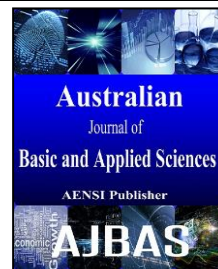




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Effects of Different Extraction Conditions on The Production of Anthraquinone

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ABSTRACT

Natural dyes have been used for the coloring of textiles since pre-historic times. Nowadays, there is an increased of interest in natural dyes as the replacement of synthetic dyes due to general environmental awareness and the increase of public interest in natural products. *Morinda citrifolia* (mengkudu) was used as the source of natural dye in this study. The extracted compound from the roots of *Morinda citrifolia* is known as anthraquinone (alizarin) that gives a red color for potential textile application. This study was performed to investigate the effects of solid liquid ratio (SLR) (1:100 to 5:100), extraction time (up to 10 hours), and pH (1 to 11) on the concentration of anthraquinone. The anthraquinone extract was analyzed by using a UV-Vis spectrophotometer. The best condition to extract anthraquinone from *Morinda citrifolia* roots was at 1:400, 2 hours, and pH 7 of SLR, extraction times, and pH, respectively. The study proved that *Morinda citrifolia* can produce a natural dye that has a strong color which can be used in textile industries.

INTRODUCTION

Various industries like textiles, rubber, paper, plastics, leather, cosmetics, food, and mineral processing industries use dyes in order to color their products. Dyes in textile industries has improved the quality of human lifestyle to a certain extent. There are over 100,000 available dyes and about 7×10^5 tons of dyestuff manufactured yearly (Koti and College, 2012). Dyes can be described as colored substances which have a similarity to the substrate to which they are applied. It can be divided into two groups, which are synthetic dyes and natural dyes. Traditionally, all dyes used in textiles were from natural origin until the revelation of first synthetic dye mauviene (basic dye) in 1856. Since then, the synthetic dye industry has developed at an elevated rate and almost wiped out the use of natural dyes across the world (Hooda and Rangi, 2015). Due to its low production cost, brighter colors, better resistance towards environmental effects, and easy-to-apply factor, the discovery of synthetic dyes has overwhelmed the role of natural dyes in the society. Synthetic dyes however, are mostly toxic and carcinogenic. Moreover, this dye has become one of the key sources of grave water pollution as a consequence of the hasty development of the textile industries. The release of colorant effluent of synthetic dyes has initiated a fear on the human health and marine survives. Inefficiency in delivering these dyes onto textile fibers can cause the colorants that contain harmful substances being released together with the effluents (Wan Ngah *et al.*, 2011). While, further disposal of the synthetic dyes from the industries into the environment causes a very serious damage, because they may significantly affect the photosynthetic activity of hydrophytes by reducing light penetration and they also may be toxic to some aquatic organisms due to their recalcitrant nature (Koti and College, 2012).

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Nowadays, curiosity in natural dyes has noticeably increased as a result of the environmental safety concerns surrounding the manufacture and use of synthetic dyes. Natural dyes can be obtained from natural sources which are plants, animals, and minerals. Across the world, variety of plant sources are used for natural dyes. It is reported that about 300 plant species have been identified in India as traditional dye sources. Any plants ranging from unwanted weeds to cultivated plants can have the possibility to act as a natural dye (Hooda and Rangi, 2015). The examples of natural dyes that can be obtained from plants are beetroot, henna leaves, *Morinda citrifolia* (mengkudu), *Curcuma longa* (turmeric), *Carthamus tinctorius L* (safflower), pomegranate rind, bark of *Acacia decurrens* (black wattle), flowers of *Tagetes erecta* (Mexican marigold), *Mirabilis jalapa* (four o'clock), and *Celosia cristata* (cockscomb) (Sinha *et al.*, 2013). Different parts of plant such as seeds, flowers, stems, leaves, barks, and roots can be used to extract colors (Hooda and Rangi, 2015), where the part of the plants chosen in this study was roots of *Morinda citrifolia*.

Morinda citrifolia is the scientific name of the commercially known plant Noni. The name *Morinda citrifolia* is also denoting to the botanical name that is originally derived from the two Latin words *morus* (ascribed to mulberry) and *indicus* (ascribed to Indian). It belongs to the Rubiaceae family. In Hawaii *Morinda citrifolia* was called as Noni, while in India it was called as Indian mulberry and nuna, or ach. Malaysians call it as mengkudu and in Southeast Asia it is called nhaut, while in the Caribbean, it is called the painkiller bush or cheese fruit (Abou Assi *et al.*, 2015). *Morinda citrifolia* is acknowledged to own high concentration of anthraquinones in their roots which is well-known to be commercially important.

Anthraquinone is identified as an essential group of natural product found in bacteria, fungi, lichens, and plants (Siva *et al.*, 2012). The elementary colorant, found in the roots of *Morinda citrifolia*, is alizarin, a red-colored anthraquinone that is used as a customary dye for decades (Bhakta and Siva, 2012). Major compounds of anthraquinone that is found in *Morinda citrifolia* roots are known as alizarin and morindone. The color obtained from *Morinda citrifolia* varies depending on the types of mordant used. For example, an aluminium mordant gives a red color (Tontrong *et al.*, 2012). Various extraction methods like pressurized hot water, solvent extraction, ultrasonic assisted extraction (UAE), and microwave assisted extraction (MAE) have been conducted to extract anthraquinone from *Morinda* in previous studies. For example, Bhu and Saiki (2003) conducted solvent extraction and aqueous extraction to extract anthraquinone, while Aobchey *et al.* (2002) only used Soxhlet extraction method in their study. Subcritical water extraction and solvent extraction was conducted by Shotipruk *et al.* (2004) where Hemwimon *et al.* (2007) conducted three methods (solvent extraction, UAE, and MAE) to compare which method can extract the highest concentration of anthraquinone.

In this work, simple solvent extraction was investigated to extract anthraquinone from *Morinda citrifolia* roots because this method is the most convenient and widely used method. It is well-known as a safe and easy to perform method compared to other methods. This method contains two phases during the extraction which are liquid (solvent) and solid (plant matrix) phases. The process has two stages that are (1) swelling and hydrating of plant matrix, and (2) mass transfer of solute from plant materials to bulk solvent by diffusion and osmotic pressure (Chua, 2013). The quantity of analytes extracted from different matrices depends on the type of matrix, conditions, and techniques of extraction. There are many factors such as types of solvent, extraction time, pH, and SLR that might significantly influence the extraction efficiency. The positive or negative role of each factor in the mass transfer of the process is not always obvious. In addition, the chemical characteristics of the solvent and the dissimilar structure and composition of the natural products that ensure each material or solvent system demonstrates different behavior also cannot be predicted (Radojković *et al.*, 2012). Thus, this study was conducted to confirm the effects of SLR, extraction time, and pH on the amount of anthraquinone from *Morinda citrifolia* roots.

MATERIALS AND METHOD

Material:

Morinda citrifolia are grown locally in Malaysia and the roots of the *Morinda citrifolia* was taken from Pusat Perusahaan Kg. Soi, Kuantan, Pahang. Distilled water was used to wash the samples (Baque *et al.*, 2012). The samples were separated before being chopped into pieces afterwards (Zin *et al.*, 2002). Alizarin, which is also known as 1, 2-dihydroxyanthraquinone, was used as reference. All chemicals were purchased from Sigma Sdn Bhd and used as received or otherwise stated.

Plant material preparation:

The roots of *Morinda citrifolia* were collected in fresh condition (Brist *et al.*, 2014). It was harvested, washed, and oven-dried at 50 °C for 2 days. By using mortar and pestle, the dried sample was ground to an average size of 0.2 mm in diameter to increase surface area (Hemwimol *et al.*, 2006). The ground samples were kept in a dry place until use (Anekanukul *et al.*, 2007)

Process of solvents extraction:

Acetone was used as a solvent in this study. In a beaker, 2 g of the roots of *Morinda citrifolia* with 200 ml of 80 % (v/v) of acetone were mixed. SLR values were varied from 1:100 to 5:100 and the extraction was carried out for 10 h (Bhu and Saiki, 2003). This extraction was performed at room temperature (25 °C) (Hemwimon *et al.*, 2007) with stirring. The pH values were varied from 1 to 11. Acetic acid was used to maintain pH in the range of 1–7, while sodium hydroxide was used to maintain pH in the range of 8–12. The extract was filtered through a filter paper (0.45µm Whatman, no.1, USA) to remove all unextractable matters, including cellular materials and other constitutions that were insoluble in the extraction solvent. The concentration of anthraquinones were measured by using UV-Vis spectrophotometer (Baque *et al.*, 2012; Hemwimon *et al.*, 2007; Mann *et al.*, 2010). The extraction was repeated three times.

Measurement of anthraquinones concentration:

In order to measure the concentration of anthraquinone in each sample, a standard curve of alizarin solutions in acetone was used as a reference (Hemwimol *et al.*, 2006). The standard curve was prepared by plotting the blank (acetone) corrected 435 nm reading for each standard (Alizarin) versus its concentration in g/L (Baque *et al.*, 2012). The range of alizarin used were 0 to 1.0 g/L and the final volume in each test tube was 5 mL (Noor Suzana, 2012). UV-Vis spectrophotometer (Hitachi U-1800) was used to determine the concentration of anthraquinone in the samples. By measuring the absorbance at 435 nm, the concentrations of anthraquinones extracts were analyzed following the spectroscopic method by Hemwimol *et al.* (2006).

RESULTS AND DISCUSSION**Effect of SLR on the production of anthraquinone:**

In order to study the effect of SLR on the anthraquinone concentration, 1:100 to 5:100 of ratio were tested at room temperature (25°C) for 10 hours of extraction time. Acetone was used as the solvent because acetone were the best solvent used to extract anthraquinone from *Morinda citrifolia* roots based on the previous study (Hemwimol *et al.*, 2006). Tontrong *et al.* (2012) supported the choice of solvent as it also found that acetone extract of *Morinda citrifolia* roots provided the highest absorbance at the maximum absorption wavelength of 435 nm. The addition of a certain amount of water in acetone contributes to the creation of a moderately polar medium that ensures the extraction of anthraquinone and thus improves the overall extracting efficiency. This was because, acetone was a low polar solvent while water was a strong polar solvent so that they can be blended with each other in any proportion. With the addition of water to acetone, the polarity of complex solvent will increase continuously. So the acquired ratio of more polar anthraquinone compounds in *Morinda citrifolia* extract increases with increasing water content agreeing to “like dissolves like” principle. Another possible reason for the increased efficiency with the presence of some water might be due to the increase in swelling of plant material by water, which increases the contact surface area between the plant matrix and the solvent (Tan *et al.*, 2013). A moderately polar solvent of 80% acetone (v/v) was chosen for the determination of various effects of the anthraquinone extract from *Morinda citrifolia* (Tontrong *et al.*, 2012). While the range of SLR was taken from the previous study which was Hemwimon *et al.* (2007) and Pongnaravane *et al.* (2006).

Figure 1 shows that the highest anthraquinone concentration achieved was 0.1081 g/L which was obtained from the curve of 1:400 SLR. Meanwhile, the lowest anthraquinone concentration achieved was 0.0779 g/L resulted from 1:100 SLR. Anthraquinone content increased generally when the SLR was increased from 1:100 to 4:100 and then decreased at 5:100. According to Nyamien *et al.* (2013), dissolution of bioactive components into the solvent is a physical process. When the amount of *Morinda citrifolia* roots increases, the chance of anthraquinone coming into contact with the acetone increases, which leads to higher leaching-out rates. On the other hand, for SLR value exceeding 4:100, the content of anthraquinone decreases mainly due to the saturation phenomenon. In fact, when the solvent is saturated with bioactive components, molecular diffusion stops and the extraction process rate remains the same. The differences observed in anthraquinone content is related to the amount of sample used and the solubility of anthraquinone in the proportion of the solvent available (Nyamien *et al.*, 2013).

This study has shown that SLR has a positive effect on the amount of anthraquinone extracted. If the SLR is too low, anthraquinone in raw material cannot be fully extracted. If the SLR is too high, this will cause a high amount of materials wasted. This might be due to the reason that *Morinda citrifolia* roots could be excessively saturated with concentrated solvent and contributed to the waste of product collection (Jiang *et al.*, 2014). An ideal SLR should be selected to avoid waste of solid and bulky handling in the subsequent processes for commercial application (Yinshi *et al.*, 2011).

Ahmad *et al.* (2015) reported that the productivity of extraction improved with higher SLR where the mass transfer rate was enhanced by producing a concentration difference between inside of the plant cells and the outside solvent. Liquid circulation such as stirring also improved the extraction by allowing greater diffusion of solvent into the sample matrix. Khoddami *et al.* (2013) recommended to determine the best ratio of SLR so that

solvent input and saturation effects of solvent by anthraquinone were minimized and increased diffusivities of the plant materials into the solution. Also, the interactions of the extracted compounds with the solvent could have modified the activity coefficients and thus the solubility of the compounds (Radojković *et al.*, 2012).

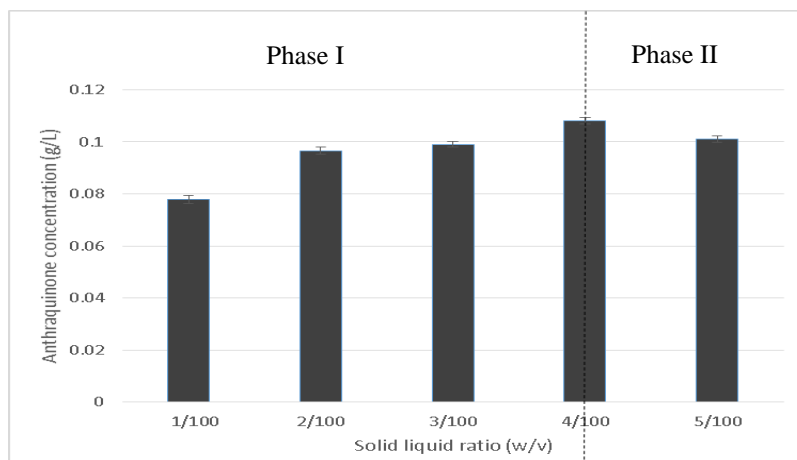


Fig. 1: Effect of anthraquinone concentration on SLR

Effect of extraction time on the production of anthraquinone:

Traditionally, higher extraction yield involves a longer extraction time (Yinshi *et al.*, 2011). To investigate the influence of extraction time on the anthraquinone concentration from *Morinda citrifolia* roots, the SLR used was 1:400 while the extraction time used was from 0 to 10 hours respectively. As illustrated in Figure 2, the anthraquinone concentration increased with extraction time. It could be due to swelling and hydration of *Morinda citrifolia* roots that it disturbs and diffuses the solvent (acetone) into the plant matrix (Mansour *et al.*, 2016). However, beyond the most favorable condition of extraction time, the anthraquinone concentration started to decrease. The rate of anthraquinones extraction was high during the first two hours (0.1090 g/L), and then decreased considerably thereafter. It was due to the large difference between the initial anthraquinones concentration of the extraction solvent (acetone) and its solubility. Another reason for the initial high rate could be that anthraquinones located at the outside region were more reachable than that in the inner part in which the plant tissues were more intact. The extraction from the outer part is attributed to external mass transfer, which in this case was convective because the liquid motion was allowed as an effect of stirring. At a later time, anthraquinones from the inner part of the root particles diffuse through the pores of the root materials, resulting in a much slower extraction rate (Hemwimol *et al.*, 2006).

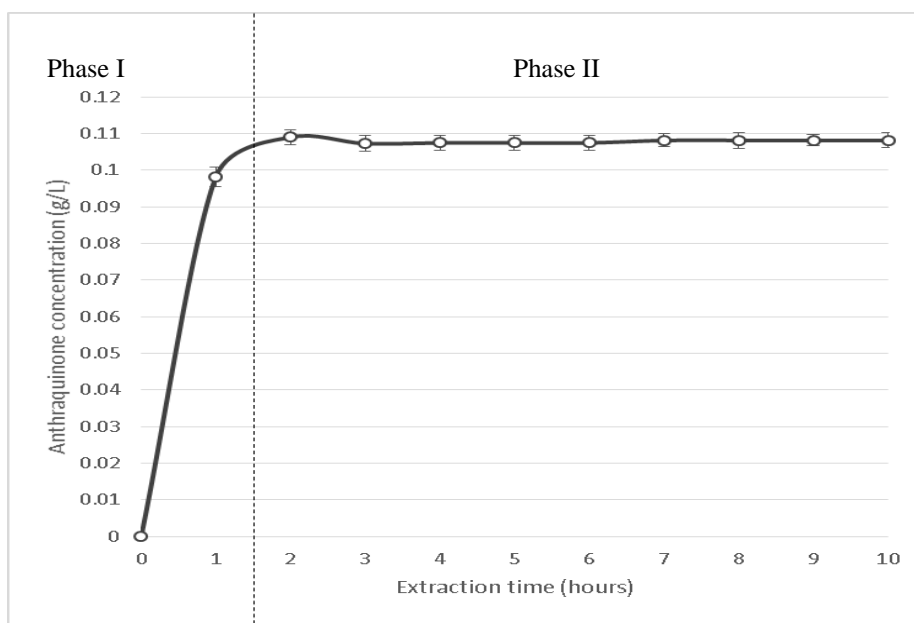


Fig. 2: Effect of anthraquinone concentration on extraction times

The result was found to be similar with previous study by Zhang *et al.* (2009), which reported that the extraction rate increased with longer extraction time because higher availability of the acetone to extract the anthraquinone. However, further increment of extraction time beyond the favorable time resulted in decreased anthraquinone concentration. These phenomena also could be well explained by the Fick's second law of diffusion in which a final equilibrium between the solute concentrations in the solid matrix (plant matrix) and in the bulk solution (solvent) might be reached after a certain time, leading to deceleration in the extraction yield. Moreover, prolonged extraction time increases the chance of decomposition and oxidation of anthraquinone due to the extensive contact to unfavorable environmental factors. On the other hand, from the industrial point of view, the increased extraction time is time consuming, uneconomical, and also a potential loss of solvent by vaporization which directly affects the loss of SLR of extraction (Tan *et al.*, 2013). Khoddami *et al.* (2013) suggested that increasing the extraction time promotes analyte solubility. However, plant phenolics like anthraquinone are mostly degraded or undergo undesirable reactions such as enzymatic oxidation.

There were many studies investigated the influence of extraction times towards their product from different raw materials. Xiaowei *et al.* (2013) discussed the extraction of procyanidins from *Larix gmelinii* bark, where a significant increase in extraction efficiency was obtained after soaking the bark and left for three hours. However, longer extraction time did not increase the extraction efficiency. The same result was obtained in this work where the solvent must have access to the cellular structure to extract anthraquinone. An intact cell structure constrains accessibility of the solvent to anthraquinone. The increase in anthraquinone extraction efficiency after soaking the *Morinda citrifolia* roots in the solvent was possibly because of the increased diffusion of the solvent into the cellular structure, allowing enhanced solubilization of anthraquinone. Meanwhile, Yinshi *et al.* (2011) suggested that as the diffusion front moves towards the interior of the tissues, the diffusion area is reduced, the diffusion distance is increased, and the diffusion rate is decreased accordingly.

Effect of pH on the production of anthraquinone:

This study also shows an interest in the effects of pH on the anthraquinone extract from *Morinda citrifolia* roots. 2 hours and 1:400 of extraction times and SLR was performed to investigate the influences of pH on anthraquinone extract respectively. The effect of pH on the color value of the dye from *Morinda citrifolia* roots was determined by recording the UV-vis spectrophotometer of the dye solution at different pH values ranging from pH 1 to 11 (Arora and Rastogi, 2012). According to Thorpe *et al.* (1887), the color given by *Morinda citrifolia* roots ranges from a reddish yellow through pink and various shades of red to a dark brown-red. The tint primarily depends on the age of the root and the proportion of root-bark to root-stem. The root-bark gives the best reds and the dye in the woody part of the root is yellow. Hence, when the woody part dominates over the bark, the resulting dye is reddish yellow. Figure 3 shows the variation of color obtained from different range of pH. The extraction using pH 1 and 3 yielded yellow solutions, while the remaining tested pH values resulted in brick red color. Depending on the strength of alkali or acid used, the color of anthraquinone extract (as alizarin) can vary from scarlet to pink to red with a bluish tint. A strong alkali creates a violet-blue color, whereas a diluted alkali create a violet-red color. On the other hand, a strong acid yields a yellowish red extract. The alcohol and aqueous solutions produce a rose color, while the ethereal solvent gives golden-yellow extract (Santis and Moresi, 2007). However, in this study, the colors obtained were only yellow and brick red. Bluish tint was not obtained might be because the basic pH used was not strong enough.



Fig. 3: The color obtained at different range of pH

As shown in Figure 4, the highest concentration of anthraquinone was obtained at pH 7 (0.1090 g/L) while the lowest was at pH 11 with 0.0493 g/L. It is clear that at 25 °C the anthraquinone extract decreased with an increase in pH from neutral to alkaline (Mansour *et al.*, 2016). Arora and Rastogi (2012) revealed that under neutral to alkaline conditions, the color of the solution deepens and it is supported by the fact that some natural anthraquinones exhibit this behavior at different pH values. The anthraquinones would display a characteristic red color in a weak basic solution. The decrease in concentration of anthraquinone may be due to the net charge effect. Changing of the pH probably alters the charge of the anthraquinones (Zhijian *et al.*, 2011). The change in

color may be attributed to changes in conjugation of the extracted compounds at different pH values (Mishra *et al.*, 2012). As the aim of this work was to obtain a reddish brown color with highest concentration possible, neutral pH 7 was chosen for subsequent experimental works.

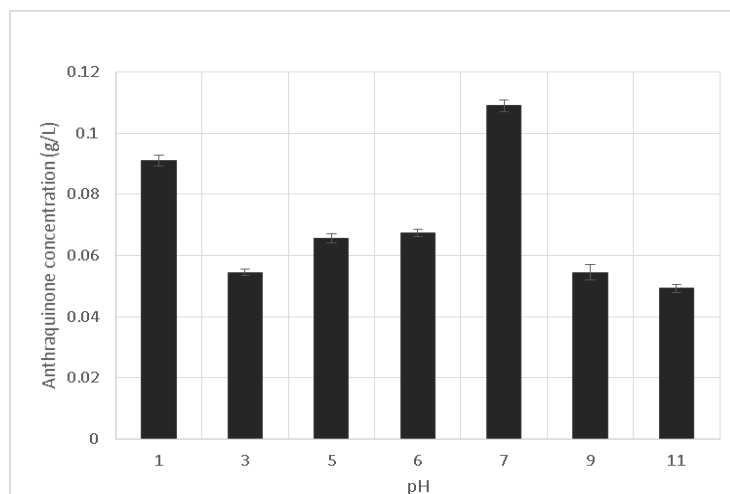


Fig. 4: Effect of anthraquinone concentration on pH

Conclusion:

The effects of SLR, extraction time, and pH on the extraction of anthraquinone from *Morinda citrifolia* roots were investigated. The highest anthraquinone concentration was obtained at 1:400, two hours, and pH 7 of SLR, extraction times, and pH, respectively. This condition yielded a reddish brown color solution which is suitable for coloring almost all types of natural fibres in the textile application. They also can be used to dye some synthetic fibres. Apart of their application in textile, the reddish brown color solution might be use in handicraft items and toys, and in leather processing.

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