The Effect of Organic Acids Concentration Extracted from the Fruit on the Conversion of Starch to Glucose Using Microwave-Assisted Acid Hydrolysis (MAAH)

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ABSTRACT

Fruits such as lemon (Citrus limon L.), starfruit (Averrhoa bilimbi L.), and pineapple (Ananas comosus (L.) Merr) have high organic acid content which can be studied for their use as acid catalysts in starch hydrolysis. The goal is to produce glucose syrup that is safer for consumption. After extraction and determination of the total titrated acid concentration (TTA), the acid was added as a catalyst for starch hydrolysis. The purpose of this study was to determine the effect of the TTA concentration of the three fruit extracts on the conversion of starch into glucose by heating using the Microwave-Assisted Acid Hydrolysis (MAAH) method at various power (40 and 50 Watt) for 5, 10 and 15 minutes. The hydrolyzed starches were sorghum and cassava with various extract volumes of 15-25 mL. To determine the highest sugar content of each variation of the hydrolysis process is determined from the increase in the value of % Brix at room temperature. In the hydrolysis process with the highest % Brix value, glucose levels were determined quantitatively by the Luff-Schoorl method. When compared with conventional hydrolysis, the sugar resulting from the MAAH hydrolysis process provides a sugar content value that is twice as high as with a shorter processing time of 15 minutes vs. 3 hours. The results of the conversion of 4% cassava starch into glucose using pineapple extract is 1,62% of reducing sugar.

Keywords: *hydrolysis, starch, glucose, organic acids, total titrated acid.*

1. INTRODUCTION

Starch hydrolysis breaks starch molecules into simpler constituent parts with shorter chains such as dextrin, isomaltose, maltose, and glucose [1]. Hydrolysis can be in the form of complete hydrolysis, namely the hydrolysis process that produces D-glucose sugar or commonly known as dextrose, and incomplete or partial hydrolysis which results in the form of dextrin, maltose, or other monosaccharides.

Starch hydrolysis solution, better known by the trade name glucose syrup, is a thick and clear liquid with the main component of glucose that can be obtained from starch hydrolysis through chemical or enzymatic means. Glucose syrup can be made from wheat starch, potato, sweet potatoes, corn, sorghum, and other ingredients that contain high starch [2,3].

$$\underset{\text{starch}}{(C_6H_{10}O_5)_n + nH_2O} \xrightarrow[\text{catalyst}]{} \underset{\text{glucose}}{(C_6H_{12}O_6)}$$

The hydrolysis reaction occurs very slowly without a catalyst, so it is necessary to add a catalyst. The catalysts commonly used in starch hydrolysis are acids or enzymes. The use of strong acid catalysts has the advantage of sugar recovery and high reaction rates but has environmental and corrosion problems as well as high costs for utility and recovery of acid use [4,5]. The process of starch hydrolysis using acid is influenced by the size of the material, acid concentration, temperature, time, the ratio of ingredients, and stirring [6]. Enzymatic hydrolysis has almost equal efficiency seen from the yield of reducing sugar produced, but economically the material needed for acid hydrolysis is much cheaper, and the time needed for the process is shorter via enzymatic [7].

Many fruits or flower plants contain quite high organic acids, including pineapple, starfruit, and lemon. Organic acids commonly found in fruit or flower plants include citric acid, malic acid, tartaric acid, oxalic acid, hibiscus acid, and ascorbic acid. Organic acids such as citric acid and malic acid can interact with the hydroxyl groups of tapioca starch and act in the acid hydrolysis of starch molecules, reducing the starch chain connections [6]. This is an opportunity to study starch hydrolysis using natural organic acid catalysts to produce glucose syrup that is safe from chemicals. The advantage of using this organic acid catalyst does not require a neutralizing step because the acids are good for health. More than that the content of vitamins and natural dyes will add value to glucose syrup.

Pineapple, starfruit, and lemon are known to contain a high percentage of organic acids such as citric acid, hydroxy citric acid, hibiscus acid, malic acid, and tartaric acid as the main compounds, as well as oxalic acid and ascorbic acid as minor compounds [8]. In addition, there are organic components such as phenolic (protocatechuic acid [PCA] and eugenol), flavonoid type polyphenol components (3-glucoside anthocyanins, anthocyanidins; flavonol quercetin), organic acids and their derivatives, vitamin C (ascorbic acid), B1 (thiamin), B2 (riboflavin), and carotenoids (β-carotene) [9].

Organic acids, such as citric acid and malic acid, can interact with the hydroxyl groups of starch and act in the acid hydrolysis of starch molecules, reducing starch chain connections [10–12]. This is an opportunity to study starch hydrolysis using acid-catalyzed natural organic acid. The advantages of using organic acid catalysts do not require a neutralization step because the acids are good for health. More than that, the content of vitamins and natural dyes will add value to glucose syrup.

In a previous study, a study on the effect of the acidity concentration of rosella flower extract on the conversion of sorghum starch into glucose obtained glucose levels through a conventional heating process at a temperature of 96°C for 3 hours obtained a glucose level of 18% [13]. In this study, the starch hydrolysis process was carried out by heating using a microwave (MAAH). It was reported that when compared with conventional heating methods, the MAAH method increased the reaction rate by 50-100 times [14]. According to Hou et al.[15], microwave heating is an alternative method for hydrolysis of biomass into simple sugars with the potential as a fast, efficient and selective method compared to conventional methods. Microwave heating can increase the solubility and decrease the crystallinity of starch and fiber because starch undergoes gelatinization and hydrolysis into its constituent monomer compounds. The higher the acid concentration and the longer the time for hydrolysis, the higher the total sugar and reducing sugar levels that are hydrolyzed [16].

2. RESEARCH METHODS

In general, the research began with the preparation of starch and organic acid extracts from pineapple, star fruit, and lemon. Furthermore, hydrolysis was carried out by mixing starch and organic acid extracts at certain ratios and conditions using microwave and conventional heating. The results of the hydrolysis were tested qualitatively for glucose levels based on the increase in % Brix. In the best conditions, glucose levels were quantitatively analyzed using the Luff Scroll method.

2.1 Hydrolysis of Starch Sorghum with Organic Acid Catalyst

One gram of starch into the Erlenmeyer then distilled water added, then it heated until it dissolves and thickens. The solution was added to the acid extract with the variation of volume 15-25 mL Hydrolysis was carried out using *Microwave-Assisted Acid Hydrolysis* (MAAH) with power variation (40 dan 60 Watt) and variations in hydrolysis time of 5, 10, and 15 minutes.

2.2 Microwave-Assisted Acid Hydrolysis (MAAH)

A modified domestic microwave oven (maximum delivered power of 800 W) was used to employ MAAH with a wave frequency of 2450 MHz.

2.3 Analysis of Glucose Levels by Refractometer

Sugar content was analyzed by Refractometer to get a degree of Brix.

3. RESULT AND DISCUSSION

3.1 Determination of pH Value and TTA Extracts of pineapple, star fruit, and lemon.

Visually, these three plant extracts have a strong sour taste, indicating that their organic acid content is quite high. The sour taste is known because of the high acid content in it. hibiscus, citrate, oxalate, malic, 3-indolyl acetic acid, and ascorbic in roselle petal extract 2-9% and 8% tartaric acid, 11 mg / 100g ascorbic acid, and 9% titrated acid. [19,20]. The results of the analysis of the degree of acidity (pH) and the total value of titrated organic acids from the three plants are shown in Table 1. One of the three types of plants extracted, starfruit had the lowest pH or highest total acidity. To determine the effect of %TTA on glucose yield, the acid concentration was varied by adding the extract 15-25 mL.

Table 1. pH and TTA value.

Fruits	pH	TTA (N)
Pineapple (Ananas comusus (L.) Merr)	3,75	0,0647
Starfuit (*Averrhoa bilimbi L.)*	1,42	0,2216
Lemon (*Citrus limon*)	2,28	0,8899

3.2 Effect of Total Titrated acid on sugar content

The effect of the TTA concentration of each fruit extract on the total sugar content resulting from the hydrolysis of tapioca starch and sorghum measured from the increase in the % Brix value is shown in Figure 1. The results of the % Brix measurement are used as initial data to determine the best conditions for starch hydrolysis. The % Brix value is directly proportional to total sugar content including reducing sugars such as glucose. The higher the sugar content in the sample, the higher the % Brix.

3.3 Analysis of sugar levels from conventional hydrolysis

The increased value of %brix from sorghum starch hydrolysis can be seen in **Figure 1**. The highest sugar percentage was produced by the catalyst of lemon extract at 4 hours of hydrolysis time.

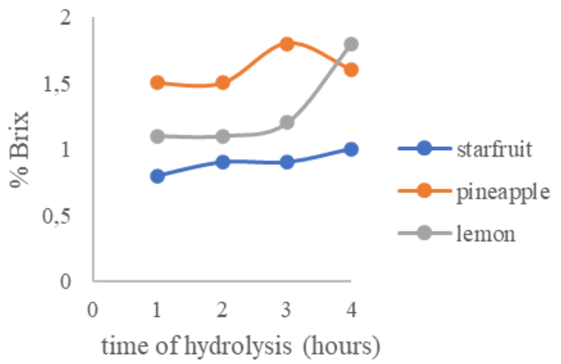

Figure 1 Effect of pineapple, starfruit, and lemon extracts on sugar levels from conventional sorghum starch hydrolysis.

The increased value of %brix from cassava starch hydrolysis can be seen in **Figure 2**. The highest sugar was produced by the catalyst of lemon extract at 3 hours hydrolysis time.

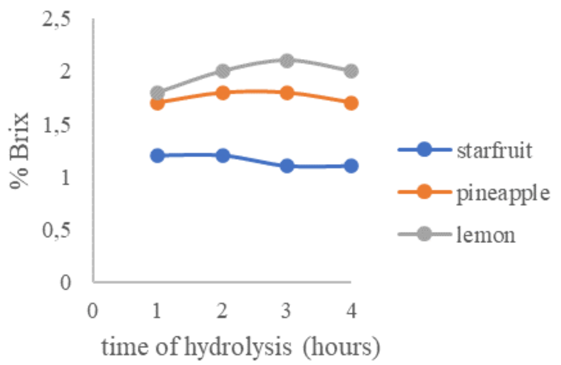

Figure 2 Effect of pineapple, starfruit, and lemon extracts on sugar levels from conventional tapioca starch hydrolysis.

3.4 Analysis of sugar levels from MAAH

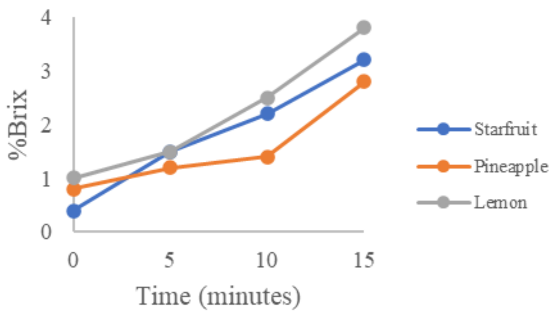

Figure 3 Effect of fruit extract on %Brix tapioca starch with MAAH

With the increase in glucose levels in hydrolyzed starch, it can be said that the organic acid extract from this material can affect the starch hydrolysis process. Figures 1 and 2 show the effect of %TTA from various fruit extracts on the glucose levels of sorghum and tapioca starch, it appears that tapioca starch provides a higher glucose yield than sorghum starch. According to SNI 01-2978-1992, the quality requirements for glucose syrup are at least 30% (w/w) so that starch hydrolysis is still needed to do more study so the level of 30% can be achieved.

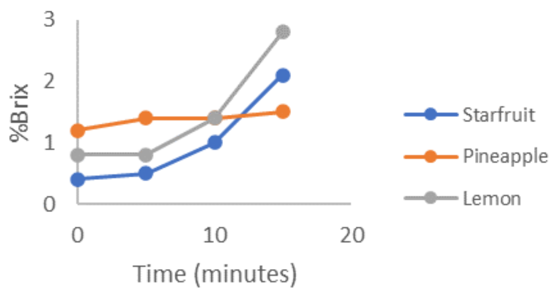

Figure 4 Effect of fruit extract on %Brix sorghum starch with MAAH

The hydrolysis yield of tapioca starch is higher than that of sorghum starch. This can be caused by the higher amylose content in tapioca flour as the starch source used, which is in the range of 30%, while sorghum starch contains amylose in the range of 21-26% [21]. Amylose is an amorphous part of starch granules and is a longer area that is easily attacked by acids [22] so that amylose will be more easily degraded when given the heat and a catalyst than amylopectin. Because tapioca starch has a higher amylose content, it allows tapioca starch to produce higher sugar content.

Overall, both conventionally and MAAH, lemon extract gave the highest % brix value to the hydrolysis of sorghum or tapioca starch. This shows that what affects the hydrolysis process is the TTA value as shown in Table 1, lemon has the highest TTA value compared to pineapple and star fruit.

The quantitative characterization of the hydrolysis product was carried out on tapioca starch samples using pineapple fruit extract including the determination of reducing sugar levels using the Luff-Schoorl method, water content, and vitamin C (Table 2). Determination of vitamin C levels aims to know the effect of the added value of the natural content of the fruit extract on the glucose syrup produced.

From **Table 2**, we can conclude that the higher the concentration of starch and TAT caused the higher the reducing sugar produced. At a starch concentration of 4% and a pineapple extract volume of 25 mL (1,6175 meq/L), the reducing sugar content was 1.62% with a water content of 91.28%. The increase in the amount of starch and extract from this limit causes process disturbances, namely, the starch solution is too thick and the formation of foam. The addition of vitamin C to the product indicates that the effect of organic acids from fruit extracts provides added value. To increase glucose levels, it is necessary to concentrate on the desired water and reducing sugar content.

Table 2. Characterization of Tapioca Starch Hydrolysis Products with Pineapple Extract

Starch (%)	Pineapple Extract Volume (mL)	Methods	Parameters tested		
			Water Content (%)	Reducing sugar (%)	Vitamin C
1	15	MAAH	98,17	0,84	9,22
2	15		83,69	1,22	9,93
	20		85,37	1,29	9,88
	25		88,98	1,35	9,86
	30		87,79	1,41	12,36
3	15		87,35	1,35	12,30
	20		87,87	1,41	12,45
	25		87,95	1,46	11,93
	30		86,78	1,51	14,30
4	15		85,67	1,45	14,79
	20		85,40	1,50	14,47
	25		84,59	1,55	14,64
	30		91,28	1,62	14,80
4	25	CONVENTIONAL	94,65	1,49	19,99
	25 (+enzim)		94,25	1,59	14,71
	0 (+enzim)		96,87	1,66	9,88
Corn syrup on the market			1,26	15,27	0

4. CONCLUSION

The conclusion of this study is the sorghum and tapioca starch hydrolysis process using natural organic acid catalysts from fruit extract can produce glucose up to 1,6% with a MAAH. With MAAH the process can be faster than conventional hydrolysis. To increase the level of glucose in the product, it is necessary to do further concentration

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