



The Effect of Co-Processed Excipient and Mannitol on Physical Properties and Release Test of Flavonoid Total of Bajakah Root (*Spatholobus littoralis* Hassk.) Water Extract in Effervescent Granule

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ABSTRACT. Bajakah root is traditionally used in herbal medicine and exhibits notable antioxidant activity ($IC_{50} = 0.155$ mg/mL) with a total flavonoid content of 32.49 ppm. However, conventional consumption by boiling is limited by poor taste, odor, and low stability. To overcome these limitations, this study developed effervescent granules to enhance acceptability and absorption. The effect of combining co-processed excipients (lactose–manihot starch in a ratio of 64.32:35.68) with mannitol on granule characteristics and flavonoid release was investigated. Bajakah root extract was prepared by hot water infusion (90 °C, 15 min; ratios 1:10 and 1:20) followed by freeze-drying. Phytochemical contents were analyzed using UV–Vis spectrophotometry. Co-processed excipients were prepared via wet granulation and characterized by SEM. Formulations with co-processed excipient–mannitol ratios of 3:1, 2:2, and 1:3 were evaluated for physicochemical properties and flavonoid release. Results showed total flavonoid and tannin contents in the extract of 2.19% and 9.04%, respectively. Excipient ratios significantly affected flow rate, angle of repose, moisture content, and disintegration time ($p < 0.05$), but not pH. All formulations met pharmacopeial requirements for $\geq 80\%$ flavonoid release within 60 min. The optimal formulation (1:3) demonstrated excellent properties, supporting its potential as an effective herbal delivery system.

INTRODUCTION

Bajakah is rooted in the Fabaceae family, which grows wild in the tropical forests of Kalimantan and propagates on wood trees from the Phaseoleae tribe. The people of Kalimantan traditionally use this plant to treat pain, diarrhea, lumps on the body, and lower uric acid levels (Sianipar *et al.*, 2023), as well as medicinal herbs, because it has anti-inflammatory and anti-cancer properties. Based on the results of the phytochemical screening carried out by Saputera and Ayuhecaria (2018), Bajakah root (*Spatholobus littoralis* Hass.) contains flavonoids, saponins, tannins, and polyphenols. The antioxidant activity (IC_{50}) of Bajakah root water extract using 2,2-diphenyl-1-picrylhydrazyl (DPPH) reagent is 155 $\mu\text{g/mL}$ (Ermawati *et al.*, 2024). In previous research, the total flavonoid levels of Bajakah roots in the stem and bark were 32.49 ppm and 15.95 ppm, respectively (Fitriani *et al.*, 2020). Traditionally, Bajakah roots are consumed by boiling. This method is considered less efficient; it has problems with taste and an unpleasant aroma of Bajakah root, and it can only be stored for a short time, so an appropriate delivery system needs to be formulated to deliver Bajakah root extract.

Effervescent granules were chosen as the delivery system because they offer advantages, including a more homogeneous distribution of active ingredients, increased fluid intake, an attractive, pleasant taste, practicality, and the elimination of a tableting process (Patel and Siddaiah, 2018). In the effervescent dosage form, active substances are released more quickly when in contact with water due to an acid-base reaction between the acid

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and the carbonate base (Mahdiyyah *et al.*, 2020). In the effervescent granule formula, the choice of filler is important because it can affect the granules' physical properties and the release of the active substance. A formula containing lactose as a filler shows reasonable drug release rates (Aprilia *et al.*, 2021). According to research by İpci *et al.* (2016), effervescent bases exhibit higher solubility and faster drug absorption than conventional tablets. Effervescent granules are released within 5 minutes (Al-Mousawy *et al.*, 2019). The bitter taste of Bajakah roots makes it less acceptable, so an effervescent granule filler is needed that can be used to improve taste. Mannitol has a sweet taste and provides a cool sensation in the mouth (Rowe *et al.*, 2006). Using large quantities of mannitol in effervescent granule formulas can improve flow properties and taste. However, it will increase the water content and the angle of repose of the granules, leading to poor granule stability (Rahmayanti, 2022).

Fillers determine the overall properties of the mixture, especially at low levels of active substances. One additional ingredient that acts as a filler and binder is the co-processed excipient (Ermawati *et al.*, 2020). Co-processed excipients are additional ingredients made by combining two or more excipients to alter their physical properties in a way that cannot be achieved by simple physical mixing alone, without significant chemical changes (Kathpalia and Jogi, 2014). Using co-processed excipients has some advantages, including avoiding chemical changes or structural alterations, improving flow properties, and increasing compressibility (İpci *et al.*, 2016). Starch composition of suweg tubers amyllum (*Amorpopallus paeoniifolius* (Dennst.) Nicolson) and lactose as a co-process excipient with a percent ratio of 64.32:35.68 produced effervescent granules that met the requirements (Ermawati *et al.*, 2020). A combination of lactose-amylum manihot in a 50:50% ratio in a Bajakah root water extract tablet produces granules that meet the requirements (Ermawati *et al.*, 2024). A 75:25 ratio of lactose and mannitol yielded the best granule evaluation (Rahmayanti, 2022). The effect of filler materials with a combination of co-processed excipient (lactose-amylum manihot) with mannitol in effervescent granules has never been studied. This research aims to determine the effects of variations in co-processed excipient and mannitol fillers on the performance of effervescent granules and on the total flavonoid release test of Bajakah root water extract.

RESEARCH METHODS

Materials: Bajakah root (Palangkaraya, Central Kalimantan), lactose (TC Jacoby Inc., USA), Manihot starch (Intelligent Materials Pvt. Ltd., USA), citric acid (Weifang Winsign Industry Co., Ltd., China), sodium bicarbonate (Inner Mongolia Ihjuchem Industrial Co., Ltd, China), PVP K-30 (JH Nanhang Life Sciences Co., Ltd., China), mannitol (USP grade, Lab Aley), taro flavor powder (PT. Matcha Muda Manggala, Indonesia), quercetin (Sigma-Aldrich, USA), aluminum chloride powder 99.99% (Sigma-Aldrich, USA), acetic acid glacial >99% (Sigma-Aldrich, USA), sodium tungstate dihydrate (Sigma-Aldrich, USA), and distilled water (Saba Kimia, Indonesia). Instrumen: Analytical balance (KERN: ABS 220-4, Germany), hot plate (IKA C-MAG, Germany), magnetic stirrer (Cole Parmer, Canada), moisture analyzer (Ohaus, USA), freeze-Dryer (alpha LD plus, Ukraine), oven (Memmert, Germany), dissolution apparatus type 2 (RC-6, China), spectrophotometer UV-Vis (Genesys 150 Thermo Scientific, USA), scanning electron microscopy (SEM) (JEOL, SU3500).

Sample Preparation

Bajakah root determination was conducted at the Biology Laboratory of the Faculty of Mathematics and Natural Sciences, UNS. Bajakah root water extract was prepared by the infusion method. The simplicia of 1000 g was boiled in 20 L of distilled water (1:20). Then, re-extraction was carried out with water solvent in a ratio of 1:10. The Bajakah roots were boiled for 15 minutes, when the water temperature reached 80 °C. The filtrate was evaporated until concentrated and then dried with a freeze-dryer (Ermawati *et al.*, 2023)

Detection of Active Ingredients Concentration

The quercetin solution at 100 ppm was diluted to 0.5 mL, then 0.5 mL of 10% AlCl₃ and 4 mL of 5% acetic acid were added, and the mixture was incubated for 30 minutes. The mixed solution is then scanned using a UV-Vis spectrophotometer over 370-450 nm. The stock solution at 1000 ppm was then diluted to 20, 40, 60, 80, and 100 ppm. Each concentration series took 0.5 mL, then reacted with 0.5 mL of 10% AlCl₃ and 4 mL of 5% acetic acid, and was incubated for 30 minutes. The absorbance was measured using a UV-Vis Spectrophotometer at the maximum wavelength obtained. A 50 mg bajakah extract was dissolved in 96% ethanol and diluted to a final volume of 10 mL. An aliquot of 0.5 mL of this solution was then mixed with 0.5 mL of 10% AlCl₃ and 4 mL of 5% acetic acid, and incubated for 30 minutes. The absorbance was measured at the maximum wavelength using a UV-Vis spectrophotometer. All measurements were performed in triplicate (Patil *et al.*, 1995).

Tannic acid stock solution of 1000 ppm. Tannic acid weighed 10.0 mg and was dissolved in 10.0 mL of distilled water. Five grams of sodium tungstate were mixed with 1.0 grams of phosphomolybdic acid, 50 mL of distilled water, and 2.5 mL of phosphoric acid. A Na_2CO_3 sample weighing 7.5 g was dissolved in distilled water and heated to 60 °C; then 100 mL of distilled water was added. The concentration series was used to generate a standard curve, and absorbance was measured over the 400-800 nm wavelength range. Measurements on a series of standard curves of 10, 20, 25, 30, and 35 ppm using a UV-Vis Spectrophotometer at the maximum wavelength obtained. Bajakah root water extract 150 mg was dissolved in 10 mL of diethyl ether, filtered, and evaporated. The extract was added with distilled water up to 10.0 mL. A 1.0 mL extract solution was taken, and 1.0 mL of Folin-Denis reagent was added. The solution was left for 3 minutes, then 1.0 mL of saturated Na_2CO_3 solution was added, followed by distilled water to 10.0 mL, diluted ten times, and incubated for 40 minutes to allow the sample solution and reagent to react completely. The final procedure involved measuring the absorption at the maximum wavelength obtained (Harbertson *et al.*, 2008).

Co-processed Excipient

Co-processed excipients (lactose–manihot starch) were prepared by wet granulation using PVP K-30 as a binder. The composition of manihot starch, lactose, and PVP K-30 was weighed according to optimal proportions researched by Ermawati *et al.* (2020). Co-processed excipients in the amount of 30 g required PVP K-30 and filler material in a ratio of 1:9. The filler material, namely lactose:manihot starch, required a percent ratio of 35.68:64.32 (Ermawati *et al.*, 2020). PVP K-30 was put in a mortar, and hot water was added while stirring until fluffy. Manihot starch-lactose was mixed thoroughly in the mortar until a moist mass was formed. Then, sieving was carried out using sieve no. 14. The granule was formed and then baked in the oven for 30 minutes at 50 °C. Dry granules were then sifted with sieve no. 16 to obtain co-processed excipients. SEM analysis was conducted on co-processed excipient compounds (lactose–manihot starch), pure lactose, and pure Manihot starch. Samples were characterized at 500-, 5000-, and 10,000-times magnification.

Granule Effervescent Formulation

Effervescent granules were made by separately processing the ingredients into acid granules (citric acid) and alkaline granules (sodium bicarbonate). Each granule component in Table 1 was weighed, and the material components were wet granulated using PVP K-30 as a binder, which had been made into mucilage. Then, the granulation results were sieved through a 14-sieve and placed in the oven for 30 minutes at 50 °C. After being oven-dried, the granules were sifted again using sieve no. 16. Then, Citric acid, sodium bicarbonate, and co-processed excipients were homogenized in dry conditions. Granule evaluation was carried out, which included organoleptic, flow rate, angle of repose, moisture content, time solubility, pH, and release of active substances (dissolution).

Table 1. The composition of the effervescent granule formula of Bajakah root water extract, using various concentration ratios of filler (co-processed excipient-mannitol), resulting in Formulas 1 – 3, which have ratios of 3:1, 2:2, and 1:3, respectively.

Ingredients	Weight (mg)			Function
	Formula 1	Formula 2	Formula 3	
Bajakah root water extract	90.0	90.0	90.0	Active ingredient
Sodium bicarbonate	891.0	891.0	891.0	Base compound
Citric acid	1164.0	1164.0	1164.0	Acid compound
Co-processed excipient	607.5	405.0	202.5	Filler
Mannitol	202.5	405.0	607.5	Filler
PVP K-30	30.0	30.0	30.0	Binder
Taro Flavour	15.0	15.0	15.0	Flavouring agent
	3000.0	3000.0	3000.0	

Physical Properties of Granule Effervescent

Organoleptic testing of Bajakah root water extract effervescent granules includes shape, color, taste, and aroma. 100 g of granules was poured slowly into the measuring funnel on the tool; the funnel was opened slowly, and the granules were allowed to flow out. The time was recorded with a stopwatch until all the granules came out. The flow rate was calculated in grams/second and replicated three times. Repose angle testing required the

height (h) of the granule pile and the radius (r) of its base. The repose of the angle was calculated using Equation 1.

$$\tan \alpha = \frac{h}{r} \quad (1)$$

Where α = angle of repose, h = height of granule pile, and r = radius of granule pile.

The moisture content of the granules was measured using a moisture analyzer. Granules weighing 2.0 g were placed on an aluminum plate in the tool and heated to 105 °C until the weight remained constant. Test results were recorded and replicated three times. Granules prepared from each formula were placed into a beaker containing 200 mL of distilled water at 25 ± 1 °C. Next, the time required for the effervescent reaction to dissolve until a clear solution formed was recorded. The pH of the effervescent solution was measured by dissolving the Bajakah root water extract effervescent granules in 200 mL of distilled water using a calibrated pH meter. The measurement results were good if the pH of the effervescent solution was close to neutral (Ipci *et al.*, 2016).

Release Test of Granule

Stock solution A was prepared by dissolving 10 mg of quercetin powder in 96% ethanol pro analysis (p.a.) in a volumetric flask to a final volume of 10 mL. Stock solution B was prepared by dissolving 0.1 mL of solution A in phosphate buffer solution in a volumetric flask to a final volume of 10 mL. The absorbance of stock solution B was measured at the maximum wavelength using UV-Vis spectrophotometry in the 200–500 nm range. Various volumes of 0.1, 1.5, 3.0, 4.5, and 6.0 mL of stock solution B were taken using a micropipette and dissolved in phosphate buffer to a final volume of 10 mL. The absorbance of the series solutions was read at the maximum wavelength of quercetin, with the buffer solution used as a blank. The dissolution test equipment used was the paddle method. The sample was dissolved in a container containing 500 mL of dissolution media at 37 ± 0.5 °C, and the mixture was stirred at 50 rpm. Dissolution was carried out in a phosphate buffer solution at pH 6.8 for 60 minutes. The absorbance was measured at 0, 2, 5, 10, 15, 20, 30, 45, and 60 minutes by taking 10 mL of the dissolved solution and measuring it with a UV-Vis spectrophotometer. The taken media was replaced with the same amount previously taken. Bajakah root water extract release levels were calculated based on the calibration curve for total flavonoid concentration (Cohen *et al.*, 1990; Ermawati *et al.*, 2023).

Data Analysis

Test results and evaluation of the Bajakah root water extract effervescent granule formulation with variations of mannitol and co-processed excipient, which includes organoleptic, flow rate, angle of repose, moisture content, solubility time, and pH, were analyzed using the ANOVA test and Kruskal-Wallis test. Total flavonoid release test results were then analyzed based on the Indonesian Pharmacopeia Edition VI requirements.

RESULTS AND DISCUSSION

The results of plant determination (number 054/UN27.9.6.4/Lab/2023) indicate that the plant used in this research was Bajakah root, belonging to the species *Spatholobus littoralis* Hassk. The freeze-dried extract yielded a total dry extract of 8.1 g (8.1%). It can be concluded that the research yield is eligible, a good yield value is not less than 7.2% (Gligor *et al.*, 2023). The infusion method yields lower amounts than other extraction methods, such as reflux and maceration. This was due to the 15-minute extraction time. However, the short extraction time can reduce the risk of destroying active substances in the sample. The appearance of the Bajakah root water extract, freeze-dried powder, effervescent granules, and effervescent solution is presented in Figure 1. The freeze-dried extract was stable, with a low density and longer shelf life (Silambarasan and Rajalakshmi, 2022).

SEM test results to observe the surface form of co-processed excipient lactose–manihot starch. A SEM test was carried out at 10.00 kV. The morphology of the materials was observed at magnifications of 500, 5,000, and 10,000, as shown in Figure 2. The morphology of the co-processed excipient made from lactose–manihot starch forms bonds, leading to larger granules. There were various shapes of the components. Lactose was round, and Manihot starch was pentagonal in shape, which then melted and formed a mass with the PVP K-30 binder into large, irregularly shaped, rough-surfaced, porous granules.

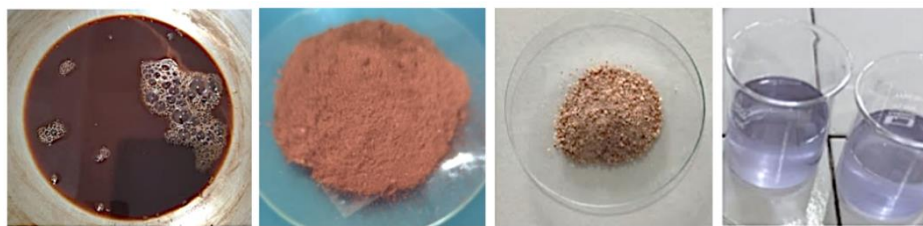


Figure 1. The results of (a) Bajakah root water extract, (b) Bajakah root dried powder after freeze drying, (c) Bajakah root effervescent granule, and (d) Bajakah root effervescent solution.

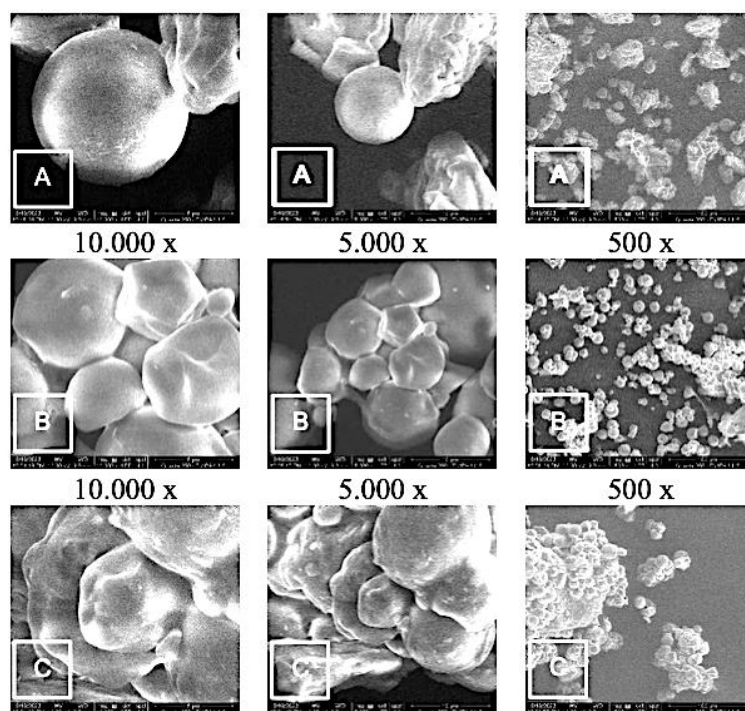


Figure 2. The SEM analysis results, where (a) lactose, (b) amyllum Manihot, and (c) co-processed excipient.

Physical Properties of Granule

A granule flow rate test is performed to determine the granule flow time, as it affects the consistency of granule movement speed. The results of the granule flow time test (Table 2) show that all formulas meet the flow time requirements of >10 g/second. A flow rate of more than 10 g/sec indicates that the granules flow easily through the hopper funnel. The granule flow speed is influenced by several factors, including granule size, shape, granule surface properties, density, and humidity. Based on the results of the ANOVA statistical analysis, it can be concluded that variations in co-processed excipient (lactose–manihot starch) and mannitol have a significant effect ($p < 0.05$) on granule flow rate. The higher the mannitol content, the slower the flow rate. This is based on research by Aprilia *et al.* (2021), who used 11.7% mannitol and reported the lowest average flow speed of 5.95 seconds. Mannitol has poor flow properties and compressibility. Therefore, the formula with Greater mannitol concentrations has longer flow times.

Table 2. The physical properties of the effervescent granule of Bajakah root water extract formulated in various formulas (Formulas 1 – 3 have ratios of filler (co-processed excipient:mannitol) of 3:1, 2:2, and 1:3, respectively).

Physical Properties	Formula 1	Formula 2	Formula 3
Granule flow rate (g/sec)	13.70 ± 1.117	12.98 ± 1.006 ^a	10.43 ± 0.435 ^a
Repose of angle (°)	28.38 ± 0.944	29.74 ± 0.379 ^b	30.69 ± 0.819 ^b
Moisture content (%)	2.42 ± 0.069	2.73 ± 0.121 ^c	3.42 ± 0.075 ^c
Time of dissolution (min)	4.14 ± 0.130	4.34 ± 0.155 ^d	4.61 ± 0.062 ^d
pH value	6.56 ± 0.057	6.53 ± 0.057	6.56 ± 0.057

*a,b,c,d...significant difference between formulas

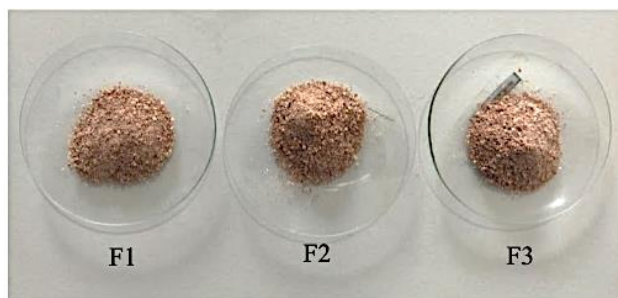


Figure 3. The photograph of the resulting Bajakah root extract effervescent granule, formulated with various ratios of filler ingredients (co-processed excipient and mannitol).

The angle of repose of granules is influenced by several factors, including shape, size, and moisture content. Moist granules will improve granule cohesiveness, which decreases granule quality, reducing granule flow speed and angle of repose (Rahmayanti, 2022). Based on the test results, the three formulas meet the requirements for a good angle of repose between 25° and 45° (Lachman *et al.*, 1994). Based on the results of the ANOVA statistical analysis, it is known that variations in the co-processed excipient (lactose–manihot starch) and mannitol had a significant effect ($p < 0.05$) on the angle of repose of the granule. According to Rahmayanti (2022), the greater the concentration of mannitol in a formula, the higher the granules' water content and angle of repose. Formula 3 has the most extensive mannitol composition, so it has the highest angle of repose. The lower angle of repose indicated better granule characteristics.

Too high granule moisture content makes the granules unstable and exhibit poor flow properties, while too low a level can make the granules fragile (Bhatia *et al.*, 2022). As shown in Figure 3, all formulations produced visually uniform granules, and all three met moisture requirements. The moisture content of granules should be below 5%, according to the Indonesian Pharmacopeia (BPOM, 2014), although values up to 10% may still be acceptable for certain formulations. Based on the results of ANOVA statistical analysis, it can be concluded that variations of co-processed excipient (lactose–manihot starch) and mannitol have significant effects ($p < 0.05$) on granule moisture content. A higher mannitol composition may increase granule moisture.

Dissolution Time

Dissolution time is the time required for the granules to completely dissolve when an acid and an alkaline source react upon contact with water, producing CO₂ that causes the granules to crumble and dissolve. The cessation of CO₂ gas production in water indicates complete solubility. The test results showed that the three formulas met < the dissolving time requirements of <5 minutes. Based on the ANOVA results, co-processed excipients and mannitol have a significant effect ($p < 0.05$) on the dissolution time of effervescent granules. Formula 1 has the fastest average time of 4.14 minutes. According to Arifuddin *et al.* (2022), there is a fast dissolution time for effervescent granules because there is no premature reaction of the effervescent caused by the granule moisture. Dissolution time is also influenced by granule porosity. The greater granule porosity means the spaces between particles are larger. That makes it easier for fluid to enter the granule structure and push granules to disintegrate. Using manihot starch as a disintegration agent in co-processed excipients can increase granule porosity, thereby reducing dissolving time.

The pH measurement aims to determine the pH of effervescent granules dissolved in water or ready for consumption. The pH measurement was carried out to determine whether the resulting solution is safe for consumption, so that it does not irritate the stomach, and to assess the taste; a low pH indicates a sour taste. Based on the results of Kruskal-Wallis's statistical analysis, variations of co-processed excipient and mannitol fillers did not have a significant effect ($p > 0.05$) on the pH of effervescent granules. Composition: The same citric acid and sodium bicarbonate in each formula causes no significant difference in pH values between formulas. The formation of CO₂ also affects the pH during the effervescent reaction. As it progresses, some will dissolve to form carbonic acid, which then decomposes to produce H⁺ ions, increasing the solution's acidity.

Release Test of Flavonoid Total

The calibration curve equation for flavonoid total in a phosphate buffer medium was used to calculate the concentration of total flavonoids released from Bajakah root water extract effervescent granules into the medium over time. Based on the calibration curve, the correlation coefficient (R) is 0.9673. The linear regression obtained is $y = 0.0069x + 0.0021$ according to the equation $y = a + bx$. The values obtained are a (intercept) = 0.0021 and

b (slope) = 0.0069; the Lambert-Beer Law shows a linear relationship between absorbance and analyte concentration. One of the test parameters required by the Indonesian pharmacopeia to ensure product quality is the dissolution test. This test aims to determine the release profile of the active ingredient from the preparation in the medium. This test has been used to ensure continuous product quality. Several factors influence the percentage dissolution of an active substance: test method, type and temperature of media, form preparation, and pH (Sathe *et al.*, 2005).

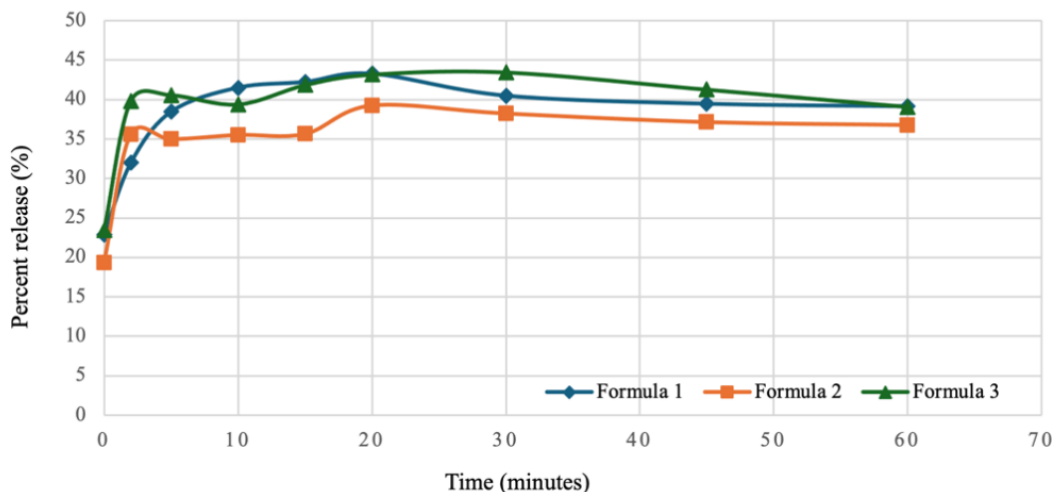


Figure 4. The profile of the percent release of flavonoid total of Bajakah root water extract effervescent granule.

The active substance concentration is determined each time using a UV-Vis spectrophotometer at a wavelength maximum (λ_{\max}) of 373 nm. The highest dissolution efficiency (%DE) value is obtained in Formula 1 (103.03%), followed by Formula 2 (96.64%) and Formula 3 (94.97%). The value exceeding 100% may be attributed to experimental variability or slight overestimation in the assay, which is commonly observed in spectrophotometric measurements (Figure 4). The %DE value is an indicator of the release capability of active ingredients from a preparation over a given period. The results of the %DE calculation show that variations in the co-processed excipient and mannitol in the barakah root water extract effervescent granule affect the dissolution rate. The entire formula meets the requirements, namely, the dissolved percentage of the active ingredient is more than 80% within 60 minutes, according to *Farmakope Indonesia Edition VI (2020)*.

Lactose fillers exhibit a good rate of drug release (Aprilia *et al.*, 2021). The bioavailability and stability of the preparation depend greatly on the excipients selected, the number used, and their interactions with the active substances or other excipients. Based on the results, Formula 1 has the highest %DE. Formula 1 has the highest concentration of co-processed excipient composition with a ratio of co-processed excipient and mannitol (1:3). This is in accordance with research by Laili *et al.* (2019), which formulated co-processed excipients using PVP K-30 as a binding agent and resulted in the excellent drug release. Co-processing of lactose with manihot starch increases granule size and porosity, making penetration through the granule pores easier and facilitating the release of active ingredients from the preparation (Bhatia *et al.*, 2022). An improved composition of co-processed excipients will enhance the release ability of active ingredients from the preparation (Bhatia *et al.*, 2022).

CONCLUSION

Variations in the co-processed excipient and mannitol have had a significant effect on the flow speed, angle of repose, moisture content, and dissolution time of effervescent granules of Bajakah root water extract, but did not affect organoleptic properties and pH. The high proportion of co-processed excipients results in a higher flow rate, a slight angle of repose, a lower moisture content, and faster dissolution. The formula with variations in co-processed excipient and mannitol in a ratio of 1:3 produces a good evaluation of effervescent granules with a granule flow rate of 13.70 ± 1.12 g/sec; angle of repose of 28.38 ± 0.94 °; moisture content of $2.42 \pm 0.07\%$; disintegration time of 4.14 ± 0.13 min; and pH of 6.56 ± 0.06 .

CONFLICT OF INTEREST

All authors declare there is no potential conflict of interest in this article.

AUTHOR CONTRIBUTION

DEE: Conceptualization, Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation, Writing-review and editing, Visualization, Project administration, Funding acquisition; NCFT: Metodology, Software, Formal Analysis, Investigation, Resources, Data Curation, Writing-original draft, Visualization; UAD: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; NCAS: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; SR: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; MFZ: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; ADM: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; DTU: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; ANA: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; HS: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation; MKU: Methodology, Validation, Formal Analysis, Investigation, Resources, Data Curation.

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