The Extraction of Natural Dyes from Henna Leaves (Lawsonia Inermis L.) by Ultrasound-assisted Method

IOP Conf. Series: Materials Science and Engineering **543** (2019) 012082 doi:10.1088/1757-899X/543/1/012082

The Extraction of Natural Dyes from Henna Leaves (Lawsonia Inermis L.) by Ultrasound-assisted Method

L Qadariyah, N Azizah, A Q Syafa'atullah, D S Bhuana, M Mahfud
Chemical Engineering Department, FTI-ITS, Keputih, Sukolilo, Surabaya, Jawa Timur 60111, Indonesia

Email: lailatul_2008@yahoo.com

Abstract. One alternative to reduce the effect of water pollution from the textile industry is to replace the use of synthetic dye with the natural one. Henna leaves are one of the natural dye sources which has lawsone, a unique substance that produces orange colour. Some commonly used conventional extractions are soxhletation and maceration. One extraction method that is tried to be developed in this study is the Ultrasound-assisted Extraction. The effects of feed ratio to solvent, time, temperature and solvent types were investigated and discussed in this study. The highest yield from the extraction of Henna leaves was found to be 17.96% which was achieved at pH 1 with the extraction time of 10 minutes, feed/solvent ratio of 0.02, and aquadest solvent. Based on the both qualitative and quantitative analysis of the extracted colour, the ultrasound assisted method was found to be more effective and efficient than the soxhletation method.

1. Introduction

Textile demand increases with increasing appetite for colour. Most of the colouring process for the textile industries use synthetic dyes. The advantages of synthetic dyes include wider colour variations, more practical, easier to use, as well as more economical. However, synthetic dyes also have disadvantages, one of which is the waste generated after the staining process. The synthetic dye waste is harmful to aquatic ecosystems because it may contain chemicals or even heavy metals [1]. In September 2012, there was a terrible pollution in Pekalongan. The laboratory test samples from the river that passes through the Karangjompo Village, Tirto, Pekalongan showed that it contained high ammonia. It can cause water hardness that can severely affect health. Therefore, the need for alternative method to reduce the impact of textile industry waste has arisen, and replacing synthetic dye with the natural one is seems to be one of the best approaches [2]. Henna leaves contain lawsone dye that can be extracted as yellow and orange crystals. This dye can be used as a dye for skin, nail, hair, silk, and wool. Extracting the lawsone from the Henna leaves can be carried out by the mass transfer of dyestuff from solid to liquid phase (solvent) [3]. This method of extraction is commonly called as solid-liquid extraction (leaching). Some conventional methods that are commonly used for extraction include maceration, soxhletation, and reflux methods. These conventional methods have several weaknesses, such as abundant requirement of solvent, lower in yield and longer extraction time. It is necessary to using an extraction technique that safe for chemical compounds and solvent with high efficient energy such as ultrasound extraction (De Oliveira et al., 2018). Ultrasound assisted extraction is simple method and eco-friendly than conventional methods [4]. Therefore, in this study the extraction method with ultrasonic waves or Ultrasound Assisted Extraction (UAE) is to be developed in order to optimize the extraction process.

2. Methods

2.1. Material and Equipment

The materials used in this experiment include Henna in powder form, aquadest, ethanol, CH_3COOH, and NaOH. The Henna was imported from Saudi Arabia. The equipments used in this experiment are as follow: three neck flask with flatbase, beaker glass, analytical balance, pH meter, thermometer, water jet pump, filter paper and ultrasonic cleaner

2.2. Optimation of extraction condition

Various trials were carried out for the extraction of natural dye from Henna leaves in aquadest solutions, ethanol 20%, ethanol 40%, ethanol 60%, ethanol 80%, and ethanol 96%. The ratios of feed to solvent were 0.01, 0.02, 0.04, 0.06, 0.08, and 0.1. The extractions were carried out at various temperature and time durations as well, i.e. at 30^{o}C, 40^{o}C, 50^{o}C, and 60^{o}C for 5 min, 10 min, 20 min, 30 min, and 60 min. As far as to obtain the optimum pH, the extractions were carried out at pH 1, pH 4, pH 8, and pH 10.

2.3. Ultrasound assisted extraction

100 ml of solvent with Henna powder in it was placed in the ultrasonic bath and was extracted by the ultrasound for 10 minutes. The extraction time was set up and the frequency of the ultrasonic bath was set up at 40 kHz. Then solvent was then separated from the extract using water jet pump and a filter paper on top of it. The solid was separated and sticked on the filter paper. The solvent became clear from solid and the color of the solvent was orange. CH_3COOH or NaOH was then added into it to control the pH of this extract solution. The lawsone contained in the solvent made the solvent turn orange and was separated with the solvent by heating. It was heated at 80^{o}C using waterbath until only the dye extract left.

2.4. Soxhelation

500 ml of aquadest and ethanol 60% were prepared and placed in a one neck flask roundbed flat of 1000 ml. Ten grams of Henna powder was prepared and put in the filter paper which was placed in the soxhlet accordingly. It was heated at 80^{o}C for ethanol 60 % and at 110^{o}C for aquadest. The extraction occured until the solvent in the soxhlet become clear or colorless. The soxhletation extraction was counted to be one cycle when the solvent filled in soxhlet and then turned back to the one neck flask roundbed

3. Results and discussion

3.1. Ethanol concentration effect

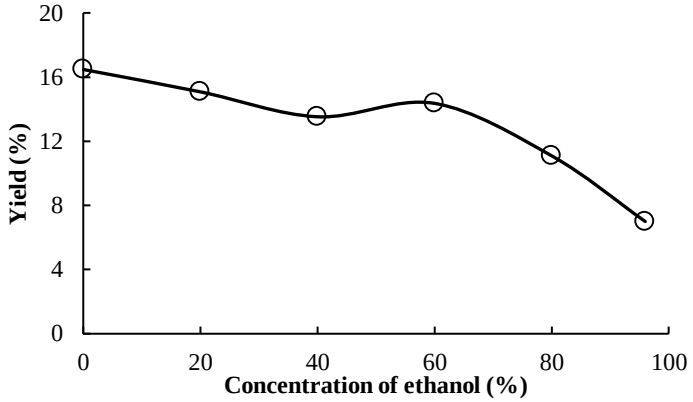


Figure 1. The effect of ethanol concentration (w/w) on the yield of henna extract (w/w) at 10 minutes and at temperature solvent 30^{o}C

The extraction with the UAE method at various concentrations of ethanol is shown in Figure 1. showed optimum results when aquadest was used as a solvent. When higher ethanol concentration was used, the obtained extract was not as abundant. However, at a concentration of 60% ethanol there was an increase in the extraction yield. In other words, 60% ethanol solvent gave higher yield compared to that with 40% ethanol solvent. it was found using 70 % ethanol as solvent give highest rate of yield extracting colorant from sorghum husk [5]. Furthermore, if the ethanol contains more water then the solvent polarity index becomes higher and increases the efficiency of the extraction process [6]. The water polarity index is 9.0 whereas ethanol is 5.2. So it can be said that water is more polar than ethanol [7]. The aquadest solvent was found to give 16.47% yield, which was higher than 60% ethanol's 14.36% yield

3.2. Effect of Extraction Time

Figure 2 shows the effect of extraction time on the yield of Henna extract (w/w) at ratio feed to solvent 0.02 and at temperature 30°C. The extraction with UAE method using 60% ethanol solvent and aquadest gave the optimum yield at the extraction time of 10 minutes. At the extraction time between 5 minutes and 10 minutes, the yield has greatly increased but after the extraction time of 10 minutes the extract yield started to decrease and finally started to stay constant from 30 minutes onwards. This point indicates that the solvent binding capacity of the extract reaches maximum (saturated). The water content of the solvent causes the swelling of the material so that the contact area between the plant matrix and the solvent becomes larger, thus, causing the extraction results to become larger accordingly [8].

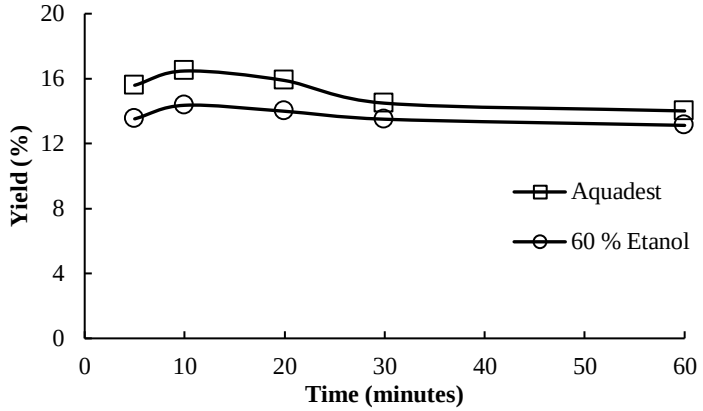


Figure 2. The effect of extraction time on the yield of henna extract (w/w) at ratio feed to solvent 0.02 and at temperature 30°C

3.3 Effect of of Ratio Feed to Solvent

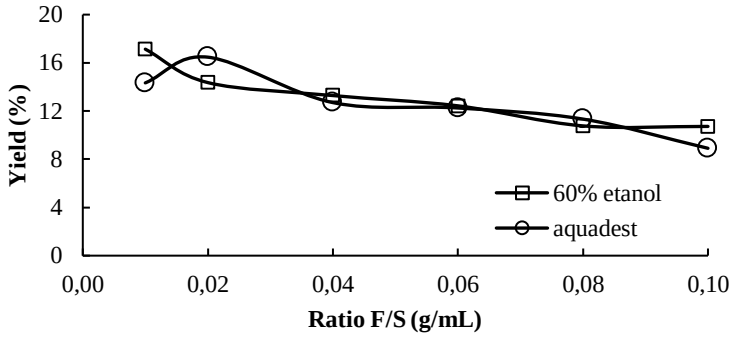


Figure 3. The effect of ratio extraction (F/S) on the yield of henna extract (w/w) at 10 min and at temperature 30°C

3.4. Effect of Temperature

The effect of temperature was investigated with a feed-solvent ratio of 0.02 and 10 minutes extraction time is shown by Figure 4. The optimum yield at room temperature was found to be 16.47% in water solvent. At higher temperatures, the yields of the resulting extracts decrease and began to rise back up at 60°C. This decrease is due to the fact that the lawsone substance in the Henna leaves is vulnerable to heat. The higher the temperature of the extraction can cause more damage in the lawsone. Thus, the optimum temperature of the solvent in Henna extraction was 30°C.

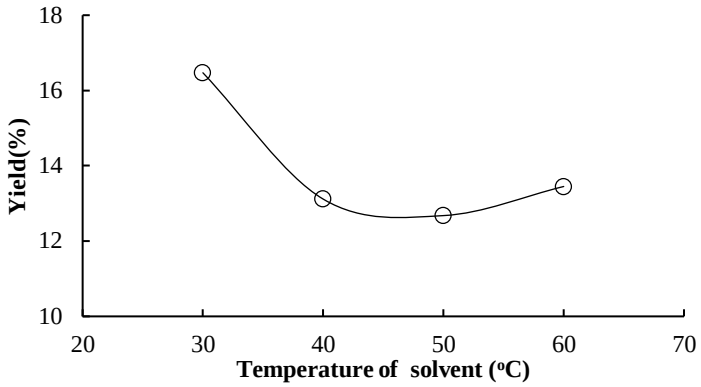
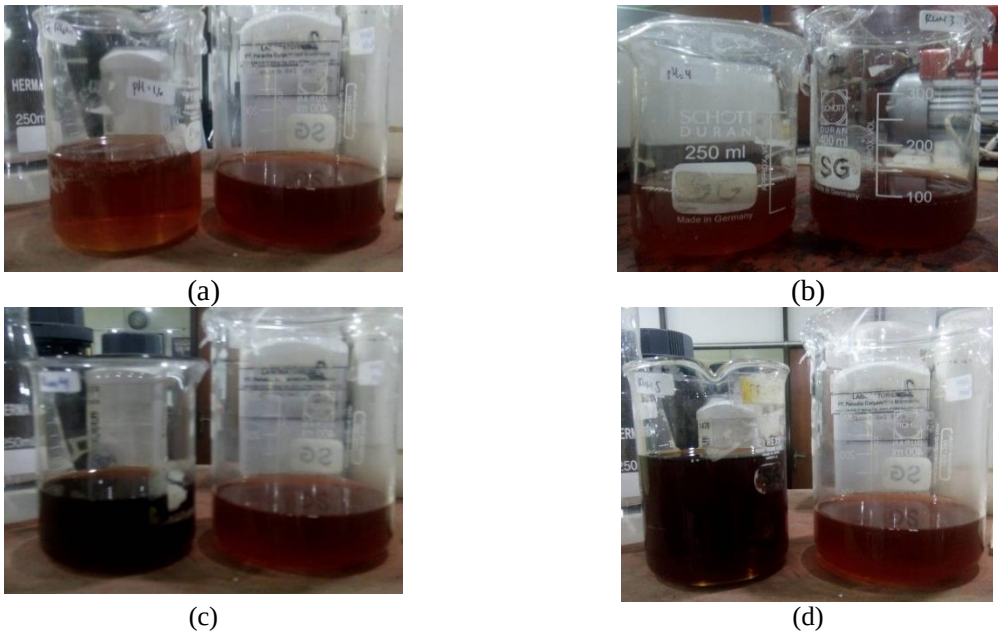


Figure 4. The Effect of temperature of solvent on the yield of henna extract (w/w) with at ratio (F/S) 0.02, time 10 min and solvent of aquadest

3.5 Effect of pH



(a) (b)

(c) (d)

Figure 5. The effect of pH in color change. The left is acidic or alkaline solution and the right is solution at original pH, (a) at pH 1.08, (b) at pH 4, (c) at pH 8, and (d) at pH 10

Figures 5 and 6 show the effect of pH on colour change and Henna extract yield. Various pH level used in this experiment was 1.09; 4.0; 6.49; 8.0; and 9.94. The pH started to be controlled after the extraction with ultrasonic. HCl was added to the extract solution to make acidic solution and NaOH to make alkaline solution, respectively. In both acidic and alkaline conditions, no significant yield changes were recorded. Rather, it only affects the colour of the extract solution. When the extract

solution was on acidic condition, the colour became fade. When the extract solution was on alkaline condition, the colour became dark.

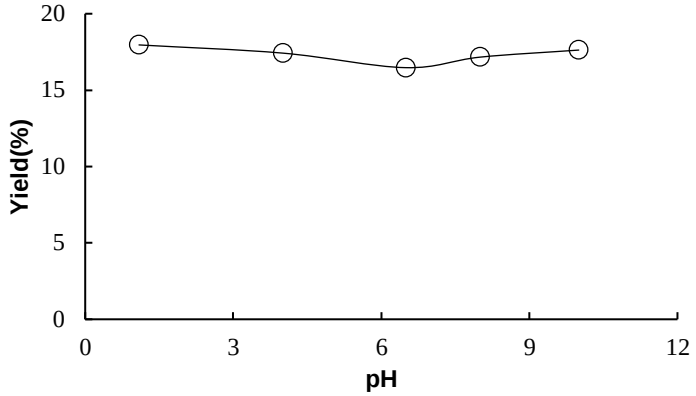


Figure 6. The effect of pH on the yield of henna extract (w/w) at ratio (F/S) 0.02, time 10 min, temperature of 30°C and solvent of aquadest

3.6 Soxhlet Extraction

Based on the experiment results, the highest yield for the UAE method with aquadest solvent was found to be 16.47% whereas 60% ethanol solvent gave 17.13% yield with 10 minutes extraction time. For soxhlet extraction method, the yield with aquadest solvent was 15.98% while 60% ethanol solvent gave 12.06% yield. The duration for soxhletation per cycle was 5-6 hours. When aquadest solvent was used, it required 10 cycles whereas 60% ethanol solvent required 12 cycles. Based on this, the soxhlet extraction method is less effective to extract dye from Henna as far as the time needed for extraction is concerned

3.7 UV-Visible Spectrophotometric Analysis of Extract Natural Dye Henna

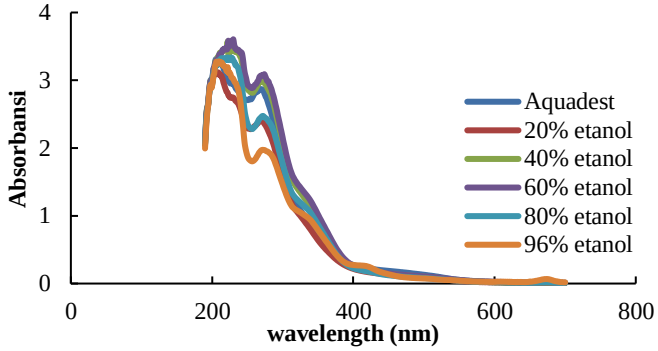


Figure 7. UV-Visible absorption spectrum in liquid extract sample with 10 min extraction time and ratio to solvent 0.02

Figures 7 and 8 show UV-Visible absorption spectrum at liquid and powder sample. The absorbance test was carried out to find out what kind of dye were present in the extracted material. The absorbance test was performed by using Thermo Scientific UV-Visible Spectrophotometry by comparing the graph of the relationship between absorbance and wavelength (nm). Aquadest and 60% ethanol solution were used as solvents for the Henna leaf extract. The light absorbed by a substance is different from the light captured by the human eye. The maximum absorption of the coloured solution occurs in the opposite colour areas. Based on the results of the UV-Vis spectrophotometric test that the maximum absorbance level on 60% ethanol solvent was 3.6 in wavelength 230 nm and 3.09 at 274 nm wavelength for the result of Henna leaf extract in liquid form, while absorbance level maximum on a powder form of 3.03 at wavelengths of 206 nm and 2 at a wavelength of 270 nm. In the previous

research, it was explained that Lawson had the followings absorbance in ethanol solvents: 246, 277, and 333 nm, respectively. In wavelength of 246 nm shows that the presence of intramolecular hydrogen bonds from benzoquinone, in wavelength of 277 nm indicates that the electron delocalization of benzoquinone becomes a quinone ring, whereas in wavelength of 333 nm indicates that the presence of naphthoquinone in the form of hydrogen bonds with carbonyl [9].

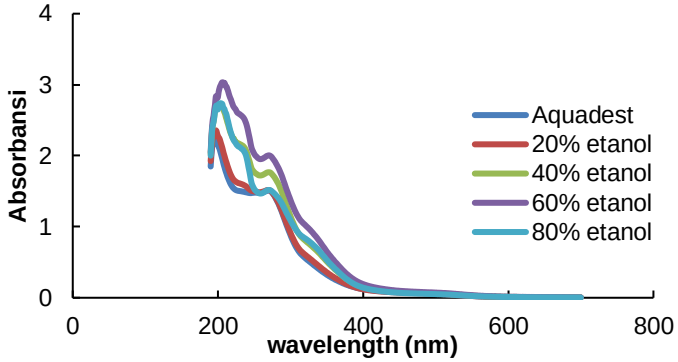


Figure 8. UV-Visible absorption spectrum in powder extract sample with 10 min extraction time and ratio to solvent 0.02

4. Conclusion

The Henna extract yield were strongly influenced by concentration of solvent, ratio of feed to solvent, pH and temperature, while time extraction has no significant effect on Henna extract yield. The highest yield was 16.47% for the UAE method with aquadest solvent whereas for 60% ethanol solvent gave 17.13% yield with 10 minutes extraction time and feed to solvent ratio of 0.02, temperature of 30^{o}C. The soxhlet extraction method is less effective to extract dye from Henna as far as the time needed for extraction is concerned.

Acknowledgments

This work is financially supported by the Ministry of Research, Technology, and Higher Education of the Republic of Indonesia (KEMENRISTEKDIKTI) under the scheme of Penelitian Dasar Unggulan Perguruan Tinggi (PDUPT) 2018.

References

[1] Gonzalez V, Wood R, Lee J, Taylor S and Bussemaker M J 2018 Ultrasound-enhanced hair dye application for natural dyeing formulations *Ultrason. Sonochem* **52** 294-304.

[2] Anastasia F, Karolina S, Sukmann T, Iŕen J J and Andrea E 2019 Application of natural dyes on diverse textile materials *Optik* **181** 215–19.

[3] Bhuiyan M A R, Islam A, Ali A, and Islam M N 2018 Color and chemical constitution of natural dye henna (Lawsonia inermis L) and its application in the coloration of textiles *J. Clean Prod.* **167** 14–22.

[4] Yingcheng W and Wei W 2015 Optimization of Ultrasound-assisted Extraction of PhysalisalkekengiL var francheti seed oil *Chem. Eng. Tran.s* **46** 1387–92.

[5] Yiqi Y, Lu W, Boumou M, Xiuliang H, Jakpa W and Yongying T 2018 Ultrasound-microwave assisted extraction of natural colorants from sorghum husk with different solvents *Ind. Crops. Prod.* **120** 203–13.

[6] Chandrasekhar J, Madhusudhan M C and Raghavarao K S M S 2012 Extraction of anthocyanins from red cabbage and purification using adsorption *Food Bioprod. Process.* **90** 615–23.

[7] Muhamad N, Muhmed S A, Yusoff M M and Gimbun J 2014 Influence of solvent polarity and conditions on extraction of antioxidant, flavonoids and phenolic content from Averrhoa bilimbi

J. Food Sci. Eng. **4** 255–60.

[8] Wen H, An X, Hai N, Zhen J and Jiawen W 2009 Optimised ultrasonic-assisted extraction of flavonoids from Folium eucommiae and evaluation of antioxidant activity in multi-test systems in vitro *Food. Chem.* **114** 1147–54.

[9] Chandrakalavathi T, Sudha V, Sindhuja M, Harinipriya S and Jeyalakshmi R 2018 Photosonoelectrochemical analysis of Lawsonia inermis (henna) and artificial dye used in tattoo and dye industry *J. Photochem. Photobiol. A: Chem.* **360** 44–57.