	0.76	1-methyl-2-(1-methyl ethyl) benzene	
10.06	0.92	D-limonene	
16.94	0.83	Copaene	

Note: Retention time, % relative, component by GC/MS. Concentration ethanol by GC-FID

Based on the results of the GC-FID, the percentage of ethanol content was very low. In the propolis production process, ethanol used for the propolis extraction process has an ethanol content of 96%. In the propolis production process, water was added to the ethanol extract of propolis 96% until the ethanol concentration became 70%, which aims to form propolis wax deposits. Also, the process of adding sodium carbonate solution was carried out to increase the pH of the ethanol extract of propolis. In the propolis production process, glycerin is added as a solvent in the distillation process, which is carried out using a bubbling vacuum evaporator which aims to regenerate ethanol from the production of propolis [17]. Based on the odor sensory test, ethanol from the by-product of propolis production does not have the distinctive odor of ethanol but has a very strong and pungent distinctive odor of propolis.

Based on the results of the GC/MS, with the characteristics of ethanol that is easily oxidized, it is suspected that an oxidation process occurs in ethanol to acetaldehyde compounds in the ethanol by-product of propolis production. Furthermore, it is suspected that acetaldehyde undergoes an oxidation process to become an acetic acid compound and then an esterification reaction occurs between ethanol and acetic acid so that the formation of ethyl acetate compounds. The acetic acid compound formed is also thought to have reacted with ethanol to form 1,2–propanediol compounds. Besides, it is also suspected that there was a reaction to the breakdown of amino acid arginine from propolis dissolved in ethanol to form a methylamine compound. D-limonene compounds, copaene compounds, and 1,3,3-trimethyltricyclo compounds [2.2.1.0(2,6)] heptane are compounds found in plants; these compounds are thought to have been carried by bees when looking for food from plants and fruits so that the compound this is contained in the propolis content and dissolved in the ethanol by-product of propolis production. The compound 1-methyl-2-(1-methyl ethyl) benzene is thought to be the result of the decomposition reaction of phenolic acids, which is a type of polyphenol that is widely contained in propolis [6, 28–30].

Analysis of recycled ethanol using synthetic zeolite 3A adsorbent

The process of increasing the ethanol content of by-products of propolis production is carried out by the adsorption distillation method using 3A synthetic zeolite, which has been activated. The analysis results are presented in table 2. Based on analysis with the GC-FID instrument, the percentage of ethanol content that has been recycled by the adsorption distillation process using synthetic zeolite 3A adsorbent has increased when compared to the percentage of initial ethanol content. In addition, this study is more better if compared with previous studies, where pure ethanol concentration increased from 92% to 99.5% [31]. The GC-FID analysis results are in line with the test results using the GC/MS instrument, where the impurities are successfully absorbed by the adsorbent so that the recycled ethanol constituent components are detected less when compared to the initial constituent components and cause an increase in the percentage of ethanol content. Based on the sensory odor test, the recycled ethanol with 3A 30 grams synthetic zeolite adsorbent has a strong characteristic odor of ethanol, but it still has a weak characteristic odor of propolis. Recycled ethanol with synthetic zeolite adsorbent 3A 60 grams has the characteristic odor of ethanol, which is very strong and does not have the characteristic odor of propolis anymore. Recycled ethanol with 90 grams of synthetic zeolite adsorbent 3A has the characteristic odor of ethanol, which is very strong and sting then does not have the characteristic odor of propolis anymore.

Table 2: Results of analysis of recycled ethanol using synthetic zeolite 3A adsorbent

Mass adsorbent	Retention time	% Relative area	Component	Concentration ethanol
30 Grams	1.50	6.60	Acetaldehyde	82.3%
	1.53–1.55	12.31	Methylamine	
	1.58–1.66	75.18	Ethanol	
	7.57	1.22	1-methyl-2-(1-methyl ethyl) benzene	
60 Grams	1.48	5.91	Acetaldehyde	87.1%
	1.54	8.26	Methylamine	
	1.56–1.67	80.84	Ethanol	
	7.55	1.15	1-methyl-2-(1-methyl ethyl) benzene	
90 Grams	1.47–1.50	6.35	Acetaldehyde	90.9%
	1.52–1.55	9.38	Methylamine	
	1.59	69.70	Ethanol	
	7.56	1.87	1-methyl-2-(1-methyl ethyl) benzene	

Note: Retention time, % relative, component by GC/MS. Concentration ethanol by GC-FID

Analysis of recycled ethanol using white silica gel adsorbent

The process of increasing the ethanol content of by-products of propolis production is carried out by the adsorption distillation method using white silica gel, which has been activated. The recycled ethanol was analyzed for ethanol content quantitatively with the GC-FID instrument and analyzed qualitatively with the GC/MS instrument. The analysis results are presented in table 3. Based on analysis with the GC-FID instrument, the percentage of ethanol content that has been recycled by the adsorption distillation process using white silica gel adsorbent has increased when compared to the percentage of initial ethanol content. In addition, this study is more better if compared with previous studies, where the pure ethanol concentration increased from 90.8% to 96.7% [24]. The GC-FID analysis results are in line with the test results using the GC/MS instrument, where the impurities are successfully absorbed by the adsorbent so that the recycled ethanol constituent components are detected less when compared to the initial constituent components and cause an increase in the percentage of ethanol content. When compared with 30 grams of white silica gel adsorbent, based on the GC/MS analysis, 60 grams and 90 grams of white silica gel adsorbent are better because they can absorb methylamine components previously detected in analysis with 30 grams of white silica gel adsorbent. Based on the sensory odor test, the recycled ethanol with 30 grams of white silica gel has a weak characteristic odor of ethanol and a strong propolis odor.

Recycled ethanol with 60 grams of white silica gel adsorbent has a characteristic odor of ethanol and still has a distinctive smell of propolis. Recycled ethanol with 90 grams of white silica gel adsorbent has a strong characteristic odor of ethanol and still has a distinctive odor of propolis even though it is weak.

Table 3: Results of analysis of recycled ethanol using white silica gel

Mass adsorbent	Retention time	% Relative area	Component	Concentration ethanol
30 Grams	1.00–1.49	6.22	Acetaldehyde	72.0%
	1.53	8.20	Methylamine	
	1.66	65.60	Ethanol	
	2.05	0.97	Ethyl acetate	
60 Grams	1.53	12.91	Acetaldehyde	79.2%
	1.59–1.66	75.49	Ethanol	
	2.05–2.08	0.65	Ethyl acetate	
	1.53	12.91	Acetaldehyde	
90 Grams	1.53	15.29	Acetaldehyde	82.2%
	1.60–1.66	64.87	Ethanol	
	2.05	0.66	Ethyl acetate	

Note: Retention time, % relative, component by GC/MS. Concentration ethanol by GC-FID

Effect of a mass addition of adsorbent on ethanol content

In the process of increasing the percentage of ethanol content, it can be influenced by the factor of increasing the mass of the adsorbent, the type of adsorbent used, and the adsorbent pore size. Based on the research that has been done, the effect of adding the mass of each adsorbent on ethanol content can be shown in fig. 2.

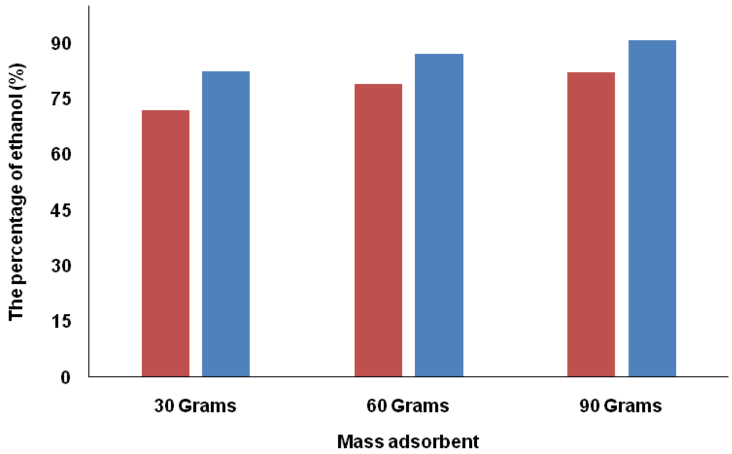

Fig. 2: Effect of mass addition of adsorbent on ethanol content orange is white silica gel and blue is synthetic zeolite 3A

It can be seen that synthetic zeolite adsorbent 3A is the best adsorbent when compared to white silica gel adsorbent. The greater the adsorbent mass used is directly proportional to the increase in the percentage of ethanol content obtained. This is because the amount of mass used affects the surface area of the resulting adsorbent. The surface area is an important factor in the adsorption process because the more the surface area of the adsorbent, the greater the resulting adsorption power so that the amount of adsorbate that will be absorbed also increases [32]. Synthetic zeolite 3A has an adsorption surface area of up to 1000 m^2/g [33]. In general, the pore size of white silica gel ranges from 3-4 Armstrong [22]. If seen in fig. 2, the mass condition of the adsorbent 30 grams of white silica gel, has a lower percentage of level than synthetic zeolite. This is because white silica gel has an adsorption surface area ranging from 400 to 800 m^2/g [34], which is lower when compared to synthetic zeolites. Besides the adsorbent surface area factor, the pore area also affects the selectivity level of an adsorbent. This causes different dissolved components in each recycled ethanol, which uses different adsorbents. The synthetic zeolite used in this study has 3 Armstrong specific pores. The general size of the ethanol molecule is 4,4 Armstrong. Ethanol, which has a large molecular size will be stuck and only pass between particles, while other compounds that have a smaller molecular size will be absorbed in the adsorbent. This is why synthetic zeolite adsorbent 3A is superior when compared to other adsorbents [23].

CONCLUSION

The ethanol content of the by-products of propolis production was successfully increased by the adsorptive distillation method. This is evidenced by the number of essential and non-essential compounds contained in the ethanol by-product of propolis production that has been successfully adsorbed so the percentage of ethanol content increases. 90 grams of synthetic zeolite adsorbent 3A is the best formulation because it can increase the recycled ethanol content by up to 90.9306%. Ethanol produced with this formulation has the characteristic odor of ethanol, which is very strong, sting, and does not have the characteristic odor of propolis.

ACKNOWLEDGMENT

The authors would like to thank the DRPM Universitas Indonesia for financial support through Grant Publikasi Terindeks Internasional (PUTI) Q3 2020 No: NKB-1403/UN2. RST/HKP.05.00/2020.

FUNDING

Funding from DRPM Universitas Indonesia

AUTHORS CONTRIBUTIONS

All the authors have contributed equally.

CONFLICT OF INTERESTS

The authors declare no conflict of interest.

REFERENCES

1. Bankova V. Chemical diversity of propolis and the problem of standardization. J Ethnopharmacol 2005;100:114–7.
2. Bankova V, Castro S, Marcucci M. Propolis recent advances in chemistry and plant origin. Apidologie 2000;31:3–15.
3. Simone M, Spivak M. Propolis and bee health: the natural history and significance of resin use by honey bees. Apidologie 2010;41:295–311.
4. Huang S, Zhang C, Wang K, Li G, Hu F. Recent advances in the chemical composition of propolis. Molecules 2014;19:19610–32.
5. Mahadewi A, Christina D, Hermansyah H, Wijanarko A, Farida S, Adawiyah R. Selection of discrimination marker from various propolis for mapping and identify anti-*Candida albicans* activity. AIP Conf Proc 2018;1933:020005.
6. Pratami DK, Munim A, Hermansyah H, Gozan M, Sahlan M. Microencapsulation optimization of propolis ethanolic extract from *Tetragonula Spp* using response surface methodology. Int J Appl Pharm 2020;12:197-206.
7. Miyata R, Sahlan M, Ishikawa Y, Hashimoto H, Honda S, Kumazawa S. Propolis components from Stingless bees collected on South Sulawesi, Indonesia, and their xanthine oxidase inhibitory activity. J Nat Prod 2019;82:205–10.
8. Sahlan M, Mandala D, Pratami D, Adawiyah R, Wijarnako A, Lischer K. Exploration of the antifungal potential of Indonesian propolis from Tetragonula biroi bee on *Candida sp* and *Cryptococcus neoformans*. Evergr J 2020;7:118–25.
9. Rahmi S, Sarwono A, Soekanto S. Effect of propolis honey candy consumption on the activity of lactoperoxidase in stimulated saliva. Int J Appl Pharm 2019;11:134–6.
10. Iqbal M, Fan T, Watson D, Alenezi S, Saleh K, Sahlan M. Preliminary studies: the potential anti-angiogenic activities of two Sulawesi Island (Indonesia) propolis and their chemical characterization. Heliyon 2019;5:e01978.
11. Farida S, Sahlan M, Rohmatin E, Adawiyah R. The beneficial effect of Indonesian propolis wax from Tetragonula sp. as a therapy in limited vaginal candidiasis patients. Saudi J Biol Sci 2020;27:142-6.
12. Pratami D, Mun A, Sundowo A, Sahlan M. Phytochemical profile and antioxidant activity of propolis ethanolic extract from Tetragonula bee. Pharmacogn J 2018;10:73–80.

13. Sahlan M, Devina A, Pratami D, Situmorang H, Farida S, Munim A. Anti-inflammatory activity of Tetragronula species from Indonesia. Saudi J Biol Sci 2018;26:1531–8.
14. Sulaiman G, Sammarrae K Al, Ad'hiah A, Zucchetti M, Frapolli R, Bello E. Chemical characterization of Iraqi propolis samples and assessing their antioxidant potentials. Food Chem Toxicol 2011;49:2415–21.
15. Sforcin J, Bankova V. Propolis: is there a potential for the development of new drugs? J Ethnopharmacol 2011;133:253–60.
16. Gonzalez M, Guzman B, Rudky R, Romano E, Molina M. Spectrophotometric determination of phenolic compounds in propolis argentine. Lat Am J Pharm 2003;22:243–6.
17. Sahlan M, Akhmariadi A, Pratama D, Hermansyah H, Wijanarko A. Scale-up production Indonesia liquid propolis from raw propolis and wild beehive using bubbling vacuum evaporator. MATEC Web Conf 2018;248:04001.
18. Tadayon F, Motiee F, Erfani A, Baghbani B. Design of adsorptive distillation for separation of an ethanol-water azeotropic mixture using bio-based adsorbents. Stud UBB Chem 2014;59:65–74.
19. Kumar S, Singh N, Prasad R. Anhydrous ethanol: a renewable. Renew Sustain Energy Rev 2010;14:1830–44.
20. Mujiburohman M, Sediawan W, Sulistyo H. A preliminary study: distillation of isopropanol-water mixture using fixed adsorptive distillation method. Sep Purif Technol 2006;48:85–92.
21. Chen W, Sheng C. Designing and constructing an optimization operating model for a bioetanol production system. Life Sci J 2013;10:2.
22. Silviana A. Pengambilan air dari sistem isopropil alkohol-air dengan adsorptive-distillation menggunakan zeolit alam dan gel silika. Reaktor 2008;12:29–32.
23. Al-Asheh S, Banat F, Al-Lagtah N. Separation of ethanol water mixture using molecular sieves and biobased adsorbents. Chem Eng Res Des 2004;82:855–64.
24. Megawati, Jannah RA, Rahayuningtiyas I. The influence of white and blue silica gels as adsorbents in adsorptive-distillation of ethanol-water mixture. AIP Conf Proc 2017;1788:30114
25. Dos Santos Costa CL, Ramos DP, da Silva JB. Multivariate optimization and validation of a procedure to direct determine acetonitrile and ethanol in radiopharmaceuticals by GC-FID. Microchem J 2019;147:654–9.
26. Del Mar Contreras M, Aparicio L, Arce L. Usefulness of GC-IMS for rapid quantitative analysis without sample treatment: Focus on ethanol, one of the potential classification markers of olive oils. LWT 2020;120:108897.
27. Nur H. Direct synthesis of NaA zeolite from rice husk and carbonaceous rice husk ash. Indones J Agric Sci 2001;1:40–5.
28. Abdulrazzaq H, Schwartz T. Catalytic conversion of ethanol to commodity and specialty chemicals. In: Basile A, Iulianelli A, Dalena F, Veziroglu TN. editors. Ethanol: Science and Engineering. 1st ed. Amsterdam: Elsevier BV; 2019. p. 3–24.
29. Sun J, Wang Y. Recent advances in catalytic conversion of ethanol to chemicals. ACS Catal 2014;4:1078–90.
30. Wagner F. Acetic acid. Kirk-Othmer Encycl Chem Technol; 2000. p. 1‑21.
31. Taufany F, Soewarno N, Yuwono K, Ardiyanta D, Eliana M, Raisa I, *et al*. Feed plate and feed adsorbent temperature optimisation of the distillationo-adsorption process to produce absolute ethanol. Can Cent Sci Ed 2015;9:140–7.
32. Zhang Z, Zhang W, Chen X, Xia Q, Li Z. Adsorption of CO_2 on zeolite 13X and activated carbon with the higher surface area. Sep Sci Technol 2010;45:710–9.
33. Stadie NP, Vajo JJ, Cumberland RW, Wilson AA, Ahn CC, Fultz B. Zeolite-templated carbon materials for high-pressure hydrogen storage. Langmuir 2012;28:10057–63.
34. Chua HT, Ng KC, Chakraborty A, Oo NM, Othman MA. Adsorption characteristics of silica gel+water systems. J Chem Eng Data 2002;47:1177–81.