EXTRACTION OF BETACYANIN FROM *Bougainvillea glabra* BRACT AND THE ADSORPTION ONTO FIBRES

NUR ATHIRAH BINTI SABARUDIN

Master of Science

UNIVERSITI MALAYSIA PAHANG

SUPERVISOR'S DECLARATION

We hereby declare that We have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

_____

(Supervisor's Signature)

Full Name : PROFESSOR DATIN DR. MIMI SAKINAH BINTI ABDUL MUNAIM

Position : PROFESSOR

Date :

_____

(Co-supervisor's Signature)

Full Name : PROFESSOR DATO' DR. ZULARISAM BIN AB WAHID

Position : PROFESSOR

Date :

STUDENT'S DECLARATION

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

_____

(Student's Signature)

Full Name : NUR ATHIRAH BINTI SABARUDIN

ID Number : MKB15001

Date :

EXTRACTION OF BETACYANIN FROM *Bougainvillea glabra* BRACT AND THE ADSORPTION ONTO FIBRES

NUR ATHIRAH BINTI SABARUDIN

Thesis submitted in fulfillment of the requirements
for the award of the degree of
Master of Science

Faculty of Engineering Technology
UNIVERSITI MALAYSIA PAHANG

2018

ACKNOWLEDGEMENTS

In the name of Allah, the most gracious and the most merciful.
May the peace and blessings be upon prophet Muhammad (SAW).

Alhamdulillah, thanks to Allah the Almighty because only with His pure blessing and mercy, this research managed to be accomplished successfully. First and foremost, I would like to thank my beloved parents, Mr. Sabarudin bin Mohd Arif and Mrs. Wan Nor Aspalaila binti Wan Muda for their continuous support, sacrifice and love throughout my life. My sisters and brothers, and also other family members for all loves and support.

I would like to express my special appreciation and thanks to my supervisor, Prof. Datin Dr. Mimi Sakinah binti Abdul Munaim for her supervision, invaluable guidance from the very early stage of this research until the finish writing phase. I would like to thank her for never ending support during my tenure as research student under her guidance, would be a difficult one. Without her advice and assistance this thesis would not have been possible. Her ideas and knowledge contributed to the success of this research. I sincerely thank for the time spent proofreading and correcting my mistakes.

I am also indebted to the Ministry of Higher Education for funding my fees studies with MyBrain15. Millions of thanks to my Co. SV Prof. Dato' Dr. Zularisam bin Ab Wahid for funding me with his grant during my study in UMP with grant no:RDU150801.

I would also like to thank my buddy Nurul Ain, my housemates (Kak Sab and Kak Erin), labmates (Kak Baiyi and Kak Zimah), and friends for the help, support, knowledge that being shared together and motivate me to strive towards my goal. My sincere thanks go to staff of Universiti Malaysia Pahang, especially to all lecturers and staff of Faculty of Chemical Engineering and Natural Resources and Faculty of Engineering Technology for providing me a platform to pursue my degree Master of Science and help me in many ways for going through my education in Universiti Malaysia Pahang. Finally, I would like to give my sincere appreciation to everybody who was involved directly or indirectly in the realization of this thesis.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF SYMBOLS

C_B	Solute bound per unit amount of adsorbent
C_e	Equlibrium dye concentration
C_i	Initial dye concentration
C_u	Unbound solute concentration
K_F	Freundlich adsorption constant
K_L	Affifnity constant for Langmuir isotherm
£	Sterling pound
kHz	Kilo hertz
q_m	Maximum capacity of adsorption
λ_{max}	Maximum wavelength

LIST OF ABBREVIATIONS

ANN	Artificial Neutral Network
ANOVA	Analysis of Variance
CCD	Central Composite Design
COD	Chemical Oxygen Demand
FTIR	Fourier Transform Infra-Red
HCL	Hydrochloric
HPLC	High Performance Liquid Chromatography
ITO	Indium tin oxide
KBr	Dry potassium bromide
MLR	Material-liquid ratio
MS	Mass spectrophotometer
OFAT	One Factor At Time
RPM	Rotation per minute
RSM	Response surface methodology
SEM	Scanning electron microscopy
SFE	Supercritical fluid extraction
SLR	Solid-liquid ratio
sp.	species
UV	Ultra violet
IP-HSCCC	Ion –pair high-speed countercurrent chromatography
ESI-MS-MS	Electrospray ionization mass-spectrometry

EXTRACTION OF BETACYANIN FROM *Bougainvillea glabra* BRACT AND THE ADSORPTION ONTO FIBRES

NUR ATHIRAH BINTI SABARUDIN

Thesis submitted in fulfillment of the requirements
for the award of the degree of
Master of Science

Faculty of Engineering Technology
UNIVERSITI MALAYSIA PAHANG

2018

ABSTRAK

Kepentingan terhadap penggunaan pewarna asli semakin meningkat di seluruh dunia. Ini disebabkan oleh persepsi terhadap peningkatan risiko kepada alam sekitar dan kesihatan yang berkait dengan pemprosesan dan penggunaan pewarna sintetik. Bunga kertas merupakan bunga taman yang berwarna merah-ungu mengandungi betacyanin, iaitu pigmen betalain. Walau bagaimanapun, penggunaan bunga kertas sebagai pewarna semula jadi tekstil masih belum diketahui kerana kekurangan maklumat mengenai keserasian dalam pencelupan tekstil. Kajian ini dijalankan untuk mengkaji proses pengekstrakan betacyanin dari kelopak bunga kertas dan penjerapan pewarna pada benang. Fokus kajian ini adalah untuk mencari keadaan pengekstrakan pigmen yang terbaik menggunakan Metodologi Gerak Balas Permukaan (RSM) dan menentukan isoterma penjerapan pewarna pada benang. Dalam usaha untuk menentukan keadaan proses pengekstrakan yang terbaik, faktor pada satu masa (OFAT) telah dikaji iaitu pH (1.0, 4.0, 7.0, dan 10.0), nisbah pepejal-cecair (5:100, 7:100, 9:100, 11:100, 13:100, dan 15:100) dan masa pengekstrakan (0-180 min) menggunakan proses pengekstrakan pepejal-cecair. Keamatan warna pewarna dan kumpulan berfungsi bagi sampel telah dianalisis menggunakan Spektrofotometer UV-Vis dan Spektroskopi Infra-Merah Transformasi Fourier (FTIR). Kajian terhadap kesan kepekatan awal pewarna dan masa keseimbangan untuk proses penjerapan juga telah dijalankan. Hasil kajian mendedahkan bahawa keaadaan terbaik bagi proses pengekstrakan adalah dalam keadaan berasid pada pH 4.0 berbanding dengan alkali, dengan nisbah pepejal-cecair 11:100. Masa keseimbangan dicapai selepas 60 minit proses pengekstrakan bermula. Penemuan mendapati 100 gram kelopak bunga dapat menghasilkan sekitar 68.12 g betacyanin. Bagi proses penjerapan, benang sutera mempunyai kecenderungan yang tinggi dalam mengekalkan warna berbanding dengan kapas polyester dan rayon. Proses penjerapan memberitahu bahawa benang sutera adalah lebih sesuai dicelup dengan pewarna semula jadi berbanding dengan benang sintetik. Penerapan model isoterma dalam meramalkan kapasiti penjerapan pewarna semula jadi menunjukkan bahawa model Freundlich menunjukkan padanan yang lebih baik berbanding model Langmuir untuk menggambarkan interaksi antara pewarna dan benang.

ABSTRACT

The importance of using natural dyes is increasing worldwide. This is due to the perception of increased environmental and health risks associated with the processing and use of synthetic dyes. *Bougainville glabra* is known as a red-violet flower garden that contains betacyanin, a betalain pigment. However, the use of *Bougainvillea glabra* as a natural colouring of textiles is still unknown because of the lack of information on the compatibility in textile dyeing. This research carried out the extraction process of betacyanin from *Bougainvillea glabra* bracts and the adsorption of the dyes on the fibres. The focus of this study was to find the best dye extraction condition using Response Surface Methodology (RSM) and to determine the adsorption isotherm of dyes on the fibres. In order to determine the best condition of extraction process, one factor at time (OFAT) was studied that is pH (1.0, 4.0, 7.0, and 10.0), solid-liquid ratio (5:100, 7:100, 9:100, 11:100, 13:100, and 15:100) and extraction time (0-180 min) using solid-liquid extraction process. Colour intensity of the dyes and the functional groups of the samples were analyzed using UV-Vis Spectrophotometer and Fourier Transform Infra-Red Spectroscopy (FTIR). The determination of the effects initial concentration of dye and contact time for the adsorption process was studied. The findings have shown that extraction process was better in acidic condition at pH 4.0 than in alkaline, with solid-liquid ratio (SLR) of 11:100. The equilibrium time was achieved after 60 min of extraction process begins. The optimization process revealed that the best condition for betacyanin dye extraction was at 10:100 solid-liquid ratio (SLR) and extraction time of 60 min. From the results, it was estimated that 100 gram of bract can produce about 68.12 g of betacyanin. For adsorption process, spun silk has the highest affinity in retaining the dye compared to rayon and polyester-cotton. The adsorption process tells that spun silk is favourable to be dyed with natural dye compared to the synthetic fibre. The application of the isotherm model in predicting the adsorption capacity of natural dye shows that Freundlich model was fitted better than Langmuir model for describing the interaction between the dye and the fibres.

REFERENCES

Abdul Hamid, N. A. (2013). *Natural dye products business plan*, (Master's thesis). Universiti Malaysia PAhang, Gambang.

Aljeboree, A. M., Alshirifi, A. N., & Alkaim, A. F. (2014). Kinetics and equilibrium study for the adsorption of textile dyes on coconut shell activated carbon. *Arabian Journal of Chemistry*, 1-13.

Azami, M., Bahram, M., & Nouri, S. (2013). Central composite design for the optimization of removal of the azo dye, Methyl Red, from waste water using Fenton reaction. *Current Chemistry Letters, 2*(2): 57–68.

Azeredo, H. M. C. (2009). Betalains: properties, sources, applications, and stability. *International Journal of Food Science & Technology, 44*(12): 2365–2376.

Azimah, S. (2014). *Separation of Betacyanin from Hylocerus Polyrhizus peel using ultrafiltration membrane*, (Master's thesis). Universiti Malaysia Pahang.

Baishya, D., Talukdar, J., & Sandhya, S. (2012). Cotton dying with natural dye extracted from flower of Bottlebrush (*Callistemon citrinus*). *Universal Journal of Environmental Research and Technology, 2*(5): 377–382.

Beeby, J. L. (2006). The theory of desorption. *Critical Reviews in Solid State and Materials Science, 7*(2): 153–166.

Bhute, K. H. P. & A. S. (2012). Plant based dyes and mordant : A Review. *Journal of Natural Products and Plant Resources, 6*(November): 649–664.

Blainski, A., Lopes, G. C., & de Mello, J. C. P. (2013). Application and analysis of the folin ciocalteu method for the determination of the total phenolic content from *Limonium Brasiliense* L. *Molecules, 18*: 6852–6864.

Bogdanov, M. G., & Svinyarov, I. (2013). Ionic liquid-supported solid – liquid extraction of bioactive alkaloids. II. Kinetics, modeling and mechanism of glaucine extraction from Glaucium flavum. *Separation and Purification Technology, 103*: 279–288.

Boonsong, P., Laohakunjit, N., Kerdchoechuen, O., Thonburi, T., & Matta, F. B. (2011). Detection of pigments and natural colorants from Thai herbal plants for possible use as coloring dyes. *Hort Science, 46*(2): 265–272.

Branch, B. (2010). Functional dyeing of wool with natural dye extracted from *Berberis vulgaris* wood and *Rumex Hymenosepolus* root as biomordant. *Iran Journal Chemical Chemical Engineering, 29*(3): 55–60.

Camenzind, M., Freitag, R., & Furter, R. (2013). *Raw Silk and Spun Silk Quality and Measurement*.

Cao, S., Liu, T., Jiang, Y., He, S., Harrison, D. K., & Joyce, D. C. (2012). The effects of host defence elicitors on betacyanin accumulation in *Amaranthus mangostanus* seedlings. *Food Chemistry, 134*(4): 1715–1718.

Chan, L. S., Cheung, W. H., Allen, S. J., & McKay, G. (2012). Error analysis of adsorption isotherm models for acid dyes onto bamboo derived activated carbon. *Chinese Journal of Chemical Engineering, 20*(3): 535–542.

Chandrasekhar, J., Sonika, G., Madhusudhan, M. C., & Raghavarao, K. S. M. S. (2015). Differential partitioning of betacyanins and betaxanthins employing aqueous two phase extraction. *Journal of Food Engineering, 144*: 156–163.

Chavan, R. (2001). Indian textile industry-environmental issues. *Indian Journal of Fibre and Textile Research, 26*: 11–21.

Chemat, F., Vian, M. A., & Cravoto, G. (2016). Green extraction of natural products : Concept and principles. *International Journal of Molecular Science, 13*(7): 8615–8627.

Chowdry, S., Chakraborty, S., & Das Saha, P. (2013). Response surface optimization of a dynamic dye adsorption process: a case study of crystal violet adsorption onto NaOH-modified rice husk. *20*: 1698–1705. doi: 10.1007/s11356-012-0989-7

Chung, H., Kim, W., Park, J., Cho, J., Jeong, T., & Park, P. (2015). Application of Langmuir and Freundlich isotherms to predict adsorbate removal efficiency or required amount of adsorbent. *Journal of Industrial and Engineering Chemistry, 28*: 241–246.

Cordeiro de Azeredo, H. M., Pereira, A. C., Rodrigues de Souza, A. C., Gouveia, S. T., & Barbosa Mendes, K. C. (2009). Study on efficiency of betacyanin extraction from red beetroots. *International Journal of Food Science & Technology, 44*(12): 2464–2469.

Deka, A., Chougule, B. S., Parveen, A., & Lahan, J. P. (2014). Natural pigment betacyanin as tracking dye for gel electrophoresis. Indian Journal of Natural Product and Resources, *6*(March): 23–26.

Delgado-Vargas, F., Jimez, A. R., & Paredes-Lopez, O. (2014). Natural pigments: carotenoids, anthocyanins, and betalains — Characteristics, biosynthesis, processing, and stability. *Critical Reviews in Food Science and Nutrition, 40* (3):173-289. doi: 10.1080/10408690091189257

Denni, M., & Rt, S. (2012). Identification and characterization of a betacyanin pigment from the semi-arid plant *Aerva lanata* juss ex . Schultes. *International Journal of Research in Pharmaceutical and Biomedical Sciences*, *3*(1): 212–214.

Desta, M. B. (2013). Batch Sorption Experiments : Langmuir and Freundlich Isotherm studies for the adsorption of textile metal ions onto Teff Straw (*Eragrostis tef*) Agricultural Waste. *Journal of Thermodynamics, 2013*: 1-6. doi: 10.1155/2013/375830

Dikshith, T. S. S. (2008). Tin and its compounds. *Hazardous Chemicals: Safety Management and Global Regulations* (pp. 388–390).New York. CRC Press.

Elmorsi, T. M. (2011). Equilibrium isotherms and kinetic studies of removal of methylene blue dye by adsorption onto miswak leaves as a natural adsorbent. *Journal of Environmental Protection*, *2011*(August): 817–827. doi: 10.4236/jep.2011.26093

Fouda, I. M., Seisa, E. a., & Eid, F. M. (2015). Characterizations of the optical and structure properties of rayon acetate fibers due to different thermal conditions. *Optik - International Journal for Light and Electron Optics*, *126*(2): 258–264. doi: 10.1016/j.ijleo.2014.08.169

Gamal, A. M., Farha, S. A. A., Sallam, H. B., Mahmoud, G. E. A., & Ismail, L. F. M. (2010). Kinetic Study and Equilibrium Isotherm Analysis of Reactive Dyes Adsorption onto Cotton Fiber. *Nature and Science*, *8*(11): 95–110.

Garcia, E. R., Medina, R. L., Lozano, M. M., Perez, I. H., Valero, M. J., & Franco, M. M. (2014). Adsorption of azo-dye orange II from aqueous solutions using a metal-organic framework material: iron- benzenetricarboxylate. *Materials,7*: 8037–8057. doi:10.3390/ma7128037

Gaurav, P., Kumar, J. N., Narendra, N., & Chatap, V. K. (2010). Bougainvillea glabra – A Natural Indicator. *Pharmacognosy Journal, 2*(5): 25–28.

Ghasemi, E., Raofie, F., & Najafi, N. M. (2011). Application of response surface methodology and central composite design for the optimisation of supercritical fluid extraction of essential oils from *Myrtus communis* L . leaves. *Food Chemistry*, *126*(3): 1449–1453.

Ghosh, R. (2006). Overview of bioseparation engineering. *Principles of bioseparations engineering* (pp. 1-12).Singapore: World Scientific Publishing Co. Pte. Ltd.

Grifoni, D., Bacci, L., Zipoli, G., Albanese, L., & Sabatini, F. (2011). The role of natural dyes in the UV protection of fabrics made of vegetable fibres. *Dyes and Pigments, 91*(3): 279–285.

Gupta, V., George, M., Joseph, L., Singhal, M., & Singh, H. P. (2009). Evaluation of antibacterial activity of *Bougainvillea* glabra 'snow white' and *Bougainvillea* glabra 'choicy'. *Journal of Chemical and Pharmaceutical Research, 1*(1): 233–237. Retrieved from http://www.jocpr.com/articles/evaluation-of-antibacterial-activity-of-bougainvillea -glabra-snow-white-and-bougainvillea-glabra-choicy.pdf

Ha, G., & Kim, J. (2016). Kinetic and thermodynamic characteristics of ultrasound-assisted extraction for recovery of paclitaxel from biomass. *Process Biochemistry, 51*(2016): 1664–1673. doi: 10.1016/j.procbio.2016.08.012

Habip, D., Dilek, K., Nigar, M., & Seyda, C. (2015). The effect of dyeing properties of fixing agent and plasma treatment on silk fabric dyed with natural dye extract obtained from *Sambucus Ebulus* L. plant. *Procedia- Social and Behavioral Sciences, 195*(2015): 1609–1617. doi: 10.1016/j.sbspro.2015.06.201

Haji, A. (2012). Antibacterial dyeing of wool with natural cationic dye using metal mordants. *Materials Science, 18*(3): 267–270. doi: 10.5755/j01.ms.18.3.2437

Hamid, N., Abdul Munaim, M. S., & Abu Seman, M. N. (2016). Regression analysis for the adsorption isotherms of betacyanin extracts from the dragon fruit peel onto the spun silk yarn. *International Journal of Engineering Technology and Sciences, 6*(1): 44–53. doi: 10.15282/ijets.6.2016.1.7.1057

Haque, N., Hannan, M. A., & Rana, M. (2015). Compatibility analysis of reactive dyes by exhaustion-fixation and adsorption isotherm on knitted cotton fabric. *Fashion and Textiles, 2*(3): 1-12. doi: 10.1186/s40691-015-0026-9

Hasan, M., Nayem, K. A., Yousuf, A., Anwarul, M., & Ghosh, N. C. (2015). Application of purified lawsone as natural dye on cotton and silk fabric. *Journal of Textiles,2015*: 1–8. doi: 10.1155/2015/932627

Hassan, S. S. M., Awwad, N. S., & Aboterika, A. H. A. (2009). Removal of synthetic reactive dyes from textile wastewater by Sorel's cement. *Journal of Hazardous Materials, 162*: 994–999. doi: 10.1016/j.jhazmat.2008.05.138

Hemmatzadeh, R., & Mohammadi, A. (2013). Improving optical absorptivity of natural dyes for fabrication of efficient dye-sensitized solar cells. *Journal of Theoretical and Applied Physics, 7*(57): 1-7.

Hernandez, A. R., Estevez, M., Vargas, S., Quintanilla, F., & Rodriguez, R. (2011). New dye-sensitized solar cells obtained from extracted bracts of *Bougainvillea* glabra and spectabilis betalain pigments by different purification processes. *International Journal of Molecular Sciences, 12*(9): 5565–76.

Hosseini, M., Montazer, M., & Damerchely, R. (2013). Enhancing Dye-ability and Antibacterial Features of Silk through Pre-treatment with Chitosan. *Journal of Engineered Fibers and Fabrics, 8*(3): 102–111.

Hou, X., Chen, X., Cheng, Y., Xu, H., Chen, L., & Yang, Y. (2013). Dyeing and UV-protection properties of water extracts from orange peel. *Journal of Cleaner Production, 52*(2013): 410–419.

Ibrahim, U. K., Muhammad, I. I., & Salleh, R. M. (2011). The effect of pH on color behavior of *Brassica oleracea* anthocyanin. *Journal of Applied Sciences, 11*(13): 2406–2410.

Jerz, G., Wybraniec, S., Gebers, N., & Winterhalter, P. (2010). Target-guided separation of *Bougainvillea* glabra betacyanins by direct coupling of preparative ion-pair high-speed countercurrent chromatography and electrospray ionization mass-spectrometry. *Journal of Chromatography. A, 1217*(27): 4544–4554.

Jothi, D. (2008). Extraction of natural dyes from african marigold flower (T*agetes Erecta* L). *AUTEX Research Journal, 8(*2): 49–53. Retrieved from http://www.autexrj.com/cms/zalaczone_pliki/4-08-2.pdf

Jung, J. Y., Choi, M. S., & Yang, J. K. (2013). Optimization of concentrated acid hydrolysis of waste paper using response surface methodology. *Journal of Korean Wood Science and Technology, 41*(2): 87–99. doi: 10.5658/WOOD.2013.41.2.87

Jyothi, S. N. & S. (2012). A comparative phytochemical analysis of *Bougainvillea* glabra choisy and calforina gold. *International Journal of Pharma and Bio Sciences 3*(3): 247–250.

Kalirajan, A., Mariselvam, R., Michael, J. S., Narayanan, K. R., Narayanan, G. A., & Singh, A. J. A. R. (2012). Utilization of *Bougainvillea* glabra for prepared natural colouring agent and biopesticides. *International Journal of Current Research, 4*(9): 9–11.

Kaur, M. (2014). *Adsorption study of betacyanin of fibre*. URP Thesis. Universiti Malaysia Pahang. Gambang.

Kechi, A., Chavan, R. B., & Moeckel, R. (2013). Dye yield, color strength and dyeing properties of natural dyes extracted from ethiopian dye plants. *Textiles and Light Industrial Science and Technology, 2*(3): 137–145.

Khataee, A. R., Zarei, M., & Moradkhannejhad, L. (2010). Application of response surface methodology for optimization of azo dye removal by oxalate catalyzed photoelectro-Fenton process using carbon nanotube-PTFE cathode. *Desalination, 258*(2010): 112–119.

Killedar, S., Pawar, A., Nadaf, S., Nale, A., Tamboli, U., & Pishawikar, S. (2014). Novel analytical method development for some amide group containing drugs using *Bougainvillea* spectabilis bract extracts. *Asian Pacific Journal of Tropical Medicine, 7*(Suppl 1): S560–S567.

Kiurski, J., Adamovic, S., & Krstic, J. (2011). Adsorption efficiency of low-cost materials in the removal of Zn (II) ions from printing developer. *ACTA Technica Corviniensis - Bulletin of Engineering, 4*(2011): 61–66.

Koleva, V., & Simeonov, E. (2014). Solid liquid extraction of phenolic and flavonoid compounds from *Cotinus coggygria* and concentration by nanofiltration. *Chemical, Biochemical Engineering, 28*(4): 545–551. doi: 10.15255/CABEQ.2014.2006

Kreuzer, H. J. (1988). Theory of Desorption Kinetics. *Chemistry and Physic of Solid Surfaces VII*, 259–260.

Kugler, F., Stintzing, F. C., & Carle, R. (2007). Characterisation of betalain patterns of differently coloured inflorescences from *Gomphrena globosa* L. and *Bougainvillea* sp. by HPLC-DAD-ESI-MS^n. *Analytical and Bioanalytical Chemistry, 387*(2): 637–648.

Lee, J., Kang, M. H., Lee, K., & Lee, Y. (2013). Characterization of natural dyes and traditional korean silk fabric by surface analytical techniques. *Materials, 2013*(6) 2007–2025. doi: 10.3390/ma6052007

Li, F., Yang, Q., Hong, C., & Zhong, Z. (2010). Color analysis on natural polyphenolic dyed cotton cellulose fibers. *Journal of Fiber Bioengineering and Informatics 3*(2): 100–105. doi:10.3993/jfbi09201007

Mahmoud, A. S., Ghaly, A. E., & Brooks, S. L. (2007). Influence of temperature and pH on the stability and colorimetric measurement of textile dyes. *American Journal of Biotechnology and Biochemistry, 3*(1): 33–41.

Mai, S. D., Thuan, T. N., Trinh, L. T. N., Trinh, T. T. N., & Ngo, T. X. (2012). Survey the Betacyanin extraction from the flower of purple. *Journal Food Sicence and Engineering, 2*:(2012): 254

Manjunath, M. S., & Sannappa, J. (2011). Variation of amino acids in some biological and pharmaceutical sample using FT-IR study. *International Journal of Pharmacy and Technology, 3*(1): 1946–1951.

Maran, J. P., Priya, B., & Nivetha, C. V. (2015). Optimization of ultrasound-assisted extraction of natural pigments from *Bougainvillea* glabra flowers. *Industrial Crops and Products, 63*(2015): 182–189. doi: 10.1016/j.indcrop.2014.09.059

Martinez, M. P., & Tena, M. T. (2011). Focused ultrasound solid-liquid extraction and selective pressurised liquid extraction to determine bisphenol A and alkylphenols in sewage sludge by gas chromatography-mass spectrometry. *Journal of Separation Science, 34*: 2513-2522. doi: 10.1002/jssc.201100159

Miraboutalebi, S. M., Nikouzad, S. K., Peydayesh, M., Allahgholi, N., Vafajoo, L., & McKay, G. (2017). Methylene blue adsorption via maize silk powder: Kinetic, equilibrium, thermodynamic studies and residual error analysis. *Process Safety and Environmental Protection, 106*: 191–202.

Mirjalili, M., & Karimi, L. (2013). Extraction and characterization of natural dye from green walnut shells and its use in dyeing polyamide : focus on antibacterial properties. *Journal of Chemistry, 2013*:1-9

Mishra, N., Joshi, S., Tandon, V. L., & Munjal, A. (2009). Evaluation of anti-fertility potential of aqueous extract of *Bougainvillea* in swiss albino mice. *International Journal of Pharmaceutical Sciences and Drug Research, 1*(1): 19-23

Moghaddam, S. S., Moghaddam, M. R. A., & Arami, M. (2010). Coagulation / flocculation process for dye removal using sludge from water treatment plant : Optimization through response surface methodology. *Journal of Hazardous Materials, 175*(2010): 651–657. doi: 10.1016/j.jhazmat.2009.10.058

Munipan, M. A. (2013). *Effect Of Enzyme Concentration And Temperature On Viscosity And Betacyanin Content From Pitaya Waste Extract*. URP Thesis. Universiti Malaysia Pahang. Gambang.

Naderi, N., Ghazali, H. M., Shobirin, A., & Hussin, M. (2012). Characterization and quantification of dragon fruit (*Hylocereus polyrhizus*) betacyanin pigments extracted by two procedures. *Pertanika Journal of Tropical Agricultural Science 35*(1): 33–40.

Narayan, S., Kumar, A., Ritesh, S. K., Sharmila, G., & Muthukumaran, C. (2013). Extraction optimization and characterization of water soluble red purple pigment from floral bracts of *Bougainvillea* glabra. *Arabian Journal of Chemistry*. doi: 10.1016/j.arabjc.2013.07.047 (in press)

Noda, N., & Adachi, T. (2000). Induction of betacyanin synthesis and pigment accumulation in cell suspension cultures of *Portulaca*. *Plant Biotechnology*, *17*(1): 27-34.doi: 10.5511/plantbiotechnology.17.27

Norasiha, H. (2011). *Inclusion complex formation between natural dye extracted from pitaya fruit skin and beta -cyclodextrin:kinetic and thermodynamic studies.* Universiti Malaysia Pahang. Gambang.

Orsini, F., Pasquier, A. Du, Beaudoin, B., & Tarascon, J. M. (1998). In situ scanning electron microscopy (SEM) observation of interfaces within plastic lithium batteries. *Journal of Power Sources, 76*(1998): 19–29.

Owen, T. (2000). Fundamentals of modern UV-visible spectroscopy. Retrieved from https://www.agilent.com/cs/library/primers/Public/59801397_020660.pdf

Padamwar, M. N., & Pawar, A. P. (2004). Silk sericin and its applications : A review. *Journal of Scientific and Industrial Research, 63*(April): 323–329.

Patil, P. D., Rao, C. R., Wasif, A. I., & Nagla, J. R. (2014). Taguchi optimization for efficient extraction of a natural dye from *Bougainvillea* glabra bracts. *International Journal of Research in Engineering and Technology, 3*(4): 23–30.

Phebe, D., Chew, M. K., Suraini, A. A., Lai, O. M., & Janna, O. A. (2009). Red-fleshed pitaya (*Hylocereus polyrhizus*) fruit colour and betacyanin content depend on maturity. *International Food Research Journal, 242*(16): 233–242.

Pisitsak, P., Hutakamol, J., Jeenapak, S., & Wanmanee, P. (2016). Natural dyeing of cotton with *Xylocarpus granatum* bark extract : dyeing , fastness , and ultraviolet protection properties. *Fibers and Polymers 2016, 17*(4): 560–568.

Pradesh, A. (2014). Mass transfer enhancement through optimized extraction of a natural dye from *Bougainvillea* glabra Juss . bracts. *Indian Journal of Natural Products and Resources, 5*(4): 332–337.

Prusty, A. K., Das, T., Nayak, A., & Das, N. B. (2010). Colourimetric analysis and antimicrobial study of natural dyes and dyed silk. *Journal of Cleaner Production, 18*(16–17): 1750–1756.

Radojkovic, M., Zekovic, Z., Jokic, S., Vidovic, S., Lepojevi, Z., & Milosevic, S. (2012). Optimization of Solid-liquid extraction of antioxidants from black Mulberry leaves by response surface methodology. *Food Technology Biotechnology, 50*(2): 167-176.

Rajendran, R., Balakumar, C., Kalaivani, J., & Sivakumar, R. (2011). Dyeability and antimicrobial properties of cotton fabrics finished with *Punica Granatum* extracts. *Journal of Textile and Apparel, Technology and Management, 7*(2): 1–12.

Raymond H. M., Douglas C. M., & Christine, M. A. C. (2009). *Response Surface Methodology: Process and Product Opimization Using Designed Experiments, 3rd Edition*. John Wiley and Sons.

Reshmi, S. K., Aravindhan, K. M., & Devi, P. S. (2012). The effect of light, temperature, ph on stability of betacyanin pigments in *Basella Alba* fruit. *Asian Journal of Pharmaceutical and Clinical Research, 5*(4): 5–8.

Ristić, N., & Ristić, I. (2012). Cationic modification of cotton fabrics and reactive dyeing characteristics. *Journal of Engineered FIbers and Fabrics, 7*(4): 113–121.

Rossi, T., Silva, P. M. S., Moura, L. F. De, Araújo, M. C., Brito, J. O., & Freeman, H. S. (2016). Waste from eucalyptus wood steaming as a natural dye source for textile fibers. *Journal of Cleaner Production,*:1-8.doi:10.1016/j.jclepro.2016.12.109

Rungruangkitkrai, N., & Mongkholrattanasit, R. (2012). Eco-Friendly of textiles dyeing and printing with natural dyes. *Proceedings of International Conference: Textiles & Fashion 2012*, Thailand, 1–17. Retrieved from http://textileconference.rmutp.ac.th/wp-content/uploads/2012/10/010-Eco-Friendly-of-Textiles-Dyeing-and-Print-with-Natural-Dyes.pdf

Sahraei, R., Farmany, A., & Mortazavi, S. S. (2013). A nanosilver-based spectrophotometry method for sensitive determination of tartrazine in food samples. *Food Chemistry Journal, 138*(2013): 1239–1242.

Samanta, A. K., & Agarwal, P. (2009). Application of natural dyes on textiles. *Indian Journal of Fibre & Textile Research, 34*(December): 384–399.

Saxena, S., &Raja, A. S. M. (2014). *Natural dyes: sources, chemistry, application and sustainability issues*. (S. S. Muthu, Ed.). Singapore: Springer Singapore.

Shah, B., Shah, A., & Shah, P. (2011). Sorption isotherms and column separation of Cu (II) And Zn (II) using ortho substituted benzoic acid chelating resins. *Archives of Applied Science Research, 3*(3): 327–341.

Shahid, M., Ull Islam, S., & Mohammad, F. (2013). Recent advancements in natural dye applications : a review. *Journal of Cleaner Production, 53*: 310–331.

Shoba, V., Krishnapriya, K.., Boopathy Raja, A., Elanchezhiyan, C., & S. (2013). Antimicrobial activity of *Bougainvillea* glabra flower extract doped mgs nanoparticles on *Escherichia coli*, *Salmonella typhi* and *Pseudomonas aeruginosa*. (1991): 2006–2009.

Singh, H. B., & Bharati, K. A. (2015). *Handbook of Natural Dyes and Pigments* (pp. 1–3).

Singh, R., Jain, A., Panwar, S., Gupta, D., & Khare, S. K. (2004). Antimicrobial activity of some natural dyes. *Dyes and Pigments*,: 5–8.(in press) Retrieved from https://pdfs.semanticscholar.org/73eb/f013f994d2956bb72d53b4004da5b7e838c2.pdf

Sinha, K., Chowdhury, S., Das, P., & Datta, S. (2013). Modeling of microwave-assisted extraction of natural dye from seeds of *Bixa orellana* (Annatto) using response surface methodology (RSM) and artificial neural network (ANN). *Industrial Crops & Products, 41(2013)*: 165–171. doi: 10.1016/j.indcrop.2012.04.004

Sinha, K., Das, P., & Datta, S. (2012). Dyes and pigments extraction of natural dye from petals of flame of forest (*Butea monosperma*) flower : Process optimization using response surface methodology (RSM). *Dyes and Pigments, 94*(2): 212–216.

Siva, R. (2015). Status of natural dyes and dye-yielding plants in India Status of natural dyes and dye-yielding plants in India. *Current Science*, 92(7): 916–925.

Sreekanth, D., Arunasree, M. K., Roy, K. R., Chandramohan Reddy, T., Reddy, G. V, & Reddanna, P. (2007). Betanin a betacyanin pigment purified from fruits of *Opuntia ficus-indica* induces apoptosis in human chronic myeloid leukemia Cell line-K562. *Phytomedicine : International Journal of Phytotherapy and Phytopharmacology*, *14*(11): 739–46.

Stamatopoulos, K., Chatzilazarou, A., & Katsoyannos, E. (2014). Optimization of multistage extraction of olive leaves for recovery of phenolic compounds at moderated temperatures and short extraction times. *Foods*, *3*: 66–81. doi: 10.3390/foods3010066

Stintzing, F. C., & Schieber, A. (2003). Evaluation of colour properties and chemical quality parameters of cactus juices. *European Food Research and Technology*, (216): 303–311. doi: 10.1007/s00217-002-0657-0

Strack, D., Vogt, T., & Schliemann, W. (2003). Recent advances in betalain research. *Phytochemistry* (pp. 247–269). doi: 10.1016/S0031?9422(02)00564?2

Vankar, P. S. (2000). Chemistry of Natural Dyes, (October): 73–80.

Vinod, K. N., & Puttaswamy. (2010). Kinetic and adsorption studies of Indian siris (*Albizia lebbeck*) natural dye on silk. *Indian Journal of Fibre & Textile Research, 35*(June): 159–163.

Wang, X., Wu, Q., Wu, Y., Chen, G., Yue, W., & Liang, Q. (2012). Response surface optimized ultrasonic-assisted extraction of flavonoids from Sparganii Rhizoma and evaluation of their in vitro antioxidant activities. *Molecules,17*: 6769–6783. doi: 10.3390/molecules17066769

Wanyama, P. A. G., Kiremire, B. T., Ogwok, P., & Murumu, J. S. (2010). The effect of different mordants on strength and stability of colour produced from selected dye-yielding plants in Uganda. *International Archieve of Applied Sciences and Technology, 1*(2): 81–92.

Wen, X., Yang, Q., Yan, Z., & Deng, Q. (2011). Determination of cadmium and copper in water and food samples by dispersive liquid – liquid microextraction combined with UV – vis spectrophotometry. *Microchemical Journal journal, 97*: 249–254.

Wybraniec, S., Jerz, G., Gebers, N., & Winterhalter, P. (2010). Ion-pair high-speed countercurrent chromatography in fractionation of a high-molecular weight variation of acyl-oligosaccharide linked betacyanins from purple bracts of *Bougainvillea glabra*. *Journal of Chromatography. B, Analytical Technologies in the Biomedical and Life Sciences, 878*(5–6): 538–50.

Wybraniec, S., Stalica, P., Jerz, G., Klose, B., Gebers, N., Winterhalter, P., Sporna, A., Szaleniec, M., & Mizrahi, Y. (2009). Separation of polar betalain pigments from cacti fruits of *Hylocereus polyrhizus* by ion-pair high-speed countercurrent chromatography. *Journal of Chromatography A, 1216*(41**)**: 6890–6899. doi: 10.1016/j.chroma.2009.08.035

Xu, Q., Shen, Y., Wang, H., Zhang, N., Xu, S., & Zhang, L. (2013). Application of response surface methodology to optimise extraction of flavonoids from *fructus sophorae*. *Food Chemistry, 138(*4): 2122–2129. doi: 10.1016/jfoodchem2012.11.099

Zhang, H., Low, W. P., & Lee, H. K. (2012). Evaluation of sulfonated graphene sheets as sorbent for micro - solid - phase extraction combined with gas chromatography – mass spectrometry. *Journal of Chromatography A, 1233*: 16–21.

Zheng, Y., & Chow, A. H. L. (2009). Production and characterization of a spray-dried complex Spray-dried hydroxypropyl-β-cyclodextrin/quercetin complex. *Drug Development And Industrial Pharmacy, 35*(6): 727-734. doi: 10.1080/03639040802526805

Zhu, Q. Y., Zhang, Q. Y., & Cao, J. (2016). Cyclodextrin - assisted liquid - solid extraction for determination of the composition of jujube fruit using ultrahigh performance liquid chromatography with electrochemical detection and quadrupole time-of- flight tandem mass spectrometry. *Food Chemistry, 213*(2016): 485–493.

Zhu, Q. yao, Zhang, Q. yun, Cao, J., Cao, W., Xu, J. jing, & Peng, L. qing. (2016). Cyclodextrin-assisted liquid-solid extraction for determination of the composition of jujube fruit using ultrahigh performance liquid chromatography with electrochemical detection and quadrupole time-of-flight tandem mass spectrometry. *Food Chemistry, 213*: 485–493.

Zularisam, A. W., Ismail, A. F., Salim, M. R., Sakinah, M., & Matsuura, T. (2009). Application of coagulation – ultrafiltration hybrid process for drinking water treatment : Optimization of operating conditions using experimental design. *Separation and Purification Technology, 65*: 193–210.