Abstract—In the present work the dielectric constant for the identification of water addition in raw milk is studied. To achieve this objective, a parallel plates capacitive cell was used, and a LCR Meter was used to measure the capacitance. The milk samples were obtained on the Upaon-Açu Island, which were diluted with water from the University supply in known proportions. Samples of approximately 120ml were used. The measurements were performed in the frequency range between 100 Hz and 100 kHz with a voltage of 1V, with a temperature of approximately 8°C. The software used for statistical analysis was SPSS version 25.0. From the results it was observed that all the frequencies presented significant differences between the groups, except the frequency of 100 Hz (p = 0.06). In the frequency of 1 kHz and 20kHz the addition of water causes a decay of the dielectric constant almost linear, whereas in the others frequencies the decay occurs smoothly, except in 10kHz, which presents a different behavior. With the obtained results, it is concluded that the dielectric constant method is feasible since it becomes possible to quickly identify the presence of water in the milk.

Index Terms—dielectric constant, raw milk, water

I. INTRODUCTION

The milk is one of the most complete foods present in the human alimentation, having in its composition a lot of compounds that are essentials for nutrition, such as proteins, lipids, carbohydrates, vitamins and mineral salts. The presence of these elements give to milk a high biological value, making it a susceptible environment to action of microorganisms if the milk is not properly acquired, processed and stored. The milk also can suffer additions in its composition, by water, density reconstituiners, neutralizers, preservatives, synthetic milk and etc. [1]-[5].

Nowadays the adulteration on dairy products are widely studied, to search to guarantee a safe product for the consumer, being very important to have a total control from milking to the processing and storing, since the added of substances not permitted by legislation can be made in any stage of production [6], [3].

The main problem in detecting adulteration in milk today, is the need for physico-chemical analyzes in laboratories with equipment and reagents that demand a high cost, since each type of adulteration has a specific type of detection [7], [5]. In addition, with new techniques of adulteration emerging, new methods for detecting adulterants should be developed to demonstrate high potential for determination milk quality and / or authenticity. Thus, the dielectric constant (K), which is an electric property present in all materials, whether solid or liquid, is defined as the ratio between the capacitance of the dielectric (C) and the capacitance of the air (Co) - K = C / Co, allows to obtain information on the composition of the material, from how it behaves in the presence of an electric field, and can be an alternative for the analysis of the constituents in milk [8].

The parallel plate capacitor was used in several studies to identify adulteration in gasoline, and for dielectric study of complex liquids obtaining results that can facilitate the quality control of these products [9], [10]. In the area of dairy products, there are works for the application of capacitance in the monitoring of microbial growth and in other, electric parameters such as admittance, conductance, impedance, resistance and capacitance were also studied to detect the addition of water in milk [11]-[14]. In this context, we have been studying increasingly methods of detecting the addition of adulterants to milk so that these measures can be fast, cheap, and reproducible in order to guarantee the purity of the milk acquired. Thus, the objective of the present study is to evaluate the efficiency of the dielectric constant in the differentiation of the raw milk from the diluted milk in different fractions after addition of water.

II. EXPERIMENTAL DETAILS

The capacitive sensor used was formed by two parallel stainless steel circular plates with radius of 25mm, these were placed inside a rectangular box also of stainless steel separated by 1mm of distance. The capacitance was measured through the Agilent 4263B LCR Meter, in the frequency bands available in this model (100Hz, 120Hz, 1kHz, 10kHz, 20kHz and 100kHz), the voltage used was 1V, since low values do not cause changes in the measurements [2]. A double terminal was used to connect the parallel plates to the LCR meter. The equipment has been calibrated for capacitance measurement (C). The assembled system for measurement is shown in Fig. 1.
The samples of raw milk were obtained of two places in the Upaon-Açu Island in the state of Maranhão-BRZ. The samples were diluted with water supply from the Federal University of Maranhão, since in the research this water is most used for dilute the milk [15]. Subsequently, they were stirred carefully to homogenize the water with the milk to avoid the formation of bubbles that could cause changes in the capacitance measurement. The samples were diluted in the following proportions (identified by their respective groups): 0% - 0; 15% - 1; 30% - 2; 45% - 3; 50% - 4; 60% - 5; 75% - 6; 90% - 7.

The samples \( (n = 307) \) with approximately 120ml were introduced into the capacitive cell so that they completely covered the circular plates, and then the apparatus was placed inside a thermal box with two ice packs, so that the temperature remained constant in approximately 8° C.

The temperature was measured at the beginning and at the end of the measurements with a spit-type digital thermometer, the temperature averaged was calculated to obtain the measurement temperature. After each analysis the cell was washed with diluted detergent and rinsed with water for approximately 1 min. And before the next sample was introduced into the cell, it was set with the sample itself to be used.

The value of the dielectric constant \( (K) \) was obtained from the relation between the sample capacitance \( (Ca) \) and the empty cell capacitance \( (Co) \) - \( K = \frac{Ca}{Co} \). For the statistical analysis, SPSS software (Statistical Package for the Social Sciences, Inc., Chicago, IL, USA) version 25.0 was used. Data was treated using descriptive procedures (median and interquartile range). The Komolgorov Smirnov (KS) test was used to verify the normality of the data. The Mann - Whitney U (MWU) and Kruskal - Wallis (KW) H test were used for comparison between the groups, for the variables that did not present normal data distribution. The results were considered statistically significant if \( p <0.05 \).

### III. RESULTS AND DISCUSSION

There was no significant difference in the comparison between the groups \( (0 \text{ to } 7) \), at the frequency of 100 HZ. Therefore, this frequency was taken from the analysis of the present study, using only the frequencies of 120 Hz to 100 kHz.
According to [7], frequencies lower than 1kHz have characteristics of the sensor’s polarization, and the frequencies of 1 kHz to 1 MHz show the milk physical characteristics of the milk. And as shown in Fig. 2 the frequency of 1 kHz and 20 kHz have a noticeably more linear inclination compared to the other frequencies. It was chosen in this case, by specifically study the frequency of 1 kHz which would be a frequency within the indicated range [7].

In the frequency of 1kHz the values of the dielectric constant of group 1 did not present significant difference in relation to group 0 as shown in Table I. In Fig. 3 it can be observed that the temperature does not remained constant, being able to be responsible for the increase of K in group 1. This aspect is discussed by [7], where it is reported that with the difference of only 1° C the conductance can be modify by up to 5%, at the frequency of 100kHz.

Although the temperature did not remain constant in all the groups, it remained in a range of 8-9°C. After statistical analysis (Table II) of the groups in relation to temperature, it was observed that the temperature does not remained constant, being able to be responsible for the increase of K in group 1. This aspect is discussed by [7], where it is reported that with the difference of only 1° C the conductance can be modify by up to 5%, at the frequency of 100kHz.

As shown in Fig. 2, the addition of water causes a decrease in dielectric constant values at all frequencies, presenting an anomaly only at the frequency of 10 kHz. This decreasing behavior of K with the water addition was also verified by [7] and [15], in the electrical admittance and electrical conductivity characteristics respectively, demonstrating that this method can be used to determine adulteration in milk.

| TABLE I. DIELECTRIC CONSTANT (K) AT 1kHz FREQUENCY, STRATIFIED BY GROUP |
|-----------------------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| K (Dielectric Constant) in 1kHz | Groups       | 0           | 1           | 2           | 3           | 4           | 5           | 6           | 7           |
|                             |              | 2.56E-12 (2.150E-12 – 2.751E-12) | 2.47E-12 (2.068E-12 – 2.615E-12) | 2.05E-12 (1.725E-12 – 2.117E-12) | 1.54E-12 (1.361E-12 – 1.618E-12) | 1.36E-12 (1.216E-12 – 1.438E-12) | 1.07E-12 (0.974E-13 – 1.135E-12) | 6.10E-13 (5.49E-13 – 6.81E-13) | 2.07E-13 (1.82E-13 – 2.37E-13) | <0.001
|                             |              | 2.56E-12 (2.150E-12 – 2.751E-12) | 2.47E-12 (2.068E-12 – 2.615E-12) | 2.05E-12 (1.725E-12 – 2.117E-12) | - | - | - | - | 0.050
|                             |              | 2.56E-12 (2.150E-12 – 2.751E-12) | - | 2.05E-12 (1.725E-12 – 2.117E-12) | - | - | - | - | <0.001
|                             |              | - | 2.47E-12 (2.068E-12 – 2.615E-12) | 2.05E-12 (1.725E-12 – 2.117E-12) | - | - | - | - | <0.001

*From group 2 all the others had a significant difference with group 0. # All other groups showed significant differences between them. **Kruskal-Wallis H. values are presented as median (interquartile range 25-75); # Mann – Whitney U. values are presented as median (interquartile range 25-75)

| TABLE II. TEMPERATURE DIFFERENCE TABLE OF ALL GROUPS AND PAIRS OF GROUPS |
|-----------------------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Temperature                 | Groups       | 0           | 1           | 2           | 3           | 4           | 5           | 6           | 7           |
|                             |              | 8.375 (7.500 – 9.1625) | 9.2750 (8.6375 – 10.1000) | 9.1500 (8.2750 – 9.4750) | 8.8750 (8.2375 – 9.6125) | 8.9500 (7.9000 – 9.6125) | 8.4500 (8.0000 – 9.3000) | 8.3500 (7.7500 – 9.6625) | 8.7250 (8.1750 – 9.6625) | >0.05
|                             |              | 8.375 (7.500 – 9.1625) | 9.2750 (8.6375 – 10.1000) | 9.1500 (8.2750 – 9.4750) | - | - | - | - | 0.002
|                             |              | 8.375 (7.500 – 9.1625) | 9.1500 (8.2750 – 9.4750) | - | - | - | - | 0.042
|                             |              | 8.375 (7.500 – 9.1625) | 8.8750 (8.2375 – 9.6125) | - | - | - | - | >0.05

*From group 3, all the others had not a significant difference with group 0; **Kruskal-Wallis H. values are presented as median (interquartile range 25-75); # Mann – Whitney U. values are presented as median (interquartile range 25-75)
IV. CONCLUSION

The present work presented a technique to detect adulteration of the raw milk from the value of the dielectric constant. For this, a capacitive cell of parallel plates was used to obtain capacitance measurements.

The results showed a significant difference in the median of the dielectric constant, between the groups, in the frequency of 1kHz, reinforcing that the addition of different contaminants causes a decrease in this value. The frequencies not evaluated will be studied in the next stage of the work. Based on these results it is still intended to carry out future studies with other adulterants, such as sodium hydroxide, urea and hydrogen peroxide, for example.

A very important step, to be structured, is the constant temperature maintenance, so that all measurements are carried out with greater control and precision. It should be further evaluated how the K measurement varies with temperature, so that a correction factor could be developed according to the temperature of the sample at the time of analysis.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Vanessa was the principal investigator of this work and wrote this paper. She had the supervision of Dr. Allan and Dr. Daniel during all research, cell construction, organization and statistical analysis of the data. Dr. Francisco Sávio participated in obtaining the data, providing the space, the necessary equipment and help. All team authors approved the final version of article.

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