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Extraction of Succinic Acid from Aqueous Mixtures by Emulsion Liquid Membrane Process using Amberlite LA-2 as a Carrier

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ABSTRACT

Background: Succinic acid is widely used in pharmaceutical, chemical and food industries with numerous applications. Despite the purification technology difficulties, the downstream processes raise important problems concerning the environmental protection. Thus, it is essential to develop an economical separation process which used less chemical consumption, in order to reduce environmental pollution and lowered the cost of purification. In this study, Emulsion liquid Membrane (ELM) process was used for succinic acid extraction using Amberlite LA-2 as a carrier. Na_2CO_3 was used as a stripping agent, kerosene as a diluent, and Span 80 as a surfactant. The Emulsion Liquid Membrane system was applied to get high selectivity of the succinic acid. Several parameters were investigated in the system such as carrier, stripping agent, surfactant concentration, and homogenizer speed. The concentration of extracted succinic acid and acetic acid was determined using High Performance Liquid Chromatography column Aminex HPX-247. Based on the result, the optimum conditions of succinic acid extraction selectivity are 0.05M Amberlite LA-2 in kerosene, 0.01M Na_2CO_3, 3% (w/v) of Span 80 and 7000 rpm of homogenizer speed.

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INTRODUCTION

Succinic acid is a dicarboxylic acid, as seen in figure 1, occur as white solid crystal at room temperature. The interest in succinic acid is justified by its biological activity, as this acid exhibits a powerful antiviral and antibacterial compound as well as good detoxifying supplement from alcohol overproduction. Succinic acid plays an important role in Krebs Cycle which helps detoxifying the human body, called as methylation. The excess of acetaldehyde produced by the human body can cause muscle destruction, lower degree liver heart, lack of testosterone hormone, and brain damage. Succinic acid, which is used in the medicine such as niacin (vitamin B3), has the ability to repair the damages in the body by allowing the body to release acetaldehyde more efficiently, makes the human being able to increase the immune system in the body.

Faced with escalating oil prices that are predictably to continue rising, the chemical industry is trying to find the alternative sources for a number of chemical ingredients derived from petroleum and other fossil sources. One such ingredient is succinic acid, which is currently made mainly from fossil-derived maleic anhydride (Zeikus *et al*., 1999). Succinic acid is industrially produced from butane, a liquefied petroleum gas. Due to the difficulties of the chemical synthesis, there is a high demand in producing succinic acid with low-cost fermentation process. The limited nature of fossil-reserves and increasing environmental concerns to replace the petroleum-based chemical processes with biobased processes. Thus, biosuccinate has given great intention worldwide due to its advantages in terms of environmental and less production cost needed.

Fermentation processes being one of the ideal methods to produce succinic acid, have been proven to be more environmental friendly and use renewable resources. The most current used strains are *Escherichia coli*, *Actinobacillus succinogenes*, *Anaerobiospirillum succiniciproducens*, and *Mannheimia succiniciproducens*

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(Song and Lee, 2006; Song *et al*., 2007; Wang *et al*., 2011). Meanwhile, *Corynebacterium* sp. and *Bacteroides fragilis* have been introduced recently as bio-succinate platform strains (Isar *et al*., 2006). The major problems of succinic acid fermentation are the low concentration of products and the high difficulties in separation process. This is due to the production of serious inhibitor in the fermentation broth which can inhibit the production of succinic acid. The classical method to separate succinic acid is by adding calcium hydroxide which formed calcium salt in order to liberate the acid. This method produced huge amount of waste salt because it consumes large amount of chemical such as sulphuric acid and lime (Inci and Aydin, 2003). Membrane filtrations including ultrafiltration, microfiltration, nanofiltration, and electrodialysis have been used for the separation and purification of succinic acid [Yao *et al*., 2008]. Reactive extraction method was also used to separate succinic acid from fermentation products mixtures or dilute aqueous stream. This method consumes more time to reach its equilibrium state to get a high succinic acid extraction.

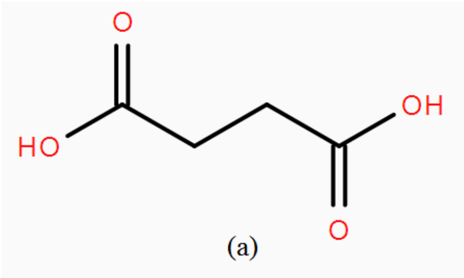

Fig. 1: Chemical structure of succinic acid.

In this research, Emulsion Liquid Membrane (ELM) system was used with several operating variables to separate succinic acid. The advantages of using Emulsion liquid membrane system are it uses less chemical and less energy consumption. The extraction and stripping process was occurred in one single step with high percentage of product extracted (Malik *et al*., 2012). Emulsion Liquid Membrane process has the potential to remove toxic substances to a very low level which applicable in the wastewater treatment process. The paper was focused on the extraction of succinic acid from the binary mixture of succinic-acetic acids solution at the feed phase using emulsion liquid membrane system. Several parameters were investigated such as carrier concentration, stripping agent concentration, and surfactant concentration. The amount of succinic acid and acetic acid extracted was analyzed using High Performance Liquid Chromatography.

Transport mechanism:

The transport mechanism for succinic acid extraction using secondary amines as a carrier from binary mixtures containing succinic-acetic acid solution;

(i) Succinic acid dissociates into H^+ and succinate ions (A^-)

$$HA = H^+ + A^-$$

(ii) Secondary amine react with succinic acid which formed amine salt

$$H^+ + A^- + R_2N = R_2NH^+ + A^-$$

(iii) Transport amine salt to the interface between the membrane and internal phase. The complex forms react with Na_2CO_3 at the membrane and stripping interface.

$$2R_2NH^+ + 2Na^+ + CO_3^{2-} \rightarrow (R_2NH)_2CO_3 + 2Na^+A^-$$

(iv) Finally, the amine carbonate form in the stripping reaction interface diffuse back to the external-internal interface

$$(R_2NH)_2CO_3 \rightarrow 2R_2N + CO_2 + H_2O$$

1. Methodology:

Chemical and Reagents:

The feed phase containing a mixture of succinic acid (purity: 90%) and acetic acid (purity: 99.7%) was purchased from Sigma-Aldrich. Lauryl trialkylmethylamine (Amberlite LA-2), sodium carbonate (Na_2CO_3, purity: 99.5%) were obtained from Merck company. Kerosene was obtained from Fluka. Sorbitan monooleate (Span 80) and sulphuric acid (H_2SO_4 , purity:95%) was obtained from Sigma-Aldrich. All the reagents were analytical grade and used without further purification.

Experimental:

Emulsion Liquid Membrane:

The mixtures solution of external phase was prepared by adding appropriate amount of succinic and acetic acid in deionized water. The concentration of succinic and acetic acid were 50 and 18.8 g/L, respectively.

Amberlite LA-2 was used as a carrier and Span 80 as a surfactant, were dissolved in kerosene as a diluent to form organic phase membrane. A stripping solution was prepared by dissolving sodium carbonate (Na_2CO_3) in deionized water. Water in oil (w/o) emulsion was made by slowly adding the stripping solution to the organic membrane phase with an equal volume of 5mL. The organic and aqueous phases were stirred for 5 minutes using homogenizer at 12000 rpm to obtain white emulsion. The emulsion must be prepared freshly for every experiment. Then, the prepared emulsion was then dispersed into the agitated vessel containing the external solution and stirred at 320 rpm for 5 minutes. After that, the samples are immediately transferred to a separation funnel. The aqueous phase will be filtered using puradisc nylon syringe filter 0.45 µm to remove any uncertainties for analysis purposes.

Analysis:

Succinic acid and acetic acid concentration were analyzed using High Performance Liquid Chromatography (HPLC, Model-Series 200, Perkin-Elmer), equipped with column Aminex Ion Exclusion HPX-87H (300 x 7.80 mm) with UV detector at 210 nm. The mobile phase used was 0.05M H_2SO_4. The flowrate and temperature for the sample was 0.5mL/min and 40°C, respectively. The typical retention time of succinic acid and acetic acid were 14.5 and 18.0 minutes, respectively.

RESULTS AND DISCUSSIONS

Emulsion Liquid Membrane:

Effect of Carrier Concentration:

The effect of Amberlite LA-2 concentration on the succinic acid extraction is shown in figure 2. Generally, amine extractant, such as Amberlite LA-2, is favorable in the organic acid extraction. Secondary amine was mostly used in the succinic acid extraction because of the higher extractability compared to tertiary amine (Hong *et al*., 2001). The result shows that the increasing of Amberlite LA-2 concentration from 0.005 to 0.05, the extraction of succinic acid increased. This is due to the high number of Amberlite LA-2 were formed complex with the succinic acid resulting in increasing diffusion of succinic acid from the external aqueous phase to the membrane phase. Lee (2011) stated that when increased the concentration of carrier, the degree of succinic acid extraction increasing as the concentration gradient of the succinic-amine complexes across the membrane enhances the flux of succinic acid through the membrane.

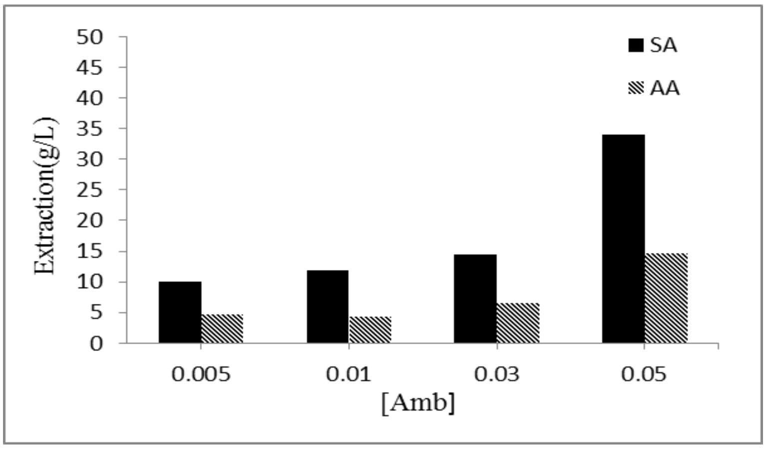

Fig. 2: Effect of concentration of Amberlite LA-2 in the membrane phase on degree of extraction of succinic acid.

Effect of Stripping Agent Concentration:

The effect of Na_2CO_3 concentration in the stripping solution on the extraction efficiency of succinic acid over the acetic acid was investigated in the range of 0.001 to 0.1M as shown in figure 3. The result shows the increment of the Na_2CO_3 concentration from 0.001 to 0.01M resulted in increase of succinic acid extraction. At low concentration of stripping agent, only a limited amount of succinate ions will be stripped because all stripping agent was completely accumulated. Meanwhile, at concentration of 0.01M Na_2CO_3 shows the highest succinic acid extraction. This is due to sufficient volume of stripping agent to strip the succinate-amine complexes into the internal phase. However, further increased the stripping concentration from 0.01 to 0.1M, the succinic acid extraction was decreased. The excess of Na_2CO_3 beyond its certain limits will lead to unstable of liquid membrane and the hydrolysis of the ester bonds of Span 80 as a part of the emulsifier in ELM process (Othman *et al*., 2013; Lin *et al*., 2002). This is resulting in decreased the succinic acid extraction efficiency.

Therefore, the high concentration of Na_2CO_3 in the stripping phase did not seem suitable for the succinic acid extraction.

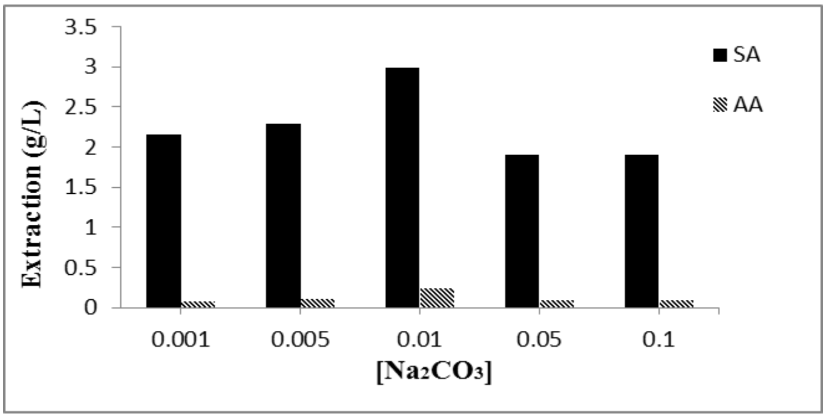

Fig. 3: Effect of concentration of Na_2CO_3 in the stripping phase on degree of extraction of succinic acid and acetic acid from a binary mixture in the ELM system.

Effect of Surfactant Concentration:

Figure 4 shows the effect of span 80 concentration in the membrane phase on separation of succinic acid from binary mixtures solution. The result shows that the extraction of succinic acid increase as increased the span 80 concentration from 1 to 3% of span 80. This is because of the decreased in interfacial tension, so smaller droplet size will formed. This will increased the mass transfer area resulted in increased of succinic acid extraction. This is in line with Sulaiman *et al*. (2014), which stated that by adding the surfactant concentration, the interfacial tension should be lowered. However, at 3 to 7% of span 80 the extraction of succinic acid is decreased. This is because of the excessive surfactant has the ability to increase the resistance at the interface and increase the viscosity of the organic phase. In addition, high surfactant levels can restrain the mass transfer of the solute by increasing the interfacial resistance. Moreover, it will interfere the carrier reaction at the interface causing in decreased of succinic acid extraction. Meanwhile, the succinic acid extraction increased at 10% of span 80. Even though the succinic acid extraction was high, it was not preferable because the emulsion formed was hard to break during recovery process. It shows an agreement with Ahmad *et al*. (2011) who stated that too stable emulsion is unfavorable because of the difficulty in demulsification process.

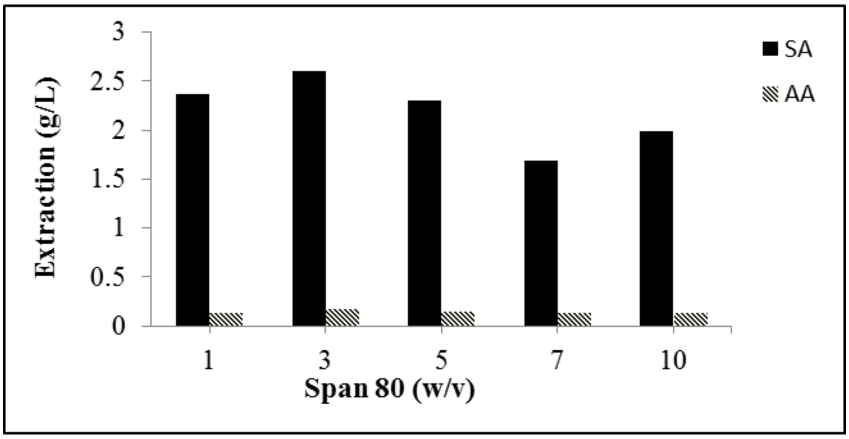

Fig. 4: Effect of concentration span 80 on degree of extraction of succinic acid and acetic acid from a binary mixture in the ELM system.

Effect of Homogenizer Speed:

Figure 5 illustrates the effect of different homogenizer speeds on the performance of succinic acid extraction from succinic-acetic mixtures solution. It was observed that an increment of homogenizer speed from 5000 to 7000 rpm, the concentration of extracted succinic acid was increased. This can be explained that an addition of homogenizer speeds provide higher dispersion rate of emulsion which increase the available number of internal droplets. This can leads to the larger membrane-internal interface thus promoting mass transfer of acid ions into the internal phase hence improving the extraction efficiency. The concentration of succinic acid extracted decreased at 7000 up to 13500 rpm. Although high number of smaller droplets was produced at higher speeds, this condition is unfavourable because these smaller internal droplets tend to coalesce among each other, thus enlarging their sizes which lead to the decrease in succinic acid extraction efficiency. This condition leads to the leakage of internal phase into the external phase hence decreasing the extraction of succinic acid. This was mentioned by Valenzuela *et al*. (2005) that an excess of homogenizer speed produced coalescence and breakdown the emulsion globules.

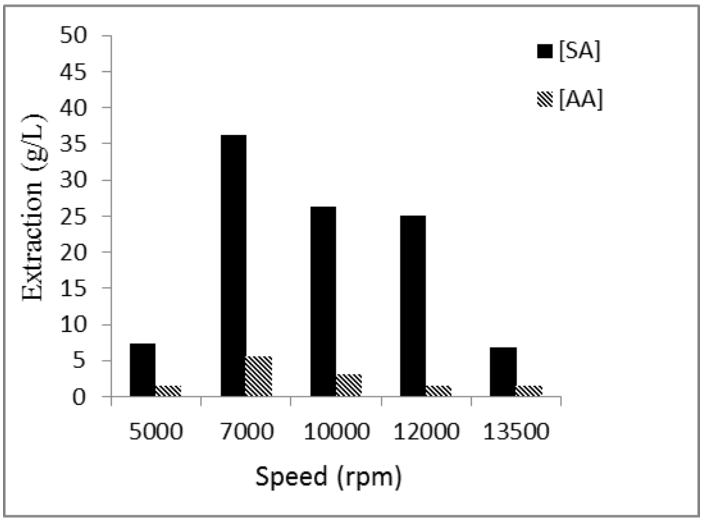

Fig. 5: Effect of homogenizer speed on degree of extraction of succinic acid and acetic acid from a binary mixture in the ELM system.

3. Conclusion:

Removal of succinic acid in the binary mixtures was carried out in batch ELM systems with a feed composition corresponding to two main organic acids in the actual fermentation broth in producing succinic acid. The effects of four operating variables were investigated to get a high degree of succinic acid extraction. The optimum experimental condition for carrier concentration, stripping agent concentration, surfactant concentration, and homogenizer speed were 0.05M, 0.01M, 3% (w/v), and 7000 rpm, respectively.

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REFERENCES

Ahmad, A.L., A. Kusumastuti, C.J.C. Derek and B.S. Ooi, 2011. Emulsion liquid membrane for heavy metal removal: An overview on emulsion stabilization and destabilization. Chemical Engineering Journal, 171(3): 870-882.

Hong, Y.K., W.H. Hong and D.H. Hong, 2001. Application of Reactive Extraction to recovery of Carboxylic Acids. Biotechnology and Bioprocess Engineering, 6: 386-394.

Inci, I. and A. Aydin, 2003. Extraction of Hydroxycarboxylic acids with MIBK/Toluene Solutions of Amines. Journal of Scientific and Industrial Research, 62(9): 926-930.

Isar, J., L. Agarwal, S. Saran and R.K. Saxena, 2006. Succinic acid production from *Bacteroides fragilis*: Process optimization and scale up in a bioreactor. Anaerobe, 12: 231-237.

Lee, S.C., 2011. Extraction of succinic acid from simulated media by emulsion liquid membranes. Journal of Membrane Science, 381: 237-243.

Lin, S.H., C.L. Pan and H.G. Leu, 2002. Equilibrium and mass transfer characterictics of 2-chlorophenol removal from aqueous solution by liquid membrane. Chemical Engineering Journal, 87: 163-169.

Malik, M.A., M.A. Hashim and F. Nabi, 2012. Extraction of metals ions by ELM separation technology. Journal of Dispersion Science and Technology, 33: 346-356.

Manzak, A. and M. Sonmezoglu, 2010. Extraction of acetic acid from aqueous solutions by emulsion type liquid membranes using Alamine 300 as a carrier. Indian Journal of Chemical Technology, 17: 441-445.

Othman, N., H. Mat and M. Goto, 2006. Separation of silver from photographic wastes by emulsion liquid membrane system. Journal of Membrane Science, 282(1-2): 171-177.

Othman, N., O.Z. Yi., S.N. Zailani, E.Z. Zulkifli and S. Subramaniam, 2013. Extraction of Rhodamine 6G dye from liquid waste solution: study on Emulsion Liquid Membrane stability performance and recovery. Separation Science and Technology, 48(8): 1177-1183.

Song, H. and S.Y. Lee, 2006. Production of succinic acid by bacterial fermentation. Enzyme and Microbiology Technology, 39: 352-361.

Song, H., Y.S. Huh, S.Y. Lee, W.H. Hong and Y.K. Hong, 2007. Recovery of succinic acid produced by fermentation of a metabolically engineered Mannheimia succinoproducens strain. Journal of Biotechnology, 132: 445-452.

Sulaiman, R.N.R., N. Othman and N.A.S. Amin, 2014. Emulsion Liquid Membrane stability in the extraction of ionized nanosilver from wash water. Journal of Industrial and Engineering Chemistry, 20: 3243-3250.

Valenzuela, F., C. Fonseca, C. Basualto, O. Correa, C. Tapia and J. Sapag, 2005. Removal of copper ions from a waste mine water by a liquid emulsion membrane method. Minerals Engineering, 18: 33-40.

Wang, D., Q.A. Li, M.H. Yang, Z.G. Su and J.M. Xing, 2011. Efficient production of succinic acid from corn stalk hydrolysates by a recombinant *Escherichia coli* with ptsG mutation. Process Biochemistry, 46: 365-371.

Yao, Z., H. Wu, S. Li, M. Jiang, 2008. Method for separation succinic acid from anaerobic fermentation broth. Chinese patent CN200610086003.7.

Yao, Z., H. Wu, H. Liu, S. Li and M. Jiang, 2008. Chinese Patent No. CN200610086003.7. CN Patent.

Zeikus, J.G., M.K. Jain and P. Elankovan, 1999. Biotechnology of succinic acid production and markets for derived industrial products. Journal of Applied Microbiology and Biotechnology, 51: 545-552.