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Original Article

Adsorption of Metanil Yellow Using Flavonoid Extract from Longan Peel (*Dimocarpus longan*): Optimization of pH and Concentration

Sri Benti Etika, Desy Kurniawati *, Melindra Mulia, Rahni Darussalam, Okta Suryani, Edi Nasra, Nada frista

ABSTRACT

Water pollution can come from industrial waste containing various toxic pollutants, one of which is dye. Hazardous dyes are stable and resistant to biodegradation due to their complex aromatic structure, so dyes need to be treated. Metanil Yellow is a dye that is harmful to aquatic life and the human body. One method that can be used is adsorption with the batch method because it has the advantage of low cost and simple processing. Flavonoid extracts have high potential to be used as biosorbents because they have hydroxyl and carbonyl groups. This study aims to determine the potential of flavonoid extracts to adsorb metanil yellow dye using optimum pH and solution concentration and to determine its absorption capacity. The results showed that the optimum pH obtained was at pH 2 and the optimum solution concentration was at 150 ppm with an absorption capacity of 9.22078 mg/g.

Keywords: Adsorption, Batch, Flavonoids, Longan Peel, Metanil Yellow.

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INTRODUCTION

Water pollution due to industrialization gives rise to a big challenge for continuity of life and sustainable development. One of the causes of waste water that is possible is dyes, which worsen our condition and cause lots of disease[1]. It has been reported that about 20% of dye textile is lost during the manufacturing process of the product[2,3]. Dangerous dyes are stable and resistant to the biodegradation process because of the structure of their complex aromatics, so that dye needs to be overcome[4].

Metanil yellow (Natrium3-[(4-*Nphenylamino*) *phenylazo*] *benzene sulfonate* and sodium salt of *methanilyazodiphenylamine*) is a dye highly toxic and dangerous monoazo. Azo dyes are most dyes used worldwide, accounting for 80% of total production dye. Methanil yellow dye is a compound aromatic synthetic with functional azo group (-N = N-) and properties coloring from dye. This is especially caused by the azo group[5]. Ordinary metanil yellow is used for industry, paint, cosmetics, etc. Impact from the dye metanil yellow is not only poisonous for wild life but also nature mutagenic (causing gene mutations) and carcinogenic (causing cancer)[2], disease peel[6,7], and others.

Until the moment, various techniques have been used to remove substances of dangerous color from water and the human body, like deposition chemistry, adsorption, ion exchange, flocculation coagulation, separation membrane, and extraction fluid supercritical[8]. Adsorption is one of the most common methods used Because the process is simple and requires little cost[8,9].

A number of biosorbents are used to absorb substance color. Because own group functions like hydroxyl, carboxyl, carbonyl, and amine can form a ligand if the dye ion is still on the surface[10]. Based on existing research done related to biosorption, it shows that peel longan (*Dimorcapus longan*) can be used as a biosorbent to absorb a color like Methylene Blue[11] was obtained capacity, the absorption was 63.6943 mg/g, and Rhodamine B[12] was obtained capacity, the absorption was 29.1806 mg/g. Longan (*Dimocarpus longan*) is a family from *Sapindaceae*, also called lungan or dragon's eye[13]. Usually peel longan no use so that becomes waste but own benefit health like own activity high antioxidant with scavenge radical free[14], improve system immunity body, guard health skin, and maintain strength hair[15]. Phytochemical studies previously reported that on the peel longans contain lots of compound active alkaloids, amino acids and their derivatives, flavonoids and their derivatives, tannins, polyphenols, terpenoids, and saponins[16,17].

Flavonoids include metabolites secondary phenolics produced by plants level high[18] and many found in many stems, leaves, flowers, and fruit in something plants[19]. Structure base propane diphenyl, where the ring phenolics (ring A and ring B) are connected by rings heterocycles (C rings) are usually form piran closed. Flavonoids are classified into a number of groups, including, among others, the following: flavones, flavanones, flavonols, isoflavonoids, and anthocyanidins[20]. Compound This is known potential to reduce degenerative diseases, cancer, and urilithiasis[21]. Possible flavonoids can adsorb substance color because they contain variation pattern hydroxylation as well as formation group carbonyl by C4[22].

Use flavonoid extract for adsorption substance color. Not yet done because that researcher is interested in the study Optimization of pH and Concentration of Metanyl Yellow Dyes Flavonoid Extract from Longan peel (*Dimocarpus longan*) using method *batches*.

METHODS

Tools used in study This is glass chemistry, bottles spray, *rotary evaporator*, dropper pipette, rod stirrer,

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paper filter, pH meter (MR *Hei Standard*), *magnetic stirrer, mortar and* pestle, balance analytical (ABS 220-4), blender and grinder, *shaker* (VRN-480), and instruments used are UV-Vis and FTIR spectrophotometer.

Materials used are methanol distillation, ethyl acetate distillation, n-hexane distillation, distilled water, paper strain, solution substance color metanil yellow, 0.5 M HCl, and 0.1 M NaOH.

Sample used in research this is peel Longan (*Dimocarpus longan*). Following is procedure work on research:

Sample Preparation

Longan peel cut small, then air-dried, and after that smoothed. After that, do a preliminary test to know if compound metabolites contained secondary compounds in the sample. Preliminary tests carried out are alkaloids, steroids, terpenoids, flavonoids, and saponins.

Extraction of Flavonoids from Longan Peel (*Dimocarpus longan*)

a. Extraction

A total of 1 kg of sample has been dry mashed and carried out a maceration process using solvent methanol until negative test results for flavonoids in *the Shinoda test* carried out on samples. Solvent filtered for separated with dregs and evaporated, use a rotary *evaporator* until obtained extract thick. Then added distilled water at 45°C, then done filtering until obtained extract was watery.

b. Fractionation

Extract watery fractionated use funnel split, fractionate done several times until n-hexane layer clear and results end form negative flavonoids in *the Shinoda test*, two fractions were obtained, namely fraction aqueous and n-hexane fractions. Fraction watery is fractionated with ethyl acetate several times. Fraction ethyl acetate was put together, and then the solvent evaporated using a *rotary evaporator* until condensed flavonoids were obtained, EtOAc (crude flavonoids). After that, characterized using FTIR.

Reagents

a) Making solution Parent Methanyl Yellow ($C_{18}H_{14}N_3NaO_3S$) 1000 ppm or 1000 mg/L

As much 0.25 grams of methane yellow is dissolved with distilled water in a glass chemistry 250 mL, then insert the solution into a pumpkin measure 1000 mL, then add with distilled water until the next limit mark is homogenized.

b) Making solution standard Metanil Yellow ($C_{18}H_{14}N_3NaO_3S$)

Pipette solution parent metanil yellow 1000 ppm as 2.5 mL, 5 mL, 7.5 mL, 10 mL, and 12.5 mL for concentrations of 50, 100, 150, 200, and 250 ppm, respectively. Then insert into the pumpkin measure 50 mL, then add distilled water until boundary marks are homogenized and included in the bottle reagent.

c) Making a 0.1 M sodium hydroxide (NaOH) solution

Weigh out 0.4 grams of NaOH and add it into the glass chemistry. 50 mL later dissolved, after that entered to in pumpkin measure 100 mL, then add distilled water until boundary marks and homogenized.

d) Making a 0.5 M Hydrochloric Acid (HCl) solution

Make 0.5 M HCl, pipette 4.14 mL of 37% HCl, then insert into the pumpkin measure 100 mL. After that, add distilled water until boundary marks and homogenized.

Treatments

a) Determine λ the maximum absorption *of metanil yellow* ($C_{18}H_{14}N_3NaO_3S$).

Solution *Metanil Yellow* 50 ppm, then measured using a UV-Vis spectrophotometer with a long wave from 350 to 600 nm, and obtained the long wave maximum absorption of methanil yellow.

b) Effect of pH

Solution *methanil yellow* as much as 25 mL with concentration of 100 ppm with pH variations of 1, 2, 3, 4, 5, 6, and 7 with addition of 0.5 M HCl and 0.1 M NaOH to adjust the pH, then each pH is contacted with 0.2 grams of crude flavonoids using system *batches*. Shake *the* solution at 200 rpm for 30 minutes, filter, dissolve, and tamp the filtrate. The resulting filtrate was then measured with a concentration of metanil yellow, which is not absorbed with the UV-Vis spectrophotometer up to optimum pH.

c) Influence concentration solution

Solution *methanil yellow* as much as 25 mL with concentrations of 50, 100, 150, 200, 250 ppm at optimum pH. Then each solution was contacted with 0.2 grams of crude flavonoids with the system *batch. The solution was* then in *a shaker* with a speed 200 rpm for 30 minutes. Then filter solution it and accommodate it in the filtrate. The resulting filtrate was measured for concentration of metanil yellow, which is not absorbed with the UV-Vis spectrophotometer, and obtained the optimum concentration.

RESULTS AND DISCUSSION

Longan skin after preliminary testing compound metabolites secondary show that skin longan positive Contains proven flavonoids when filtrate skin the previous longan heated in solution methanol for 5 minutes experience change color from yellow pale to red orange after Mg-HCl was added (*Shinoda test*).

FTIR Characterization

Transform Infrared Spectrophotometer (FTIR) is a useful tool for characterizing and identifying material chemistry or determining group existing functions with a range frequency of 4000-650 cm^{-1}. FTIR results in research can be seen in Figure 1.

From the results measurement, the IR spectrum against flavonoid extract provides absorptions important, namely 3335.55 cm^{-}

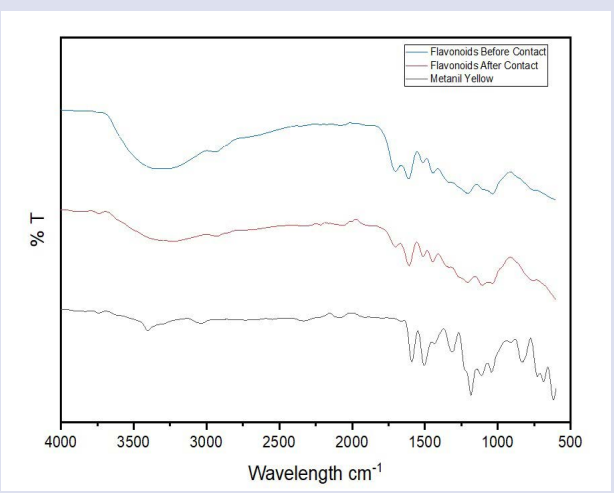

Figure 1 . Combined FTIR Spectrum.

[1], interpreted as OH stretching (Hayat et al., 2020) , This is supported strongly by vibration-bent CO alcohol in the area 1206.39 cm^{-1}. Vibration 2937.98 cm^{-1} is interpreted as aliphatic CH stretching. Other supporting characteristics exist ring aromatic indicated by absorption in the area number wave 1611.99 cm^{-1}, 1513.48 cm^{-1}, which is uptake from strain aromatic C=C ring as group typical chromophore from internal flavonoids system bond conjugated. The results obtained from FTIR are almost the same as with the previous research[23].

Characteristic substance color methanol yellow can be seen in the vibration mode sharp at 1590.20 cm^{-1} for the most certain N=N sites in identification and quantitation. Apart from that, vibration sulfate (SO_3) at 1042.27 cm^{-1} confirms the existence of methanol yellow. At vibration 3403.14 cm^{-1}, interpreted as NH. Vibration 1313.90 cm^{-1} interpreted as C=C. Because methanol yellow doesn't have a CH_3 or CH_2, the peak near 3000 cm^{-1} (3035.85 cm^{-1}) is a CH site; however, set the peak to a specific CH site still far from certain. However, it is the location that produces its peak unique CH bending (1183.94 cm^{-1}), likely big correlated with one frequency CH stretch. Peaks above 3100 cm^{-1} are too wide and often also depend on humidity, so they are not useful in a way analytical[24].

From the FTIR spectrum above, absorption has occurred between the flavonoid extract and metanil yellow, as evidenced by the change or shift of the O-H group in the FTIR spectrum of the flavonoid extract before being contacted and after being contacted with metanil yellow. The estimated mechanism or adsorption process between the flavonoid extract and metanil yellow is a hydrogen bond.

Wavelength Determination Maximum (λ max) Metanil Yellow

Determination long wave maximum aim for know area possible uptake generated as well as how many marks absorbance from solution metanil yellow uses tool UV-Vis spectrophotometer. Determination of long-wave metanil yellow using a UV-Vis spectrophotometer is carried out in the range of long waves 350-600 nm with a concentration of 50 ppm[25]. The wavelength maximum obtained is 475 nm.

Effect of Optimum pH of Metanil Yellow on Flavonoid Extract

pH of the solution dye plays a role important in the adsorption process. This is in fact a significant influence mechanism ionization or dissociation of molecule colorants, properties of surface adsorbents, and interactions electrostatic between molecule adsorbates and surfaces adsorbents[26]

The research was carried out at pH 1 to 7 to determine the optimum pH condition of the solution on absorption capacity. Effect of pH on absorption of metanil yellow can be seen in Figure 2.

The picture shows capacity absorption. The highest was at pH 2, namely 7.01299 mg/g. Absorption capacity increases at pH 2 due to the presence of H+, who dominates, so that happens style pull electrostatic with Metanil yellow anion and decrease capacity absorption occurs at pH 3 onwards matter. This is because there is rejection electrostatic between anions of substances of the color metanil yellow with the surface adsorbent, and charge negative on the surface adsorbent decreases with increasing pH[27].

Influence Concentration Metanil yellow against Flavonoid Extract

Determination concentration solution aim For know about research are 50, 100, 150, 200, and 250 ppm at pH 2 (optimum pH). From the results of the study, the optimum absorption concentration of metanil yellow by flavonoid extract is 150 ppm, with capacity absorption amounting to

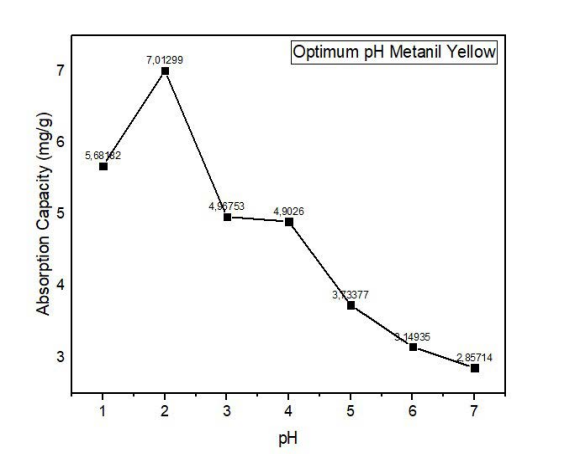

Figure 2 . pH graph of Metanil yellow against flavonoid extract.

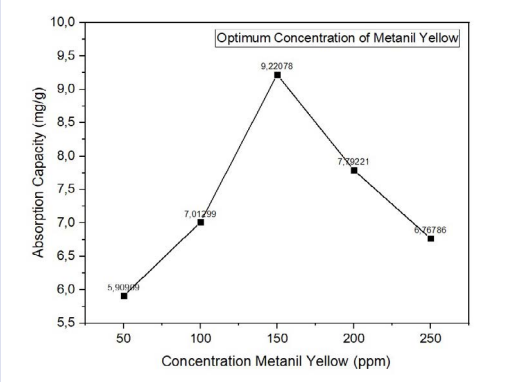

Figure 3. Graph of Metanil yellow concentration on flavonoids.

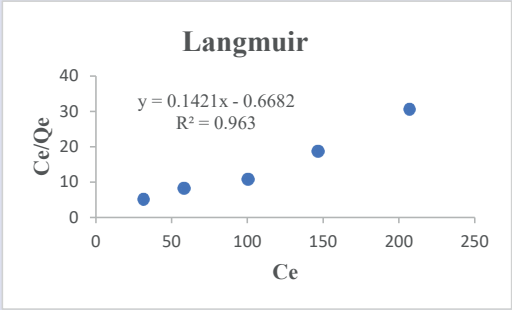

Figure 4. Langmuir isotherm of Metanil yellow adsorption using flavonoid extract.

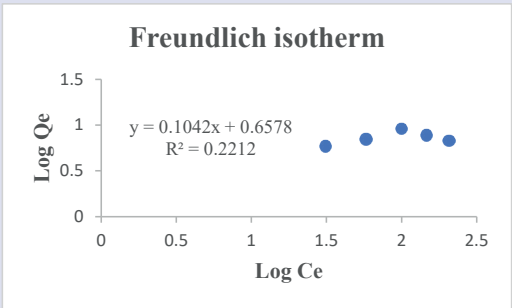

Figure 5. Freundlich isotherm of Metanil yellow adsorption using flavonoid extract.

9.22078 mg/g. Influence concentration on absorption of metanil yellow can be seen in Figure 3.

Adsorption capacity increases along with increasing concentration substance color because of increasing interaction electrostatic between molecule metanil yellow and active sites on the surface of flavonoids. Besides that enhancement, concentration also results in a big boost in strength between both of them. Declined capacity adsorption after reaching optimum is caused by saturation-side active adsorbent because of the number of active sites. Biosorbents are not comparable with amounts of adsorbate[26,28].

Isotherm Adsorption

Isotherm Langmuir and isotherm freundlich become decider mechanism adsorption used with method know price coefficient the highest determinant (R). between both of them. Determination isotherm This can be determined with the method make curve straight line equilibrium. The purpose of determination of adsorption isotherms is so you can know changes that occur in concentration adsorbate at the moment happen adsorption[29]. Equation curve isotherm Langmuir and isotherm freundlich can be seen in the picture following.

Based on the second picture, it shows that adsorption methanol yellow uses more flavonoids tend to be isotherms (Langmuir). This is because the coefficient determinant (R) is bigger than isotherm freundlich, which is 0.963. That matter shows that the methanyl yellow ion is adsorbed on the surface of its flavonoid properties homogeneous with a fixed number of sites to form a monolayer layer[29,30,31].

CONCLUSION

Based on research that has been done, it can be concluded that the optimum pH is obtained, namely at pH 2, and the concentration optimum solution is at 150 ppm with a capacity absorption of 9.22078 mg/g.

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DISCLOSURE STATEMENT

The authors have declared that no competing interests exist.

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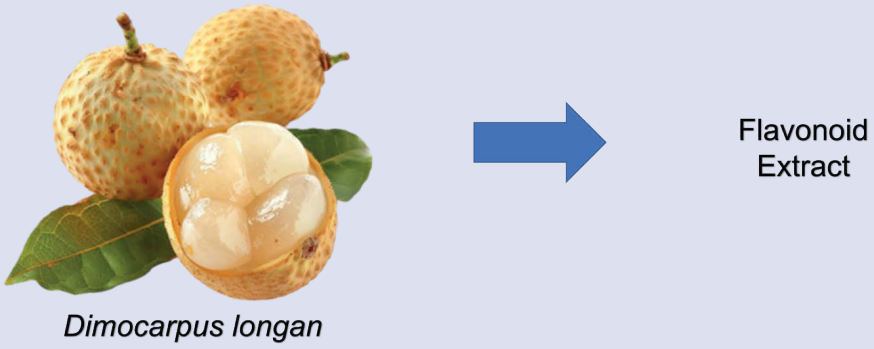
GRAPHICAL ABSTRACT



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