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The Prospect of Electrical Impedance Spectroscopy as Non-destructive Evaluation of Citrus Fruits Acidity

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Abstract **— Main objective of the present work was to investigate the Garut citrus fruits acidity using electrical impedance measurements. Furthermore, the measurements of electrical properties of Garut citrus fruits were conducted at various levels of pH and frequencies. This is done as a first step in order to evaluate the Garut citrus fruits acidity that does not damage it. The electrical parameters have significant response to acidity of Garut citrus fruits. The changes of acidity lead to changes in electrical properties of fruits. Resistance per weight, reactance per weight, inductance per weight, and impedance per weight of Garut citrus fuits declined when the pH is increased. While the capacitance per weight of the Garut citrus fruits increases as pH increases. The correlations between pH and electrical parameters are estimated by using multiple regressions equation. Their correlation was highly linear at frequency of 1 MHz. Those estimation equations are the primary basis in nondestructive determining of Garut citrus acidity with electrical impedance measurements.**

Keywords **— Electrical Impedance spectroscopy, Nondestructive evaluation, Fruits acidity, Garut citrus**

I. INTRODUCTION

Nowadays, citrus is a high importance in agriculture and a substantial source of income for several countries. Citrus is primarily valued for humans, which is either eaten alone as fresh fruit or processed into juice, added to dishes and beverages. Citrus has many other uses including animal fodder, craft and oil. Physical specifications of agricultural products are important parameters needed in the design of grading, transfer, processing, and packaging systems. Physical specifications, mechanical, electrical, thermal, light, acoustic and chemical properties are among properties of useful engineering applications [1].

One of the important chemical properties of citrus fruits is acidity. It is generally determined by destructive methods. Therefore it is necessary to explore the new methods of nondestructive evaluation of Garut citrus fruits acidity. Measurement and interpretation of electrical properties give a chance to solve the problem for nondestructive acidity evaluation of Garut citrus fruits.

Electrical measurement provides the opportunity for simple, low cost, and quick assessment of product quality such as moisture and salt content determining in smoking fish [2], soluble solids content of apples [3], or in determination of damage and loss of quality in apples [4]. Furthermore, the electrical properties of the fruits are important in the cognitive aspect, especially to find out responses of the fruits to electric fields of variable frequency. The purpose of the study is to analyze the electrical impedance properties of the Garut citrus fruit at various level of acidity by using low voltage electrical signals that do not damage it.

The measurement of impedance spectra of biological samples is called bioimpedance spectroscopy. The β-dispersion region (1 kHz – 10 MHz) is interesting for the consideration of the cell structures. If frequencies at the top of this dispersion region are chosen, the current flows through the cell. If frequencies in the lower β-dispersion region are selected, the current can only flow through the extracellular space [4]. Electrical impedance spectroscopy (EIS) measures the dielectric properties of materials, such as fruits and other agricultural products, as a function of frequency. It is based on the interaction of an external electric field with the electric dipole moment of materials [5]. EIS has been widely used to assess the in vivo conditions of animal and plant tissue because it is a rapid and easy method of measurement. In this method, Alternating Current (AC) causes polarization and relaxation in the sample leading to changes in the amplitude and phase of the applied AC signals. In biological samples, the proportion of current passing though the apoplastic and symplastic spaces in tissue depends on the AC frequency [6]. Changes of the dielectric parameters of fresh hen eggs during storage were investigated for the development of a method to assess the main quality indices [7, 8]. Ragni *et al.* [9] reported that the analyzed method based on admittance measurements is able to estimate the quality of liquid whole egg products. Zywica *et al.* [10] have found that milk fat content as a key indicator of the quality of raw milk is strongly correlated with the electrical capacitance properties of milk.

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Electrical impedance was used to measure fruit ripening such as the kiwi fruit [11], tomato [12], and nectarine [13]. Also, electrical impedance was used to measure the quality and decay of apples [3, 4], and beef meat ageing [14]. The Q value is a fairly good indicator of the loss of freshness in meat [15]. Vozáry and Benkő [16] reported that the resistance and relaxation time parameters of the second distributed element can be used for characterizing the state of apple flesh.

Afzal *et al.* [17] estimated leaf moisture content by measuring the dielectric constant of leaves in five different types of crops. They used two semi oval isolated copper plates and a Keithly 590 C-V Analyzer as the capacitance measuring instrument, which had the ability of measuring capacitance at two frequencies of 100 kHz and 1 MHz. Nelson [18] measured dielectric properties of fresh fruit, fresh chicken breast meat, and hard red winter wheat. The dielectric constant and loss factor decrease monotonically with increasing frequency, except that the loss factor can increase or decrease with frequency in regions of dielectric relaxation.

II. Materials And Methods

A. Materials

The experiment was carried out on various levels of pH of Garut citrus fruits. The fruits were taken from Garut Regency (7°13'S 107°54'E), in Indonesia. Fruits were picked at ambient temperature. The measurements of all parameters were done when the fruits were still in fresh condition.

B. Acidity and Weight Measurements

The pH of citrus juices provides an idea about the acidity of fruit, and it could be one way of expressing acidity. The pH is an important parameter from a processing point of view [19]. Acidity was determined by using a pH meter (YSI EcoSense pH 100, Xylem Inc., USA). The weight of these citrus was measured by using an electronic balance with an accuracy of 0.01 gram (Sartorius ED 822, Goettingen, Germany).

C. Electrical Measurements and Analysis

A capacitive sensor has favorable characteristics such as robustness, high speed, resistance to the bad environmental conditions, easy operation and low cost [20], so it used in this research. The samples of citrus are placed between two plate conductive electrodes as dielectric material. This method has been done by previous researchers on different products [7, 17, 21]. The conductive plates selected from copper material.

Electrical parameters of citrus fruits were measured by using LCR meter (3532-50 LCR HiTESTER, Hioki, Tokyo, Japan). These parameters are electrical impedance, resistance, reactance, inductance and capacitance. The parameter values of electricity were measured within a frequency of 0.1 kHz, 1.0 kHz, 0.01 MHz, 0.1 MHz and 1 MHz. Each measurement of different shoots repeated four times and then the average is calculated.

In this investigation, the measured electricity was divided by the weight of citrus fruits in an attempt to compensate for the weight variation. It has also been done by Zachariah and Erickson [22] in assessing the maturity of avocado fruits with a capacitance per weight measurement.

For the regression models based on electrical parameters and pH, all data were subjected to regression analysis using the Microsoft Excel program and SPSS 10.0 program. Based on the electrical properties of materials, multiple regressions are divided into two groups. The first group consists of capacitance per weight (C_{wgt}), inductance per weight (L_{wgt}), and resistance per weight (X_{wgt}) of Garut citrus fruits. The impedance per weight (R_{wgt}), resistance per weight (Z_{wgt}), and reactance per weight of Garut citrus fruit are included in the second group.

III. Results And Discussions

A. Dependence of Electrical Properties on Frequency

Typical results showing the frequency dependence of electrical parameters per weight (impedance, resistance, reactance, inductance, and capacitance) for Garut citrus fruits are shown in Fig.1. The figure shows that the all electrical properties of Garut citrus fruits decreased with increasing frequency. Especially in the lower frequency regions, electrical parameters of fruits decreased sharply as frequency increased. As an illustration, it can be reviewed on a sample of fruit that has a pH of 2.86. Decreasing of resistance per weight, reactance per weight, and impedance per weight of Garut citrus fruits at lower frequency are respectively 229.87, 127.032, and 182.794, while at higher frequency are respectively 0.00237, 0.00051, and 0.00052 ohm/ (gram Hz). Declining of inductance per weight and capacitance per weight of citrus at lower frequency are 0.24726 Henry/ (gram Hz) and 0.00032 pF/ (gram Hz), while at higher frequency are 8.5E-10 Henry/ (gram Hz) and 1.2E-7 pF/ (gram Hz).

If the frequency is enlarged, the rate of direction changes of current in the external electrical circuit will be faster. This is the external condition of the electrical signal that will affect the internal conditions of Garut citrus fruits, especially on the mobility of electric charge.

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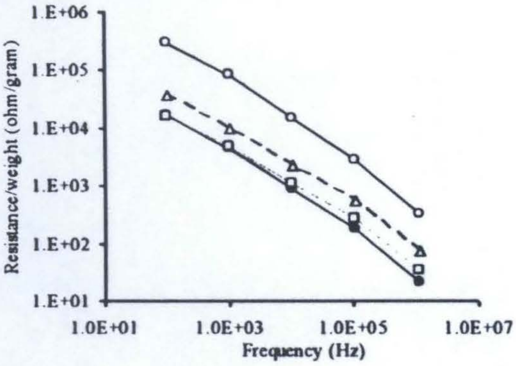
Frequency changes will affect the condition of the ions in the material. Ionic loss is inversely proportional to frequency and become critical as we go to the lower frequency. High values of electrical impedance, resistance or reactance of citrus at low frequency (Figure 1a, 1b, and 1c) could be attributed to high mobility of dipole due to free water state and electrode polarization. Dissipation of dipolar energy at higher frequencies is less dominant and ionic loss become almost absent [23].



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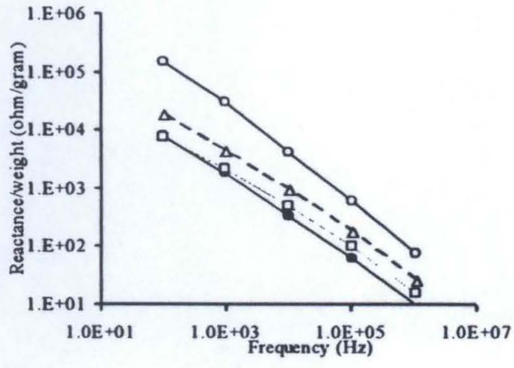

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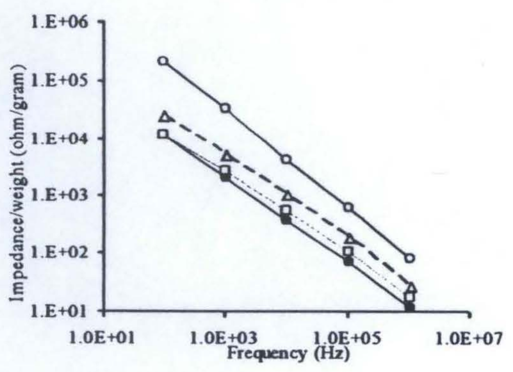

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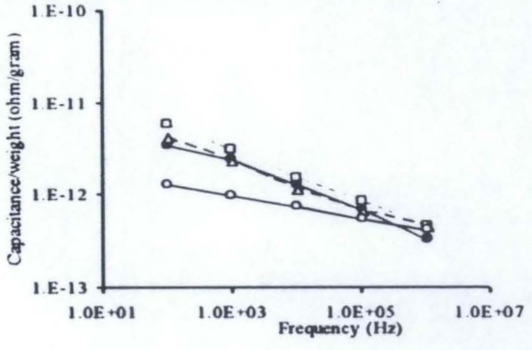

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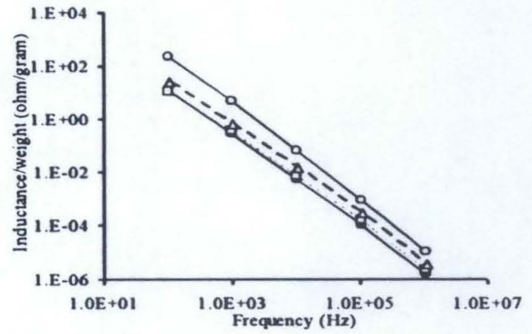

(e)

Figure 1. The electrical resistance (a), reactance (b), impedance (c), capacitance (d), and inductance (e) per weight of Garut citrus at selected acidity. Legend of (—◆—) for pH 4.49, (—□—) for pH 4.06, (—△—) for pH 3.47, and (—○—) for pH 2.86.

Total polarization processes occurring in the material require sufficient time. With the high frequency causes a short time in the process of polarization, so that the polarization does not occur completely. Thus, the increasing frequency values will lead to the decrease in the total polarization. This caused the low polarity of the materials. This is being the reason for capacitive phenomenon that occurs in Garut citrus fruits (Figure 1d). For proper description of the dielectric behavior of the Garut citrus fruits and other biological materials, contributing phenomena other than dipolar relaxation also need to be taken into account, such as the ionic conduction at lower frequencies, the behavior of bound water, and the influence of constituents other than water.

Arising from Faraday's law, the inductance may be defined in terms of the emf generated to oppose a given change in current. At low frequency conditions, opposition to change in the current flow has a fairly high value, while at high frequency has a relatively low value (Figure 1e). This is consistent with the properties of materials devoted to the impedance, reactance or resistance.

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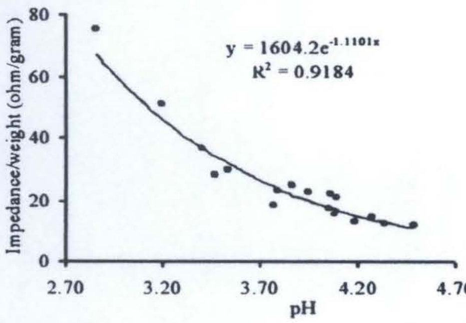
Garut citrus fruits show resistive properties at low frequency, so it is a poor conductor. Thus, the ions and electrons in the Garut citrus fruits are bound relatively stable at low frequency.

B. Dependence of Electrical Properties on Fruits Acidity

Fruit pH is a hydrogen-ion concentration and provides an estimate of the extent of acidity in the fruit juice. Garut citrus fruits have a pH range of 2.86 – 4.5. As in most of the fruits, the maturity of Garut citrus fruits heralds the accumulation of the loss of acidity and host of other biochemical changes. Increased maturity of the citrus fruit will cause the acidity is reduced and pH will increase. The decrease in acidity or increasing in pH was considered to be due to dilution as the fruit increased in size and juice content [19].

The high total acid correlated with low pH or high concentration of hydrogen ions. The pH value depends on the type of substance that is dissolved in water and its concentration. If the pH is low, it means that the substances contained in water are dominated by the acids. Organic acids are respiratory substrates in the fruits. Higher respiratory quotient indicates utilization of acids, mainly citric and malic acids through the tricarboxylic acid cycle, in which acids are oxidized and ATPs are formed for synthesis of new compounds [19].



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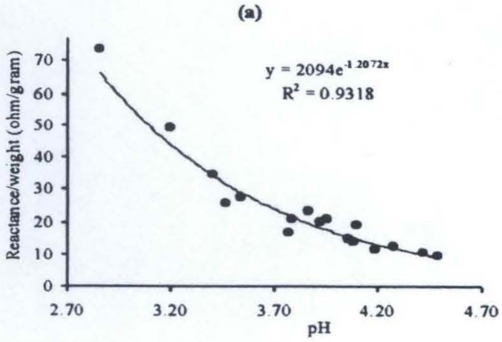

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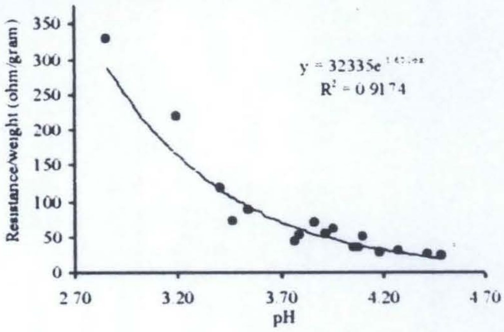

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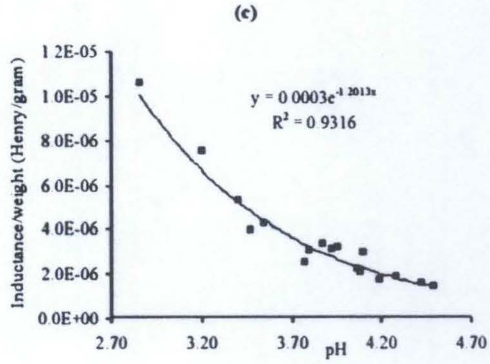

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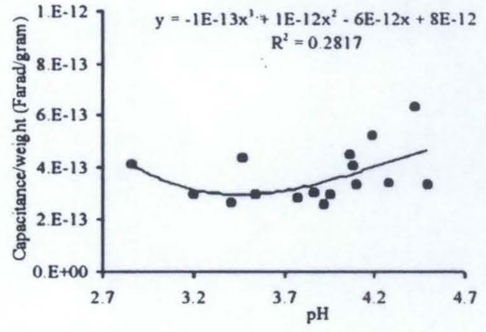

(e)

Figure 2. The effect of pH on the impedance per weight (a), reactance per weight (b), resistance per weight (c), inductance per weight (d) and capacitance per weight (e) of Garut citrus at 1 MHz.

The results showing an electrical dependence with acidity of Garut citrus fruits are disclosed in Figure 2. All of the samples fruits have same phenomenon that the decreases of electrical parameters per weight as increasing of pH. The correlation of each electrical parameters and pH did not show a linear. The best correlation equation is exponential form. All of deterministic coefficients are fairly good especially at frequency of 1 MHz (Figure 2) except on capacitance parameter, so that the correlation between electrical parameters and pH are relatively good.

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This phenomenon is possibly happened because the AC current can flow and penetrate to the sample more deeply when the signal frequency is high enough. The current flows through the cell if frequencies at the top of β-dispersion region are chosen. If frequencies in the lower β-dispersion region are selected, the current can only flow through the extracellular space [4].

The capacitance per weight of fruit showing in Figure 2 does not show a strong correlation and has a different profile with other parameters. Increase in pH values is accompanied by an increase in the electrical capacitance per weight of Garut citrus fruits.

All the constituent parts of citrus, i.e. seed, segment, peel, etc., has contributed to the electrical properties. Although the segment of citrus was a lot of water content and acidic, but it's was in a sealed condition. So the important rule of ion mobility defends on cell membrane and wall permeability. A decrease in fruit impedance, resistance, inductance, and reactance may be related to increased concentration of mobile ions in cell wall and/or increase in cross-section of the cell wall accessible to low-frequency current. Cell wall resistance declined as freshly harvested fruit ripened, and this decline was closely related to changes in fruit texture [24]. Most fresh fruits and vegetables are high in water, but normally have very poor electrical conductivity because much of the water is held immobile, trapped within the cells and the intercellular spaces within the tissue structure of these plant materials [25]. Landaniya [19] reported that the more granulation developed, the more the electrical conductivity increased. Increased conductivity and granulation have the same meaning with the decreases in resistivity and increasing cross sectional area of the membrane cell. It was support in understanding the phenomenon obtained in this study. The decreases in acidity (indicated by an increase in pH) occurs when fruit growth and maturation. In addition, there is also growth of granulation.

The changes of tissue resistance indicate the changes in mobility of ion in cell and physiological changes of fruits during ripening [26]. It is enough reasonably related to a decrease in impedance properties when the pH increased.

C. pH Estimation Using Electrical Parameters

In order to estimate the pH (acidity) of the fruit by using the electrical parameters, multiple correlation equation between them should be caught early. In this case, the SPSS program for multiple linear regressions was used. So, the pH value could be predicted by using the electricity parameters of Garut citrus fruits.

Multiple linear regression equations for pH prediction using the electrical parameters of the citrus fruits at various frequencies are shown in Table 1. Also, the linearity between pH predictions and experiments are shown in Figure 3 for the first group (C_{wgt}, L_{wgt}, and R_{wgt}) and Figure 4 for the second group (R_{wgt}, Z_{wgt}, and X_{wgt}). The level of linearity is showed by the point distribution of datas on graph of $y = x$ equation.

By reviewing its deterministic coefficients (R^2) and standard error of estimations (SE), those equations give a fairly good estimate (Table 1). The lowest correlation for the both of group was occurring at a frequency of 0.1 kHz (Figure 3a and 4a). The highest correlation for the first group occurred at a frequency of 1 MHz (Figure 3b) and the second group occurred at a frequency of 0.01 MHz (Figure 4b).

According to the average value of deterministic coefficients and standard error of estimations, the best correlation occurred at a frequency of 1 MHz which is characterized by the highest deterministic coefficient ($R^2_{average} = 0.9255$) and lowest standard error of estimation ($SE_{average} = 0.1323$). So that equation can be used as a reference to estimate the pH value of Garut citrus fruits using electrical measurements, as a nondestructive test, to the next post-harvest handling.

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Table 1
Multiple regressions for pH estimation using electrical parameters of Garut citrus fruits.

Frequencies	Equations of estimation	R^2	$R^2_{average}$	SE	$SE_{average}$
0.1 kHz	$pH = 0.0000493R_{wgt} + 0.0001314X_{wgt} - 0.000031Z_{wgt} + 4.182$	0.685	0.663	0.2724	0.2817
	$pH = -0.0000166R_{wgt} + 0.01475L_{wgt} + (2.5E+09)C_{wgt} + 4.113$	0.641		0.2909	
1.0 kHz	$pH = 0.0001349R_{wgt} - 0.0002190X_{wgt} - 0.0001890Z_{wgt} + 4.411$	0.822	0.806	0.2049	0.2138
	$pH = 0.0001313R_{wgt} - 2.682L_{wgt} - (1.1E+10)Z_{wgt} + 4.424$	0.790		0.2226	
0.01 MHz	$pH=4.531E-04R_{wgt} -7.980E-06X_{wgt} -1.990E-03Z_{wgt} +4.661$	0.93	0.9205	0.1288	0.1369
	$pH = 5.055E-04 R_{wgt} -1.492E+02 L_{wgt} -1.000E+09 C_{wgt} + 4.652$	0.911		0.1450	
0.1 MHz	$pH = 0.001154R_{wgt} + 0.009648X_{wgt} - 0.01810Z_{wgt} + 4.714$	0.894	0.900	0.1581	0.1535
	$pH = 0.001588R_{wgt} - 735.4L_{wgt} - (8.2E+09)Z_{wgt} + 4.788$	0.906		0.1489	
1 MHz	$pH = 0.01449R_{wgt} - 0.2.270X_{wgt} + 0.1310Z_{wgt} + 4.724$	0.917	0.9255	0.1401	0.1323
	$pH = 0.0125R_{wgt} - (5.484E+05)L_{wgt} - (8.6E+10)Z_{wgt} + 4.971$	0.934		0.1245	

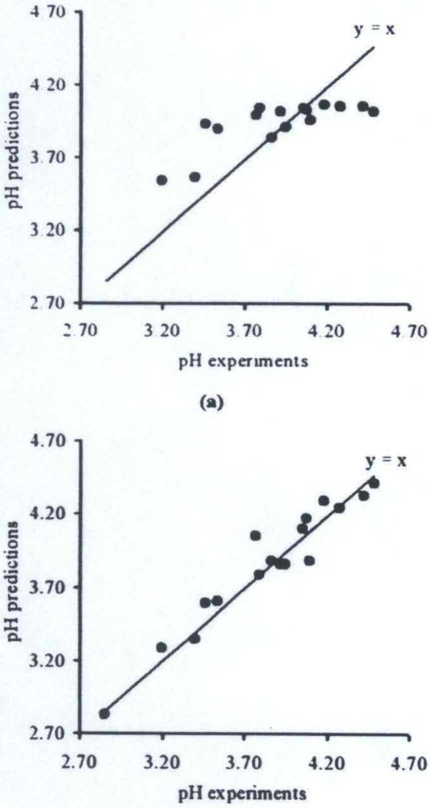

Figure 3. The linearity of pH predictions and experiments of Garut citrus using electrical inductance, resistance and capacitance per weight parameters at frequency of 0.1 kHz (a) and 1 MHz (b).

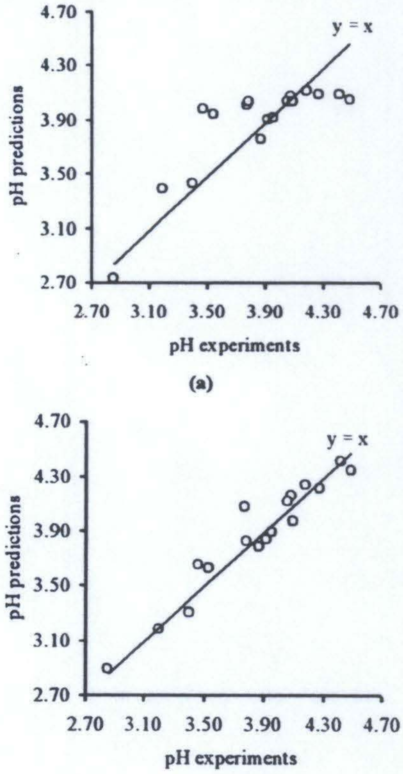

Figure 4. The linearity of pH predictions and experiments of Garut citrus using electrical impedance, resistance and reactance per weight parameters at frequency of 0.1 kHz (a) and 0.01 MHz (b).

IV. CONCLUSIONS

Electrical impedance measurements were able to determine the acidity of Garut citrus fruits and it depends on the frequency. The electrical parameters have significant response to acidity of Garut citrus fruits. The pH estimations by using multiple regressions of electrical parameters were highly linear especially at frequency of 1 MHz. Thus, the electrical impedance measurements of Garut citrus fruits have a high potential for determining the acidity of the Garut citrus fruits that is not destructive. So it becomes an alternative technique to complement and even replace existing tools. The results of this study can be utilized and further developed by agricultural technicians, farmers, chemists, other researchers and the industry to become more useful.

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