Kinetic analysis of changes in sweetness and acidity of sliced pineapples during ultrasonication

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Abstract: Changes in sweetness and acidity of sliced pineapples during ultrasonication was carried out by determining the rate of changes in degree of sweetness and pH. Half and fully ripe sliced pineapples were prepared and exposed to ultrasonic waves at the frequency of 40 kHz for 0, 5, 10, 15, 20, 25, 30, 35, and 40 minutes. Degree of sweetness, pH, and order of reaction during ultrasonication were analyzed. Sonication of half-ripe pineapples for 40 minutes resulted in the lowest total sugar content (9.53% Brix), whereas the same treatment on fully ripe pineapples resulted in the highest pH (4.73). The reduction of degree of sweetness in half and fully ripe pineapples followed a first-order reaction with rate of constants (*k*) of -0.450 and -0.612 per hour, respectively. The kinetic changes in pH during ultrasonication showed that half and fully ripe of pineapples followed a first-order reaction with rate of constants (*k*) of 0.222 and 0.216 per hour, respectively.

Keywords: kinetics changes, ultrasonication, pineapple, sugar, pH

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1 Introduction

Pineapple Queen variety is the most extensively grown pineapple in South Sumatra (Marmaini et al., 2023; Yusi, 2016). This variety is mostly consumed fresh because it tastes sweeter than other varieties and has an attractive yellow flesh with a crunchy texture. According to Arboleda et al. (2021), pineapple maturity is classified into index 1 to index 5. Index 1 (unripe) shown by 0-30% yellow pixel, index 2 (underripe) 20%-55% yellow pixel, index 3 (ripe) 45%-80% yellow pixel, index 4 and 5 (overripe) 70%-100% yellow pixel. The ripeness of pineapple has an effect on its chemical, physical and sensory characteristics. The characteristics that draw attention to pineapples are its sweet and sour taste.

The glycemic index of pineapples is high, with a glycemic index of 94.88% and a glycemic load of 47.43% in 50 grams of available carbohydrates (Okareh et al., 2021). The high glycemic index may cause pineapples to potentially increase blood sugar levels. Total sugars, ash, fibre and ascorbic acid in pineapples juice were 12.1%, 0.4%, 0.2% and 14%, respectively (Yeoh and Ali, 2017). Cordenunsi et al. (2010) reported that the total sugars found in pineapples are mostly sucrose, followed by fructose

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and glucose. The most abundant organic acids in pineapples were citric acid and ascorbic acid (Hossain et al., 2015; Lu et al., 2014). Surprisingly, the nutrient content in pineapples particularly for ash and carbohydrate contents are higher in the core than in the pineapple pulp (Santos et al., 2021), however, the core is often discarded prior to pineapple consumption.

Due to high total sugar content, there needs to be a way to reduce the total sugar content to make pineapples safe for consumption by those on a sugar diet. The method used to reduce sugar content should be environmentally friendly, without involving chemicals, and do not significantly affect the performance of the pineapple fruit. One way to reduce the sugar content in fresh pineapples is by using ultrasonic waves.

Sound waves have been used in the processing of agricultural products, where ultrasonic waves are widely used. The application of ultrasonic waves at frequency of 20 kHz - 10 MkHz is capable of causing physicochemical changes in materials in a liquid medium. Ultrasonic waves propagate in a liquid medium and has the ability to create bubbles that can burst very quickly. The burst bubbles will produce enormous energy which turns into heat energy. The creation and bursting of bubbles as well as the pressure generated during ultrasonication is called the acoustic cavitation effect which can damage cell tissue causing cells to burst. (de Carvalho Silvello et al., 2019; Muzaffar et al., 2016) used ultrasonic waves to increase the release of total sugar in sugarcane bagasse. Other works utilizing ultrasonic waves to extract total sugars or dissolved organic matters in products include those carried out by Jiang et al. (2014), Mondal et al. (2024), and Pedreschi et al. (2021).

Changes of physicochemical properties in the processing of agricultural products can be demonstrated by kinetic analysis of changes. It is, therefore, possible to predict changes that occur in a food processing process, such as changes in product quality, structure, and properties. Li et al. (2023)investigated volatile flavor changes by kinetic modelling; Vele Santos et al. (2023) reported on the kinetic changes of glucose during acid glycolysis in banana plant; Huang et al. (2023) worked on the color change kinetic during drying coffee leaves. Basically, kinetic study is the basis for explaining the speed of various changes that occur during the food processing process. The rate of a reaction changes in response to changes in the concentrations of the reactants is indicated by the reaction order. The order of a reaction is defined as the power dependence of the rate on the concentration of all reactants.

In this study, analysis of the changes in total sugar content and pH were carried out to obtain predictions of changes in degree of sweetness and pH of sliced pineapples of the Queen variety during ultrasonication.

2 Materials and methods

2.1 Materials

The pineapple Queen variety was obtained from farmers in Prabumulih district, south Sumatera, Indonesia. They were harvested in January 2023. The pineapple fruit was cut at the stalk where the pineapple fruit connects to the plant. Three pineapple fruits along with crown and stalk were tied together with the crown and stalk on. They were transferred to the laboratory of agricultural product processing with no more than 3 hours after harvested. The fully ripe pineapples were characterized by the color of the outer skin being yellow, while half-ripe pineapples were indicated by the outer half of the skin being yellow. The pineapple fruits were peeled and eyes were removed. Peeled pineapples were sliced into 2 cm thickness.

2.2 Ultrasonication process

Ultrasound treatment was carried out according to Lagnika et al. (2017) with the modification on the frequency and temperature. The frequency applied was 40 kHz and the temperature was set to 30oC.The ultrasonic bath was filled with 3 L of mineral water. The device was turned on and heated for 15 minutes at a temperature of 30^{o}C and 40 kHz frequency. Sliced pineapples were placed into the ultrasonic bath for 5, 10, 15, 20, 25, 30, 35 and 40 minutes . After the ultrasonication process was complete, the sliced pineapples were removed from the ultrasonic bath and measured for degree of sweetness and pH.

2.3 Statistical analysis

The experiments were designed as a completely randomized factorial design with two factors (level of ripeness and ultrasonication time) and each treatment was repeated three times. The first factor consisted of fully ripe pineapples and half ripe pineapples. The second factor was the ultrasonication times (0, 5, 10, 15, 20, 25, 30, 35, and 40 minutes). The data obtained were analyzed by analysis of variance (ANOVA) and the statistical significance was further analyzed by Tukey's honestly significant difference test at the 5% level.

2.3 Parameters determination

2.3.1 Degree of sweetness

Analysis of degree of sweetness was carried out using a hand-refractometer (Atago, USA). Pineapple samples were mashed and filtered using filter paper and then 5 mL of the filtrate was taken. The sample filtrate is dripped over the refractometer lens. The degree of sweetness of the sample was displayed on the refractometer monitor. The unit was expressed as degrees Brix (symbol °Bx).

2.3.2 pH measurement

pH measurements on pineapple fruits were carried out with a pH meter (Hanna, Romania). The electrode on the pH meter is standardized using an acid buffer solution (pH 4), neutral buffer solution (pH 7) and alkaline buffer solution (pH 10), then cleaned using distilled water and dried. The pineapple samples were mashed and filtered using filter paper and then placed inside a glass beaker to measure its pH.

2.3.3 Reaction order and rate of constants determination

Changes in degree of sweetness and pH, including the determination of reaction order and rate of constant was analyzed using a graphical method. The first step was plotting a graph between C versus time t, then $ln\ C$ versus time t, and $1/C$ versus time t. C denotes the measured parameters, and t denotes ultrasonication time. Equations of linear regression and R^2 were determined based on the graphs.

The reaction order was determined based on the R^2 value. If the largest value of R^2 was found in the plotting graph of C versus time t, then the reaction order followed a zero-order reaction. If the largest value of R^2 was found in the plotting graph of $ln\ C$ versus time [t], then the reaction order followed a first-order reaction. If the largest value of R^2 was found in the plotting graph of $1/C$ versus time t, then the reaction order followed a second-order reaction. The rate of constant (k) was the slope of the linear regression of the equation based on the selected reaction order. The rate of change in degree of sweetness and pH that occurred during ultrasonication was indicated by the rate of constant (k). If the k value was negative, it indicated a decrease during ultrasonication, whereas if the k value was positive, it indicated an increase during ultrasonication.

3 Results and discussion

3.1Degree of sweetness

The average degree of sweetness of treated sliced pineapples ranged from 9.53°Brix to 16.00°Bx as presented in Figure 1. Analysis of variance showed that level of ripeness, ultrasonication times and the interaction between those factors had a significant effect on the degree of sweetness of pineapple during ultrasonication. Factors that have a significant effect were further analyzed by the Tukey's Honest Significant Difference (HSD) test at the $\alpha = 0.05$ to determine treatment pairs that were significantly different.

Pineapple is classified as a non-climacteric fruit where there is no increase in taste and sugar content after harvest due to the decrease of ethylene production which functions to ripen the fruit. Half-ripe pineapples have a more dominant sour taste and lower total sugar content than fully ripe pineapples (Kamol et al., 2014). Rashima et al. (2019)reported that an increase in pineapple maturity was accompanied by an increase of total soluble solids in all pineapple varieties examined. Those findings were in line with the data shown in

Figure 1. The total soluble solids of pineapples increase due to the conversion of starch into sugars, such as glucose, sucrose, and fructose by ADP-glucose pyro phosphorylase, β-amylase, and sucrose phosphate synthase during ripening from green mature pineapple to fully yellow mature pineapple (Somasundram et al., 2018; Nadzirah et al., 2013). Furthermore, Ding and Syazwani (2016) stated that increasing fruit ripeness can increase the sugar content and yellow intensity of pineapples, as well as reduce the acid and vitamin C content of pineapple flesh.

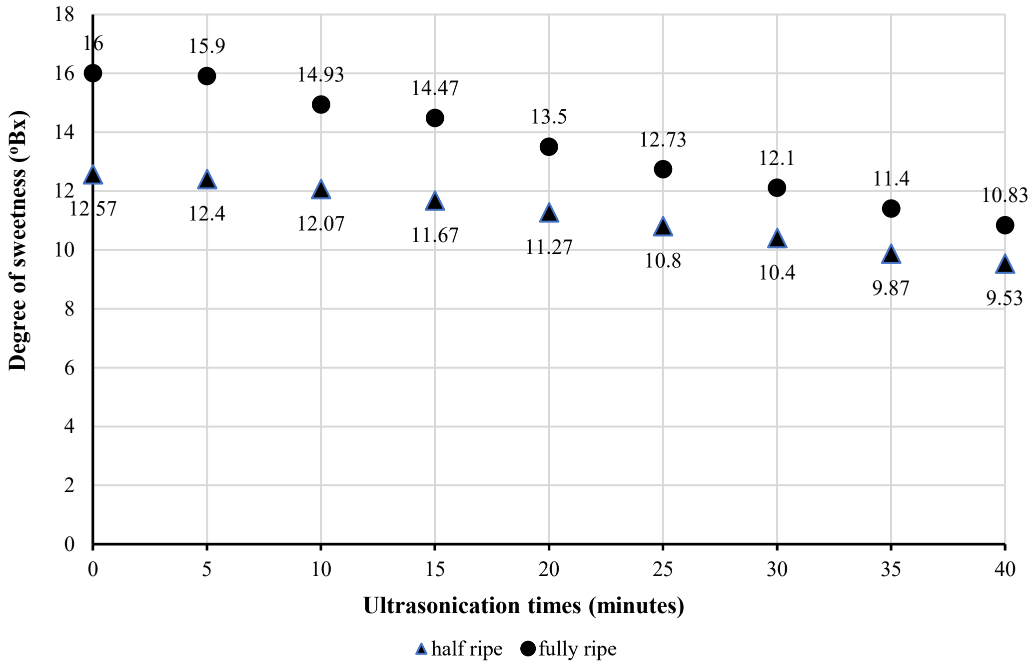

Figure 1 Degree of sweetness of sliced pineapple during ultrasonication

The longer the ultrasonication time, the lower the sugar content. During the ultrasonication, this occurs due to the release of sugar components from the vacuole of the fruit cells. The effect of acoustic cavitation can cause bubbles and pressure which damage or cause cells to burst. Pineapple fruit components, such as sugar, is then released from pineapple cells, thereby reducing the total sugar levels of sucrose, glucose, and fructose of pineapple fruit. During that time, there is rapid formation and breakdown of bubbles in the liquid due to pressure fluctuations. The cavitation effect causes damage to cell tissue and fruit structure which can increase mass transfer within the fruit so that water-soluble components in the cytoplasm of pineapple flesh cells can be removed easily and quickly (Nguyen and Le, 2012).

Analysis of kinetic changes in the degree of sugar of half ripe and fully ripe pineapple Queen variety during ultrasonication followed a first order reaction, as presented in Table 1. A first-order reaction during ultrasonication of pineapple indicates that the rate of reaction is proportional to the concentration of degree of sweetness. In other words, changes in total sugar content depend on the initial concentration before ultrasonication. The reduction of degree of sweetness during the ultrasonication process occurs due to mass transfer process in the pineapple cells. Consequently, chemical components such as sugar and water are as a result of the cells breaking due to the ultrasonication treatment.

The smallest k value reflects the smallest rate of change in degree of sweetness in pineapples. The k values for fully ripe and half ripe pineapples are -0.450 per hour and -0.612 per hour, respectively. The rate of decrease in degree of sweetness of fully ripe pineapples is smaller than that of half-ripe pineapples. This is influenced by the speed of the ultrasonic waves which is reduced due to the increasing level of ripeness of the pineapples, causing differences in the rate of decrease in the total sugar content of the pineapple fruit during ultrasonication. The liquid released from pineapples during ultrasonication was predicted as a mixture of sugar, water, and organic acids. Collectively, this caused a decrease in degree of sweetness in pineapple fruit during ultrasonication. This finding is similar to Salehi et al. (2022), whereby mass transfer due to ultrasonic waves increased water loss of cut apples.

Table 1 Profile of kinetic changes in degree of sweetness during ultrasonication of pineapples

Level of ripeness	Order	Kinetics equation	R^2	k (hour^{-1})
Half ripe	1	$y = -0.0075x + 2.5466$	0.9827	-0.450
Fully ripe	1	$y = -0.0102x + 2.8081$	0.9843	-0.612

3.2 pH changes

The average pH in pineapple ranges from 3.73 to 4.73, as depicted in Figure 2. The analysis of variance showed that level of ripeness of pineapples, and ultrasonication time had significant effects on the pH of the pineapples during ultrasonication. Conversely, the interaction between those factors had no significant effect on the pH of the pineapple fruits during ultrasonication. The significant effect was further analyzed by the Tukey's Honest Significant Difference (HSD) test at $\alpha = 0.05$ to determine treatment pairs that were significantly different.

The pH value increases in ripe fruit because organic acid content is decreased, as a result of the ripening process, where organic acids become a substrate during ripening besides sugar. According to Cordenunsi et al. (2010), pineapple contains organic acids in the form of citric acid, malic acid and oxalic acid which are quite high and are found in the skin, flesh, and pith of pineapple fruits. The content of organic acids in pineapples affects the pH of the fruit. Citric acid is the main organic acid found in pineapples and contributes to the fruit's sweet taste.

The increase in pineapple pH after ultrasonication treatment results from bubbles forming and then bursting quickly which can cause damage to fruit cells. Water soluble organic acids will come out of the fruit cells (vacuoles) as a result of the cells being damaged due to ultrasonication treatment (Ashokkumar, 2015). Figure 2 shows that the pH of pineapples after ultrasonication tend to increase.

During ultrasonication, the process of transferring the chemical components of the fruit occurs, the liquid in the pineapple fruit cells will come out, filling the space inside the polypropylene plastic as a result of the cells being damaged due to the cavitation effect produced during ultrasonication. The fluid that comes out is thought to be a mixture of organic acids, water, and other components. The release of organic acids is thought to cause an increase in the pH of pineapple fruit after ultrasonication treatment. This findings were in accordance with Muzaffar et al. (2016) who reported that the pH of cherries increased after being treated with ultrasonication (frequency 33 kHz) for 0, 10, 20, 30, and 40 minutes. Low frequency ultrasonic waves (20-100 kHz) will form microbubbles around the liquid medium and then explode due to the cavitation effect which will cause damage to the material in cells (Kushwaha et al., 2020).

The pH changes in half-ripe and fully ripe pineapples follow order 1, meaning that the pH changes depend on the initial pH concentration before ultrasonication. This change occurs due to the mass transfer process in the pineapple cells, so that chemical components in the form of organic acids in the cells will come out as a result of the cells breaking due to the ultrasonication treatment. The release of organic acids from the cells can increase the pH of pineapple fruit after ultrasonication.

The largest k value reflects the largest rate of change in pineapple pH. The k value of half-ripe pineapple (0.222) is greater than that of fully ripe pineapple (0.216). The rate of increase in pH of fully ripe pineapples is smaller than half-ripe pineapples. This could be caused by the speed of ultrasonic waves. The higher the level of maturity of the pineapple fruit, the speed of ultrasonic waves propagating in the pineapple fruit will decrease. This causes differences in the rate of increase in pH of pineapple fruits during ultrasonication with different levels of maturity.

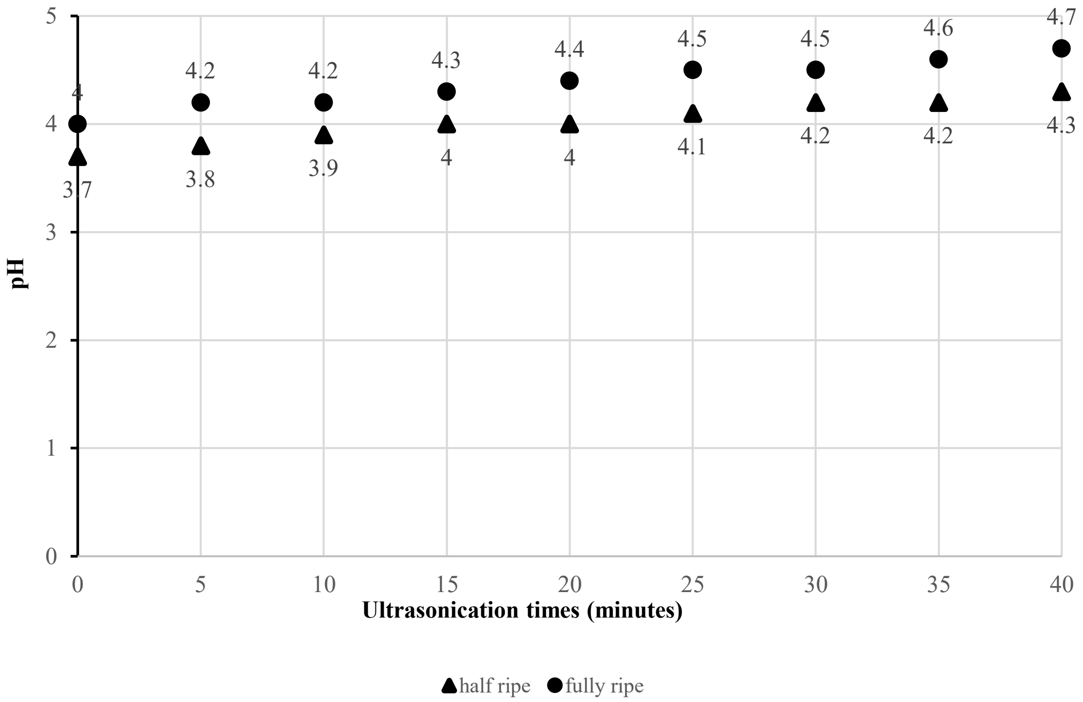

Figure 2 The pH changes of pineapple during ultrasonication time

Table 2 Profile of kinetic changes in pH during ultrasonication of pineapples

Level of ripeness	Order	Kinetics equation	R^2	k (hour^{-1})
Half ripe	1	$y = 0,0037x + 1.3203$	0.9768	0.222
Fully ripe	1	$y = 0,0036x + 1.4041$	0.9812	0.216

4 Conclusions

The level of ripeness and duration of ultrasonication have a significant effect on changes in degree of sweetness and pH of Queen pineapple variety. The interaction between the two treatment factors had a significant effect on changes in degree of sweetness, but was insignificant on changes in the pH of Queen pineapple fruit during ultrasonication. Changes in degree of sweetness and pH of semi-ripe pineapple fruits during ultrasonication follow order 1, with k values of -0.450 hour^{-1} and 0.222 hour^{-1}, respectively. Changes in degree of sweetness and pH of fully ripe pineapple fruits during ultrasonication follow order 1 with k values of -0.612 hour^{-1} and 0.216 hour^{-1}, respectively.

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