CHEMICAL ENGINEERING TRANSACTIONS

VOL. 56, 2017

A publication of AIDIC

The Italian Association
of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Jiří Jaromír Klemeš, Peng Yen Liew, Wai Shin Ho, Jeng Shiun Lim

ISBN 978-88-95608-47-1; **ISSN** 2283-9216

DOI: 10.3303/CET1756008

Water Purification of Lake Water Using Progressive Freeze Concentration Method

Norshafika Yahya, Lee Wei Jie., Zaki Yamani Zakaria, Norzita Ngadi, Zurina. Mohamad, Roshanida Abdul Rahman, Mazura Jusoh*

Faculty of Chemical and Energy Engineering, Universiti Teknologi Malaysia (UTM), 81310 UTM Johor Baru, Johor, Malaysia
mazura@cheme.utm.my

Progressive freeze concentration method using stainless steel coil crystallizer was performed to concentrate and purify water from lake in Universiti Teknologi Malaysia, UTM. The study focused on the effect of operation time in range of 5 to 25 min and coolant temperature in range of -5 to -17 °C with constant pump rotation (450 rpm), initial biological oxygen demand (16.83 mg/L), total suspended solid (566 mg/mL) and turbidity (300 NTU). In water treatment using this progressive freeze concentration setup, a layer of pure ice crystal is produced on the inner wall of the crystallizer, leaving behind concentrated mother liquor. In order to determine the efficiency of the process, values of parameters such as biological oxygen demand (BOD_5), total suspended solids (TSS) and water turbidity were measured. High system efficiency was observed at coolant temperature of -11 °C and operation time of 10 min giving BOD: 2.23 and 2.47 mg/L TSS: 28 and 36 mg/L and turbidity 1.22 and 1.25 NTU, respectively. The current technology of crystallizer design was found to be relevant to reduce BOD_5, TSS and turbidity of water lake.

1. Introduction

Planet earth is also called 'the water planet' because it has full approximately 14,108 km^3 of water. However, 97.5 % of the water exists in the seas, meanwhile the remaining water is the water in lakes (0.007 %) and in river (0.002 %) (Ministry of the Environment, 2016). Lakes and river are the best 'available' freshwater sources on Earth and it is valued as water sources for fishing, water transport, recreation, tourism and also for drinking purpose (after water treatment). Fresh lake water is one of the most vital resources used by homes, industries and businesses. Lake water mostly contains a lot of unwanted substance which come from a various kinds of pollutants cause by human activities, nature (Mohd Yusof, 1996), development activities and geological conditions (Mohd Ekhwan and Large, 2004). Based on Interim National Water Quality Standard (INWQS) at several lake in Malaysia, there are reported the range of Water Quality Index (WQI) for Biological oxygen demand (BOD5) are 342.9 mg/L to 179.4 mg/L (Adeleke Abdul Rahman et al.,2014) , Total suspended solids (TSS) are 179.4 to 5.9 mg/L and turbidity are 54.56 to 12.41 NTU (Rahaman et al.,2016) compared with the standard of INWQS for BOD5 (1 mg/L, class I; 3 mg/L, class II; 6 mg/L, class III; 12 mg/L, class IV; >12 mg/L, class V), TSS (25 mg/L, class I; 50 mg/L, class IIA; 150 mg/L, class III; 300 mg/L, class IV; <300 mg/L, class V) and turbidity (5 NTU, class I; 50 NTU, class IIA; 150 NTU, class III; 300 NTU, class IV; <300 NTU, class V) (National Water Quality Standard, 2016). The standardization measurement from INWQS is an important and most helpful in assessing and monitoring surface water (Yisa and Jimoh, 2010).

Despite not having any serious insufficient drinking water issue, provision of alternative supply for drinking water would serve as a valuable facility for UTM students. In UTM, there is a huge lake for recreation and landscape purpose and it could play a role as an alternative source of providing daily drinking water. The lake water needs to be purified for drinking purpose. There are a few methods for water purification process, namely evaporation, reverse osmosis and freeze concentration. Evaporation is the simplest and the pioneering method of removing solvent through the vaporization of the liquid. A portion of liquid molecules possess sufficient energy to break the molecular bonds and vaporize into gas phase. Although external energy is not required throughout the whole process, it is still not likely a good way for water purification since it takes long time for the process to complete.

Please cite this article as: Yahya N., Lee W.J., Zakaria Z.Y., Ngadi N., Mohamad Z., Abdul Rahman R., Jusoh M., 2017, Water purification of lake water using progressive freeze concentration method, Chemical Engineering Transactions, 56, 43-48 DOI:10.3303/CET1756008

For boiling, the process is much faster than surface evaporation. Nevertheless, this method is not favorable for water purification process as it requires a lot of heat energy to vaporize the liquid which is known as heat of vaporization (Miyawaki et al., 2012). From some research analysis, it has been known that both boiling and surface evaporation have low yield if compared to other methods as some of the solutes are entrapped in vapor while vaporization process occurs. The leftover solution will be less concentrated while the vapor will be highly impure.

Reverse osmosis hence replaced evaporation with a higher yield and the least amount of energy as no phase change occurs. Reverse osmosis is a process where a semi-permeable membrane is needed to separate solvent from solution by the help of concentration gradient. The purity of solvent removed is very high but the cost of replacing the membrane is also high (Rodríguez et al., 2000). Freeze concentration method are chosen as an advanced technology which is suitable to separate many types of solvent from solution, besides it is capable of enhancing concentration of a solution. This method has potential to treat the water which is believed to have a lower capital cost (Jusoh et al., 2008) and maintenance fees compared to reverse osmosis and uses less energy compared to evaporation (Ab Hamid et al., 2015).

Freeze concentration also removes the water component out of the solution without any heating process compared with using evaporation and it is suitable for separation of some heat-sensitive products and more important this technology not used of any fuel that can be contribute to the low carbon issues. The process are simplicity due to the nature of the water which traps very less amount of impurities and can be considered as one of the best methods among all. The freeze concentration can be divided into two different types, namely progressive freeze concentration and suspension freeze concentration. Progressive freeze concentration is a method of separating solvent and solution into a single ice layer on the wall where coolant is applied, whereas suspension freeze concentration is a process in which lots of small ice crystals are formed as a suspension in the mother liquor. Progressive freeze concentration (PFC) was used in the study due to its simpler nature which makes it easier to be operated. There are a few factors which affect the efficiency of the system such as pump rotational speed, operation time, coolant temperature etc. In this study, the effects of coolant temperature and operation time towards progressive freeze concentration were investigated due to the most effected in PFC study (Shafira et al., 2016).

2. Materials and Methods

Lake water from Tasik Ilmu in Universiti Teknologi Malaysia, Johor was used as a raw material for this experiment. The water samples were collected from about 10 cm below the water surface at the same location of lake (Sujaul Islam et al., 2012) to ensure that all the water samples were in the same condition to provide more accurate result. The water sample from the lake was then preserved in sampling bottles and stored in the freezer at temperature of 2 °C before started the process. The initial BOD_5 (16.83 mg/L), TSS (556 mg/L) and turbidity (300 NTU) were determined using BODII Trak for BOD_5 (APHA,1992), filtering apparatus with suction tube for TSS (Gupta et al., 2009) and nephelometer for turbidity (Amrita, 2011). Stainless steel coil crystallizer with hollow structure was used, which could provide higher productivity and efficiency for progressive freeze concentration process through its higher surface area. The advantages of coil crystallizer in PFC process is its high productivity and the capability of increasing productivity (Jusoh et al., 2008) by adding more cycles to the whole process or increasing the coil diameter (Ab Hamid et al., 2015).

The coil crystallizer consists of three cycles which are attached by five flanges at the edge of the coil. With the flanges, the formation of ice can be easily observed at each point. The coil crystallizer can be split into two for ice removal process. In order to monitor the temperature profile of the entire process, eight thermocouples were installed on every point on each cycle of the crystallizer recorded by Picolog recorder. In this study, the source of energy which provides cooling effect came from ethylene glycol, which was the coolant. The coolant in the water bath was cooled to a desired temperature in the range of -5 °C to -17 °C. Data from Picolog recorder software was monitored constantly to ensure the temperature was maintained at the desired value. From the feeding tank, the lake water solution was fed while 2 min of time delay was required for the solution to fill up the whole crystallizer and pipelines, with volume approximately 1 L per cycle. Pump was operated at the set circulation flow rate. Force was supplied by peristaltic pump to circulate the solution within the whole system. Pure water was also injected into the system in order to form seed ice lining on the cooling surface. It is important to prevent initial super cooling from happening which can cause high impurities in initial ice crystal formed due to contamination (Gunathilake et al., 2014). As the process proceeded, ice layer formed at the wall of crystallizer. The pressure inside the crystallizer increased and affected the whole process. A relief valve was installed at the crystallizer outlet to prevent overflow of the sample solution due to sudden increase in pressure in the crystallizer.

The concentrate collection, 5 min was set as the time delay. The outlet pipe of the crystallizer was facing upward, thus making the pumping force insufficient for peristaltic pump to pump the entire solution out of the crystallizer.

A small path was added at the lowest point of the crystallizer to aid the process of concentrate collection. Since the concentrate was still in the freezing temperature, a heater tape was wrapped around the path to ensure no occurrence of freezing process during concentrate collection. After all the concentrate had been completely removed, the coolant temperature was increased for the ice thawing process to occur. Time taken for ice thawing process was set to 60 minutes approximately while time delay was set to 5 minutes for the thawed ice collection process. After that, BOD_5, TSS and turbidity of thawed ice sample was analyzed by respective equipment using BODII Trak, filter apparatus with suction tube and nephelometer. The experiment was repeated by manipulating circulation time (5 min to 25 min) while fixing the coolant temperature at -17 °C in order to study the effect of circulation time. Meanwhile, for study the effect of coolant temperature, the experiment was varied in different coolant temperature in the range -5 °C to -17 °C and constant the circulation time at 15 min. Other operating conditions such as pump rotation speed was also kept constant which was 450 rpm. All the data collected at the end of experiment were recorded. Figure 1 shows the experimental setup of this study.

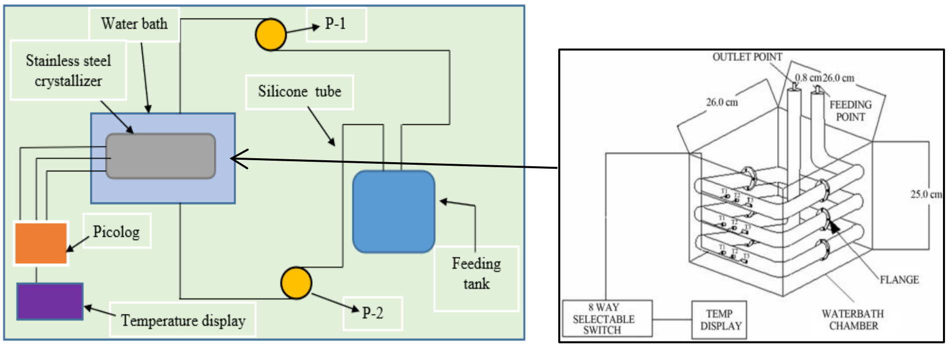


Figure 1: Schematic diagram of the complete PFC coil crystallizer system

3. Results and Discussions

3.1 Products Experiment

The comparison of the feed, concentrate and also product is shown in Figure 2 (a). From left hand side to the right, the sequence of the bottle samples are feed, concentrate and product. Originally the feed which was the lake water contained some suspended solids that make it greenish and muddy. The concentrate which acts as an unwanted waste of the process contained more suspended solids. The product is the thawed ice which is clean and without much suspended solids. During the process, the pure water formed as ice crystal layer on the inner wall of the crystallizer, while the foreign particles were repelled from the ice crystal layer. Figure 2 (b) indicates the complete formation of ice crystal during the PFC process.

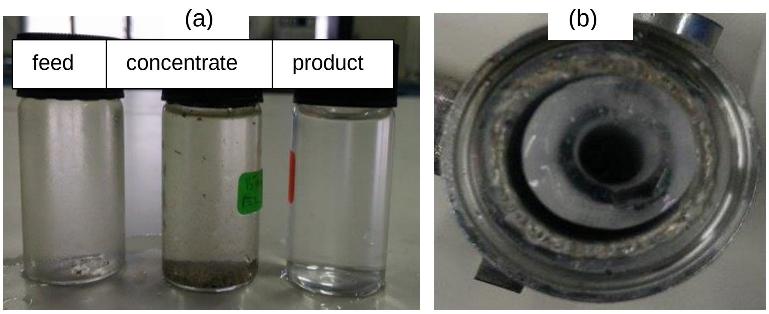
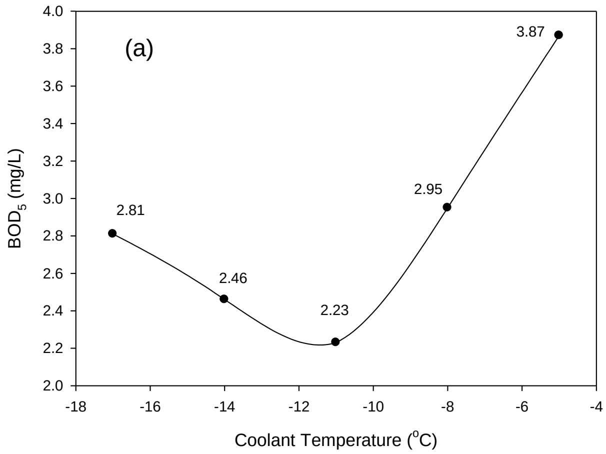


Figure 2: (a) Comparison between feed, concentrate and product (b) Ice crystal layer formed in coil crystallizer during PFC

3.2 Effect of Coolant Temperature

Based on the melting profile of pure water and lake water, the studied range of coolant temperature chosen was from -5 °C to -17 °C. As there were impurities in the water, the freezing point decreased below the freezing point of pure water (0 °C) because lake water which was impure has lower melting point. At temperature of 0 °C, pure water will freeze first leaving behind the lake water which has lower freezing point. The other operating parameters were kept constant while the coolant temperature was varied. Pump rotating speed was kept at 450

rpm while the circulation time was fixed at 15 min. After the products of ice crystal were obtained, all of them were analyzed for BOD, TSS and turbidity. The results are illustrated in Figure 3 (a), (b) and Figure 4 for all parameters. From the results obtained, coolant temperature of -11 °C is considered as the most optimum condition as it gave the lowest value of BOD_5, TSS and turbidity. The values of these parameters experienced a drastic decrease from -5 °C to -11°C, indicating that the lower the coolant temperature, the higher the system efficiency in this range of coolant temperature. Nevertheless, when the coolant temperature decreased further from -11 °C to -17 °C, the trend changed and the value of parameters started to increase gradually. This means that the system performance will start to drop if the coolant temperature goes too low. After coolant temperature of -11°C, there is a chance of the solutes in lake water originally will be trapped within the ice formed. This results in higher impurities in the thawed ice and thus lower system efficiency.
The difference in coolant temperature dictates the thickness of the ice formed as well. As the coolant temperature goes lower, the thickness of the ice formed goes higher. However, thicker ice layer formed does not ensure better quality of product. This is because the ice growth rate is influenced by the coolant temperature, whereby the ice crystal growth rate needs to be low to produce high purity of ice (Miyawaki et al., 2016). In slow freezing, most of the impurities were repelled out of the ice crystal thus producing high purity ice. When the ice crystal growth rate is too high, the solutes are more prone to entrapment in ice crystal layer. This phenomenon is due to fact that the solutes in the lake water moving towards the ice crystal layer which occurred at a faster rate than the outward movement of solutes from the ice crystal layer formed as a result of the high ice crystal growth rate.



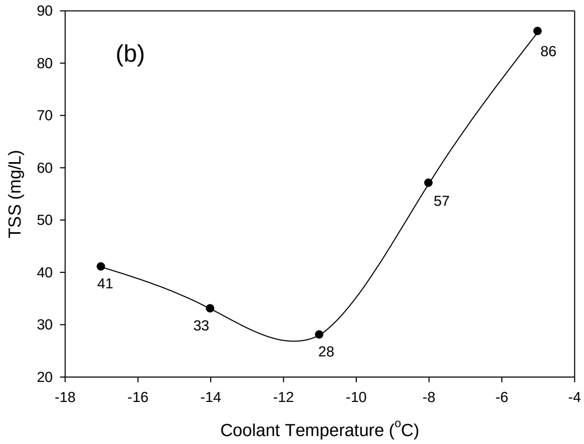


Figure 3: (a) Effect of coolant temperature on BOD_5 (b) Effect of Coolant Temperature on TSS

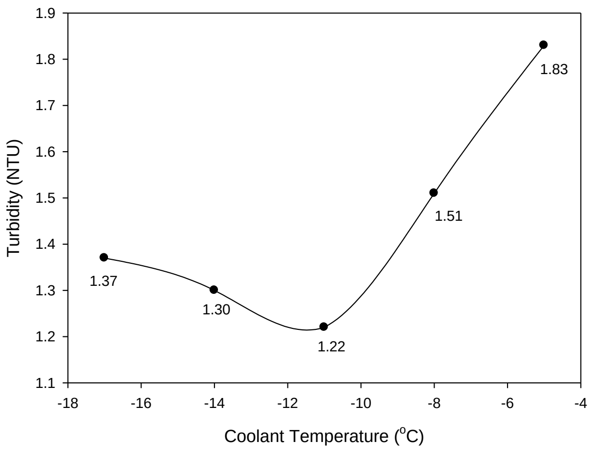


Figure 4: Effect of Coolant Temperature on Turbidity

This resulted in solid inclusion in the ice crystal layer. The BOD_5, TSS and turbidity of thawed ice would increase as a result of higher impurities entraped within the ice crystal due to the reason mentioned earlier.

3.3 Effect of Operation Time

The results of the experiment are illustrated as in Figure 5 (a), (b) and Figure 6 for all parameters. From 5 min to 10 min, the values of the parameters decreased which is highly desired. However, as the circulation time increased further from 10 min to 25 min, what can be observed was a drastic increase in the values of the parameters. It can be concluded that the optimum circulation time for the progressive freeze concentration is 10 minutes. As the circulation time became much higher than 10 min, it started to produce ice crystal with high impurities. As the solution was circulated for more than 10 min, the ice formed became eventually thicker which filled up almost the entire crystallizer. This resulted in narrow space for the unwanted concentrate to circulate smoothly around the crystallizer. When the ice crystal layer became very thick while the concentrate become stagnant, the contaminants were easily trapped in the ice crystal layer formed (Luo et al., 2010).

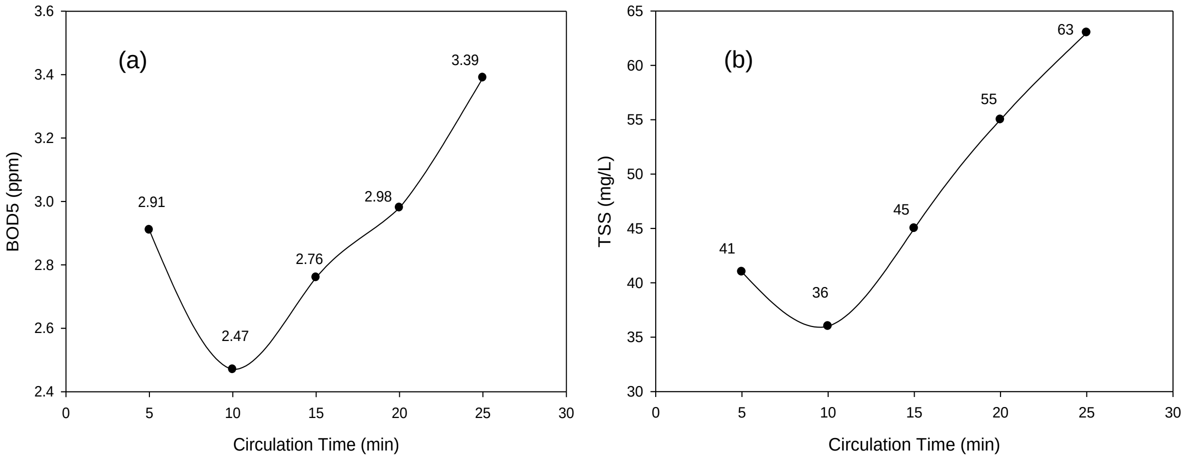


Figure 5: (a) Effect of on Circulation Time BOD_5 (b) Effect of Circulation Time on TSS

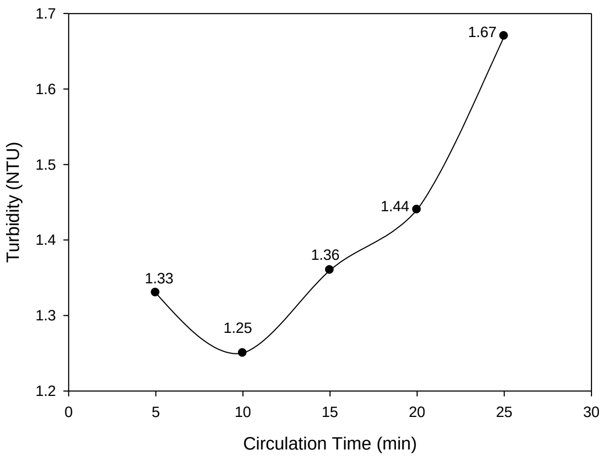


Figure 6: Effect of Circulation Time on Turbidity

It is also noted that 5 min of circulation time did not give good result as well compared to 10 min of circulation time. This is because during that time the ice crystal layer was not completely formed, which was still thin and not smooth. The dendritic structure of it will trap the solutes from lake water in the growing ice crystal layer. From the analysis of three different parameters, it is shown that a circulation time of 10 minwould be the most optimum condition to produce ice crystal of the lowest BOD_5, TSS and turbidity.

4. Conclusion

Progressive freeze concentration has a splendid potential on water purification and known as one of the most effective separation process to concentrate a solution by using the natural properties of ice crystal lattice. In this case, the most optimum condition for progressive freeze concentration to work is at coolant temperature of -11°C and circulation time of 10 min as it gave the lowest value of biological oxygen demand (BOD_5), total suspended solids (TSS) and turbidity in the ice layer produced which was reduced from 16.83 mg/L to 2.23

mg/L and 2.47 mg/L for BOD_5, 566 mg/L to 28 mg/L and 36 mg/L for TSS and while for turbidity from 300 NTU to 1.22 NTU and 1.25 NTU. Low value of all the parameters indicate that the solutes within the thawed ice were in the least amount for microorganism to survive, thus the purest. If compared with other methods such as reverse osmosis and evaporation, progressive freeze concentration has lower capital and maintenance cost. Besides, low temperature operation causes corrosion to be insignificant during the process making it more favorable. It is also give an enough evident for the result collected that is qualified in reducing of BOD_5, TSS and turbidity in this technology.

Acknowledgments

The financial support from Research University Grant (10H45) from Universiti Teknologi Malaysia and Ministry of Higher Education (MOHE) is gratefully acknowledged.

Reference

Ab Hamid F.H., Rahim N.A., Johari A., Ngadi N., Zakaria Z.Y., Jusoh M., 2015, Desalination of seawater through progressive freeze concentration using a coil crystallizer, Water Science and Technology-Water Supply 15 (3), 625-631.

Adeleke Abdul Rahman O., Nik Daud N.N., Ahsan Amimul, Biswajeet Pradhan, 2014, Water quality assessment of UPM Lake and the impact of geographic information system, International Journal of Environmental Monitoring and Analysis 2 (3), 158-162.

Amrita, 2011, Water analysis-determination of physical parameters, <vlab.amrita.edu/?sub=2&brch=193 &sim=575&cnt=1> accessed 10.10.2016.

APHA, 1992, Standard methods for the examination of water, John Wiley & Sons, Ontario, Canada.

Gunathilake M., Dozen M., Shimmura K., Miyawaki O., 2014, An apparatus for partial ice-melting to improve yield in progressive freeze-concentration, Journal of Food Engineering 142, 64-69.

Gupta A., Rai D.K., Pandey R.S., Sharma B., 2009, Analysis of some heavy metals in the riverine water:sediments and fish from river Ganges at Allahabad, Environmental Monitoring Assessment 157, 449-458.

Jusoh M., Mohd Yunus R. and Abu Hassan M. A., 2008, Effect of flowrate and coolant temperature on the efficiency of progressive freeze concentration on simulated wastewater, World Academia Science and Engineering Technology 47, 75-78.

Luo C.S., Chen W.W., Han W.F., 2010, Experimental study on factors affecting the quality of ice crystal during the freezing concentration for the brackish water, Desalination 260 (1–3), 231-238.

Ministry of the Environment, 2016, State of Japan's environment at a glance: Japanese Lake environment- the importance of lakes <www.env.go.jp/en/water/wq/lakes/why.html> accessed 31.07.2016.

Miyawaki O., Kato S., Watabe K., 2012, Yield improvement in progressive freeze-concentration by partial melting of ice, Journal of Food Engineering 108 (3), 377-382.

Miyawaki O., Gunathilake M., Omote C., Koyanagi T., Sasaki T., Take H., Matsuda A., Ishisaki K., Miwa S. Kitano S., 2016, Progressive freeze-concentration of apple juice and its application to produce a new type apple wine, Journal of Food Engineering 171, 153-158.

Mohd Yusof F., 1996, Environmental management of Lake Kenyir, Malaysia: Prevention of eutrophication, Jurnal Engineering Science 9, 43-48.

Mohd Ekhwan Toriman and Large A., 2004, Process of channel platform change on meandering channel in the Langat River, Malaysia: Implication to river management, Proceeding of 2nd Bangi, World Conference Studies, Universiti Kebangsaan Malaysia, Bangi, Selangor.

National Water Quality Standard, 2016, National water quality standards for Malaysia <www.wepa-db.net/policies/law/malaysia/eq_surface.htm> accessed 09.11.2016.

Rodríguez M., Luque S., Alvarez J., Coca J., 2000, A comparative study of reverse osmosis and freeze concentration for the removal of veleric acid from wastewaters, Desalination 127 (1), 1-11.

Rahaman A.Z., Che Rus, S.F., Omar M.A., Ismail W.S., 2016, River and lake as natural heritage: water quality status in the Northern states of Peninsular Malaysia, KEMANUSIAAN 23, 109-128.

Shafirah Samsuri, Nurul Aini Amran, Norshafika Yahya, Mazura Jusoh, 2016, Review on progressive freeze concentration designs, Chemical Engineering Communication 203 (3), 345-363.

Sujaul Islam M., Ismail B.S. Muhammad Barzani G. Sahibin A.R., Mohd Ekhwan T., 2012, Hydrological assessment and water quality characteristics of Chini Lake, Pahang, Malaysia, American-Eurasian Journal Agriculture and Environment Science 12 (6), 737-749.

Yisa J., Jimoh T., 2010, Analytical studies on water quality index of River Landzu, American Journal of Applied Science 7, 453-458.