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Modelling the Dielectric Properties of Cow's Raw Milk under Vat Pasteurization

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Abstract—An efficient microwave milk pasteurization system requires a rigorous temperature dependent dielectric model of the milk, since the performance of milk pasteurization strongly depends on its dielectric properties. This paper describes the dielectric modelling of cow's raw milk during batch (Vat) pasteurization which covers the frequencies from 0.2 GHz to 6 GHz. An open-ended coaxial sensor is used for the measurements of dielectric constant, loss factor, and ionic conductivity at temperature range of 25°C to 75°C with an interval of 5°C. Combinations of Cole-Davison and Debye equations are modified to fit the dielectric measurements. It was found that the measured dielectric constant decreased as the frequency increased, while the high temperature processed produce lower in a convergence manner toward 6 GHz. The loss factor exhibited high losses at higher temperature and lower frequencies, as well as converged at 1.9 GHz then diverged up to 6 GHz. Three relaxation processes are dominated at all temperature treatments within the frequency range. The relaxation time, τ, and the activation energy, Q, are modelled based on linear fitting of measured data according to Debye and Arrhenius approaches.

1. INTRODUCTION

Milk is an important source of protein with rate of (32 g/liter) which is an important source of amino-acids and other bio-active components, as well as calcium at rate (1.1 g/liter) which is essential for bones, blood control, and teeth [1]. Raw milk is often not recommended to be consumed directly due to its high probability of getting contaminated with micro-organisms by the cow herself, pasture, milking machine, and containers. Hence, pasteurization process of the milk is mandatory to ensure the safety, as well as to prolong shelf-life of milk. In fact, pasteurization is mandatory according to Pasteurized Milk Ordinance (PMO) standards which are established by the Food and Drug Administration (FDA), where only certified milk products are permitted for shipping and selling abroad. Thermal treatment of milk reduces the number of viable spoilage micro-organisms and inactivates undesirable enzymes [2]. The drawbacks of current commercial pasteurization machines exhibit fouling in heat exchangers, causing inefficient pasteurization process; consequently, the demand of using emerging technologies to overcome such problems gets increased [3]. Microwave heating is considered as one of the promising alternatives, and it plays an important role in the progression of pasteurization process due to its characteristics of rapid penetration through food materials which increases the rate of milk production in the industrial sector [4]. However, the development of a precise microwave-based pasteurization system requires an accurate modelling of dielectric properties of milk over wide variety of thermal treatment. Hence, modelling dielectric properties helps in selecting the optimal frequency ranges, because it covers up to optical frequencies [5]. That aside, dielectric heating highly depends on power absorption which depends on penetration depth and temperature profile during microwave heating. At the time of writing, few researches have been reported for the determination of milk's dielectric properties and its model. First,

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a study on dielectric properties of milk has been conducted from 1 to 20 GHz at temperature range of 17°C to 20°C [6]. Furthermore, a study showed the milk dielectrics at three points of frequency (0.3 GHz, 1 GHz, and 3 GHz), and four points of temperature 25°C, 35°C, 45°C, and 55°C [7]. The dielectrics were measured at four points of frequencies namely 27.12, 40.68, 915, and 2450 MHz, respectively [8]. Recently, a research reviews the measurements of dielectric properties of cow's raw milk at two industrial frequencies 915 MHz and 2.45 GHz [9]. The latest recorded measurement on dielectric properties of cow milk was at high temperature treatment (up to 70°C at 2.45 GHz) [10]. An effective model of dielectric properties is important in order to understand the behaviour of temperature distribution which ease the industrial selection of suitable waveguide solution. There are available commercial microwave waveguide systems particularly for those operating frequencies, such as waveguide models of WR975 and WR430/WR340 for 915 MHz and 2.45 GHz, respectively. In addition, the mentioned two microwave frequencies are allocated by the US Federal Communications Commission (FCC) for industrial, scientific, and medical (ISM) applications. Normally, domestic microwave heating and some industrial applications are located at operating frequency of 2.45 GHz. On the other hand, 915 MHz is preferred for industrial/commercial microwave heating due to its features of being suitable for large volumetric heating capacity and high penetration depth as compared with 2.45 GHz systems. However, the reported researches did not take into account the wide range of temperatures at three operating frequencies which affects the model of milk dielectric properties. In addition, the corresponding relaxation time and activation energy models were not been covered. Hence, this research aimed to measure and model the dielectric properties of the milk at eleven points of temperature from 25°C to 75°C at frequency ranging from 0.1 to 6 GHz. The mathematical model was developed by using a combination of modified Cole-Davidson and Debye processes. In addition, the relaxation time, τ, and activation Energy, Q, were extracted and precisely modelled.

2. MATERIALS AND METHODS

2.1. Milk Collection

Fresh (raw) milk samples were collected from local cow's farm in Johor, Malaysia. The milking time was in early morning, and the milk was stored in icebox of 3 to 6°C for 4 hours prior to the initiation of dielectric measurements. The measured pH value of milk was 6.6 at 4°C before measurements. The milk samples contained $3.8 \pm 2\%$ of fat, $2.8 \pm 1.5\%$ of protein, and $8.5 \pm 0.5\%$ of non-fat solids.

2.2. Dielectric Properties

The interaction between the material and microwave signal is represented by two mechanisms, namely energy storage or absorption and transformation of the absorbed energy into heat. Such mechanisms can be quantitatively evaluated using dielectric properties of the material, represented by its relative complex permittivity, $\varepsilon_r (= \varepsilon'_r - j\varepsilon''_r)$ in Equation (1), where ε'_r is the real part of permittivity so called dielectric constant, and ε''_r is the imaginary part of the permittivity, namely loss factor. Since the main chemical constituent of the milk is water, the milk is classified as polar solvent material, and its value of ε'_r can be modelled using Debye polar model [11].

$$\varepsilon_r = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + j\omega\tau} \tag{1}$$

where ε_s, ε_∞, and τ are the static permittivity, optical permittivity, and relaxation time, respectively. Although Debye model is used to describe pure polar molecules [12], non-linearity always exists in all polar materials. For this reason, wide variety of polar dielectric models have been modified based on Debye, such as Cole-Cole, Cole-Davidson, Havriliak-Negami models or a combination of them to conform the specific dielectric measurements [13].

2.3. Dielectric Measurements

The dielectric measurements were achieved using a Keysight 85070E dielectric probe connected to a Keysight E5071C Network Analyzer (formally Agilent Technologies) via coaxial cable and controlled

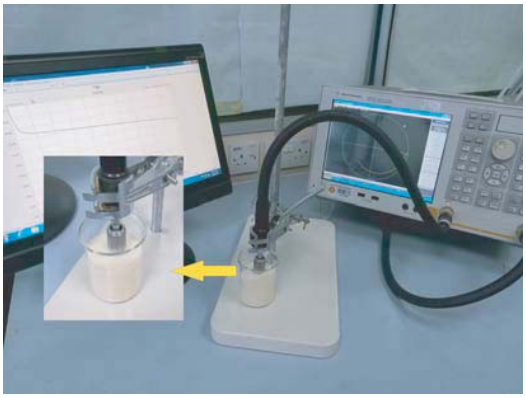

Figure 1. Measurements setup.

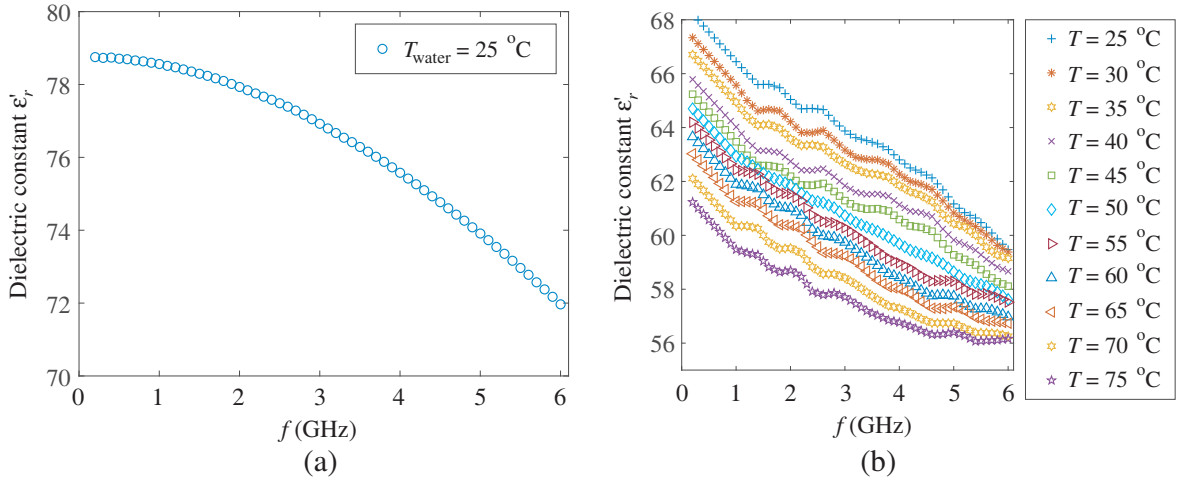

Figure 2. Shows the measured dielectric constant, ε_r' of (a) water and (b) cow's raw milk at various temperature, T.

by computer-based software as shown in Figure 1. The probe calibration was performed at the probe aperture using three dielectric calibration references: air, metal short, and deionized water at (25±1)°C, respectively. The milk samples were thermally processed using digital water bath based on Fisher Scientific Isotherm Heated Magnetic Stirrer to ensure uniform heat transfer after the calibration has been made, and the probe was diagonally immersed into the milk and totally covered the entire area of the probe aperture to ensure that there were no bubbles attached into the aperture of the coaxial probe as shown in Figure 1. The measurements were repeated at each temperature step from 25°C to 75°C starting the measurements at +0.5°C from the desired temperature to compensate the cooling margin associated with the probe interface area and to ensure that the measurements were at the desired temperature, T. The stable and uniform temperature, T, distribution of the raw milk samples was controlled by a water path surrounding the sample's container. The temperature, T, readings were monitored by thermocouple connected to Fluke 287 multi-meter. The measured dielectric constant, ε_r', and loss factor, ε_r'', are respectively shown in Figure 2(b) and Figure 3. The dielectric data in those figures are the averaged values of four repeated measurements at each temperature value. In fact, the dielectric data may slightly vary due to several environment and milk processing factors, such as season, different diary farm, milking time, cleanliness of food and dairy equipments, and storage temperature prior to measurements.

Figures 2(b) and 3 present the measured dielectric constant and loss factor of cow's raw milk at wide range of pasteurization temperatures (25 to 75°C), respectively. Figure 2(a) presents the results

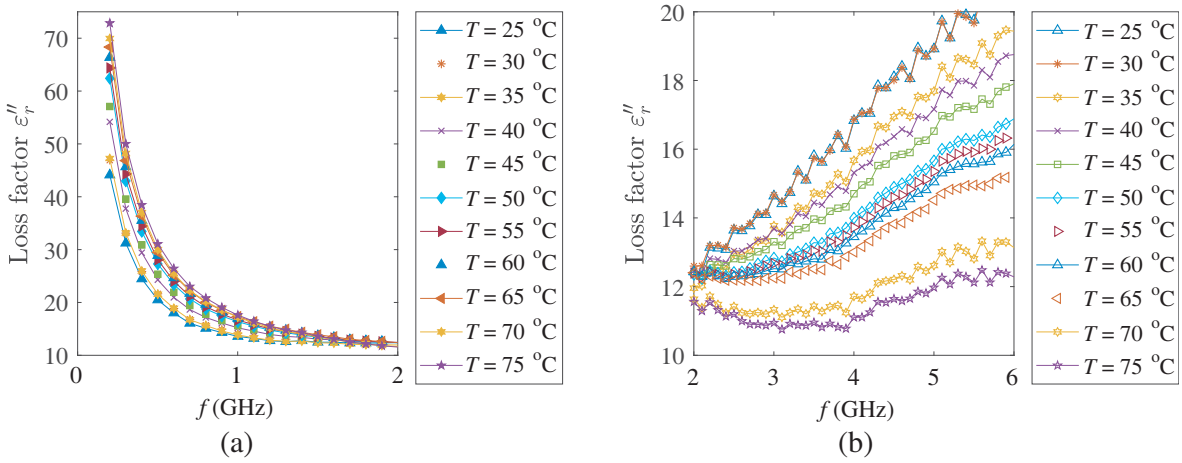

Figure 3. Measured dielectric loss factor, ε_r'' of raw milk at temperature range of 25°C to 75°C from (a) 0.2 GHz to 2 GHz and (b) 2 GHz to 6 GHz.

of dielectric measurement of deionized water. This indicates the linearity domination of water content of milk on its dielectrics. The dielectric constant, ε_r'', of milk is linearly decreased with temperature, T as well as the frequencies, f. However, the increase in the high temperature causes an increase of intermolecular vibrations. Consequently, the hydrophobic interactions of milk's protein will be increased which causes an interruption to the ordered water molecule arrangements that causes reduction in dielectric constant [14–16]. Figure 3 shows the loss factor, ε_r'', of cow's raw milk. The loss factor, ε_r'', at low frequencies ($<$ 1900 MHz) is higher as the temperature goes high and decreases exponentially over such a frequency range. These dipole losses are caused by water dipole rotation, while the higher temperature causes decrease in loss factor, ε_r'' at frequencies ($>$ 1900 MHz up to 6 GHz). This is due to ionic losses that cause migration of ions. The ionic conductivity, σ, is strongly affected by the temperature especially at lower frequencies, and physically it depends on the presence of salts in the milk. The variation in measurements is substantially affected by milk compositions and the cleanliness of milking and storage temperature.

3. MATHEMATICAL MODELLING

Although milk is a polar solvent material, and Debye relaxation is an appropriate model for pure polar solvent materials, the milk does not only consist of pure polar molecules, due to its compositions of other nonpolar materials than water. Usually the pure polar relaxation model of Debye describes that the molecules of dielectric material tend to gravitate exponentially to an equilibrium state. According to the measurements as shown in Figures 2 and 3, a set of three relaxation processes are dominated in the pasteurization of cow's milk as shown in Figure 4. In addition, the losses effect caused by ionic conductivity, σ, which is mainly presented at lower frequencies ($<$ 1900 MHz), can be determined using Equation (2).

$$\varepsilon_r'' = \frac{\sigma}{2\pi\varepsilon_o f} \tag{2}$$

Finally, by considering the ionic conductivity, σ, the model is able to handle the loss factor, ε_r'', at low frequencies.

$$\varepsilon_r = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_1}{1 + j\beta_1\omega\tau_1} + \frac{\varepsilon_1 - \varepsilon_2}{1 + j\beta_2\omega\tau_2} + \frac{\varepsilon_2 - \varepsilon_\infty}{1 + (j\omega\tau_3)^{\beta_3}} - \frac{j\sigma}{\omega\varepsilon_o} \tag{3}$$

Equation (3) presents the mathematical model corresponding to the best possible fit of the measurements. The coefficients of the developed model have been extracted and optimized based on fitting with measured data.

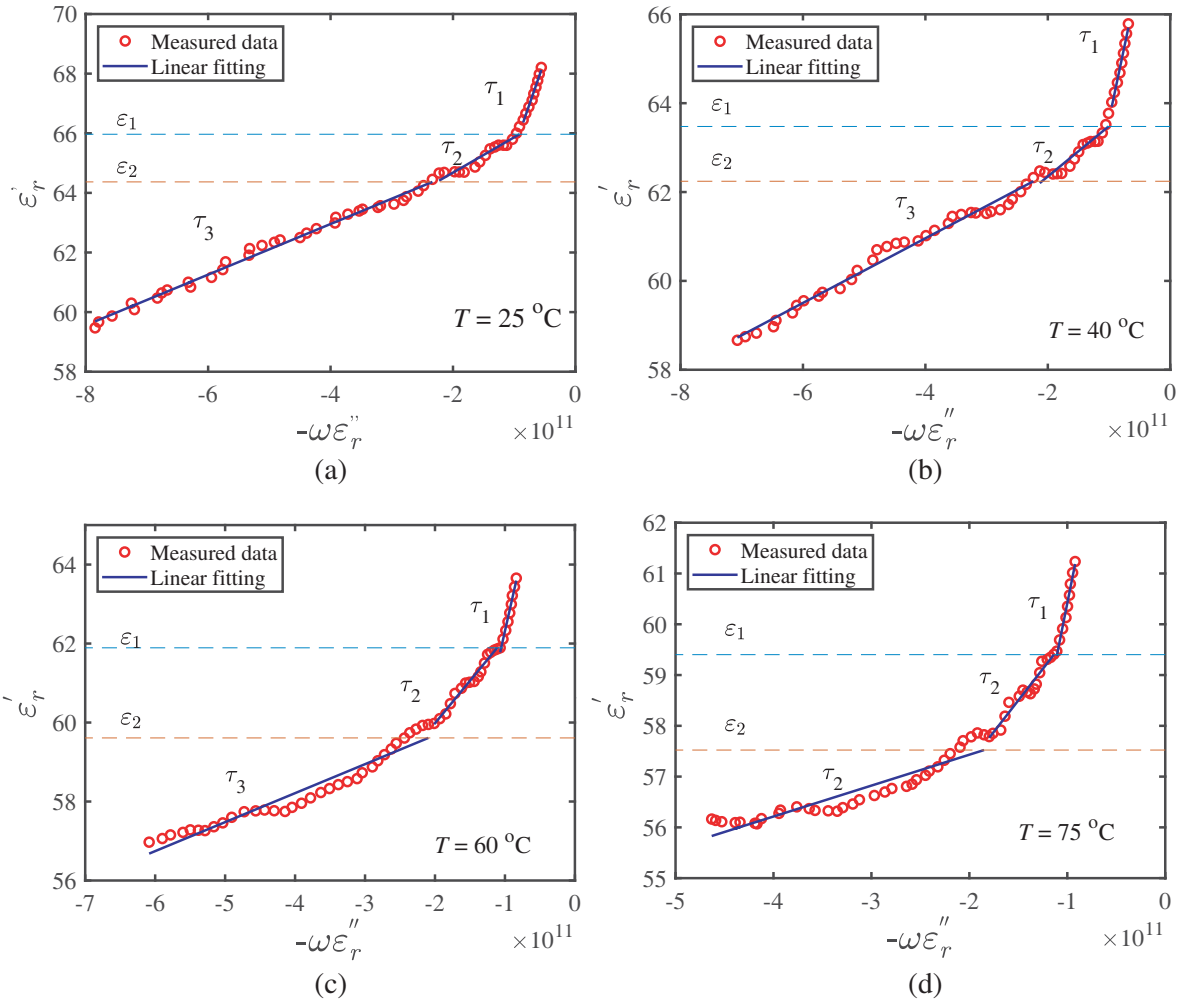

Figure 4. Linear fitting for the three relaxation times at selected pasteurization temperatures; 25°C, 40°C, 60°C, and 75°C, respectively.

3.1. Extraction of Model's Parameters

The relaxation time, τ, and static permittivity, ε_s, for each pasteurization temperature are predicted based on Equation (4) for each relaxation processes existed [17].

$$\varepsilon_r' = -\left(\omega\varepsilon_r''\right)\tau + \varepsilon_s \tag{4}$$

The linear slop of Equation (4) represents the relaxation time of the molecules objected to electromagnetic waves. The values of the static permittivity, ε_s, have been extracted when $-(\omega\varepsilon_r'') = 0$. Figure 4 exhibits the linear slopes of ε_s versus $-(\omega\varepsilon_r'') = 0$ for selected pasteurization temperature, T. The three dispersions of cow's milk pasteurization are distributed over the frequency scale respectively as low, medium, and high frequency dispersions as shown in Figure 4. The values of dielectric constants (ε_s, ε_1, and ε_2) and relaxation time (τ_1, τ_2, and τ_3) are extracted from measured data which are listed in Table 1. The corresponding regression equations for the three relaxation times as functions of temperature, T, are expressed in Equations (5), (6), and (7).

$$\tau_1 = 8.427 \times 10^{-15}(T)^2 - 1.344 \times 10^{-13}(T) + 5.768 \times 10^{-11},\ R^2 = 0.9721 \tag{5}$$

$$\tau_2 = \left\{ \begin{array}{c} 4.6844 \times 10^{-18}(T)^4 - 1.3731 \times 10^{-15}(T)^3 + \\ +1.3584 \times 10^{-13}(T)^2 - 5.1063 \times 10^{-12}(T) + 7.4601 \times 10^{-11} \end{array} \right\},\ R^2 = 0.9681 \tag{6}$$

$$\tau_3 = \left\{ \begin{array}{c} -1.7853 \times 10^{-20}(T)^6 + 4.9288 \times 10^{-18}(T)^5 - 5.4538 \times 10^{-16}(T)^4 + \\ +3.0739 \times 10^{-14}(T)^3 - 9.2194 \times 10^{-13}(T)^2 - 1.3749 \times 10^{-11}(T) - 7.0072 \end{array} \right\},\ R^2 = 0.9185 \tag{7}$$

Table 1. The approximated values of model's parameters.

T (°C)	τ_1 (ps)	τ_2 (ps)	τ_3 (ps)	ε_s	ε_1	ε_2
25	0.594	0.118	8.49	71.43	67	66.35
30	0.601	0.109	7.51	70.81	66	65.39
35	0.646	0.112	7.59	70.49	65.4	64.76
40	0.655	0.113	7.27	70.18	64.6	63.87
45	0.685	0.12	7.7	70.09	64.2	63.38
50	0.747	0.172	7.84	70.51	64.5	62.55
55	0.767	0.208	7.1	70.36	64.7	61.66
60	0.788	0.215	7.34	70.2	64.3	61.15
65	0.797	0.217	7.18	69.82	63.8	60.55
70	0.908	0.247	7.4	70.07	63.1	59.66
75	0.966	0.248	6.09	70.08	62.2	58.65

The optical permittivity ε_∞ is considered as temperature and frequency independent quantity. Hence, the value of optical permittivity, $\varepsilon_\infty^{\text{Milk}}$, is calculated based on percentage of moisture content of cow's raw milk then correlating milk infinite permittivity according to the optical permittivity of the water as given by Equation (8)

$$\varepsilon_\infty^{\text{Milk}} = \upsilon_{\text{water}} \varepsilon_\infty^{\text{water}} + (1 - \upsilon_{\text{water}})\, \varepsilon_\infty^{\text{Dried-Milk}} \tag{8}$$

where $\upsilon_{\text{water}} = 0.87(87\%)$ is the water content of the milk [18]; $\varepsilon_\infty^{\text{water}} = 4.5$ is the optical permittivity of the water [19]; and $\varepsilon_\infty^{\text{Dried-Milk}}$ equals 2.6 [20]. Consequently, the milk optical permittivity is equal to 4.25. Parameters of the developed model (β_1, β_2 and β_3) have been determined according to the fitting with measured data, and they are shown in Table 2. The parameters (β_1, β_2, and β_3) in Table 2 can be calculated at any temperature using correlated linear Equations (9), (10), and (11), respectively.

$$\beta_1 = 1.663 \times 10^{-4} T^3 - 3.15 \times 10^{-2} T^2 + 2.143T - 33.693, \; R^2 = 0.9870 \tag{9}$$

$$\beta_2 = 7.459 \times 10^{-5} T^3 - 1.732 \times 10^{-2} T^2 + 1.4584T - 25.73, \; R^2 = 0.9850 \tag{10}$$

$$\beta_3 = \left\{ \begin{array}{c} -1.3072 \cdot 10^{-9} T^6 + 3.942 \cdot 10^{-7} T^5 - 4.8253 \times 10^{-5} T^4 \\ +3.0592 \cdot 10^{-3} T^3 - 1.0555 \cdot 10^{-1} T^2 + 1.8713T - 12.293, \end{array} \right\}, \; R^2 = 0.9185 \tag{11}$$

The performance of the predicted model is shown in Figure 5.

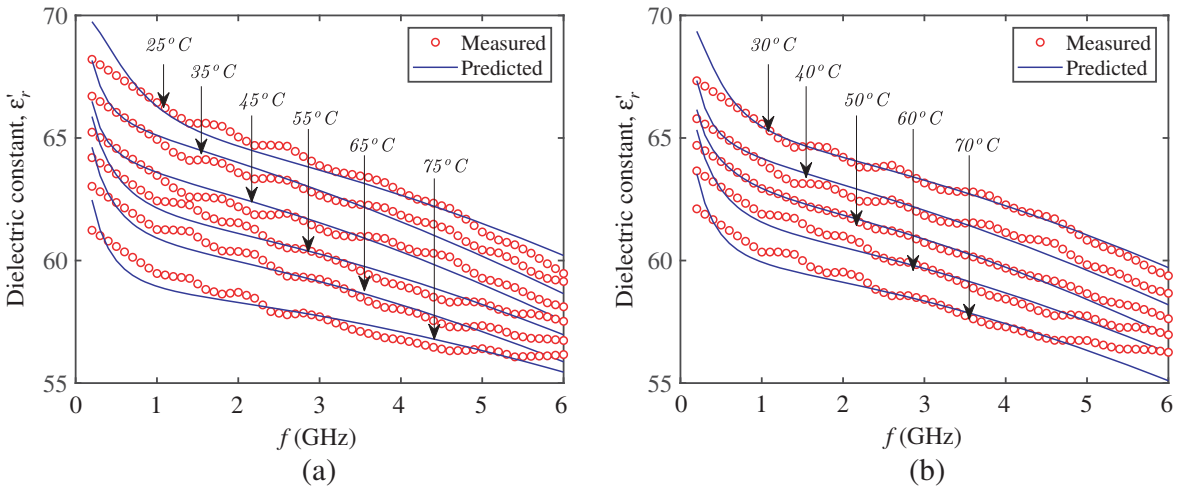

Figure 5. Evaluation and comparison of the proposed model and the measured data.

Table 2. The approximated values of model's parameters.

T (°C)	25	30	35	40	45	50	55	60	65	70	75
β_1	3.5	5	11	12	14	16	16.5	17	18	19	20
β_2	2	3	7	10	12	14	14.5	15	16	17	18
β_3	1	0.99	0.95	0.95	0.96	0.97	0.97	0.97	0.97	0.99	0.98

3.2. Conductivity Modelling

It is important to model the ionic conductivity, σ, of such dielectric properties, as long as the ionic conductivity, σ, affects the loss factor, ε_r'', of the milk during thermal treatment. Therefore, the optimization of ionic conductivity, σ, was done for accurately modelling the loss factor, ε_r'', especially at frequencies lower than 1.9 GHz. The conductivity, σ, in Table 3 has been modelled as presented in Equation (12) and Figure 6.

$$\sigma = 0.0058(T) + 0.32636, \quad R^2 = 0.9741 \tag{12}$$

Table 3. The approximated values of model's parameters.

T (°C)	25	30	35	40	45	50	55	60	65	70	75
σ (S/m)	0.46	0.49	0.51	0.58	0.6	0.63	0.67	0.67	0.7	0.72	0.75

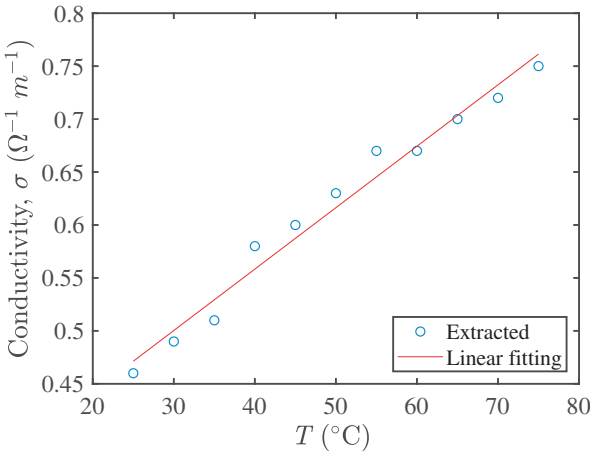

Figure 6. Variations in conductivity along side with variations the temperature and corresponding model.

3.3. Penetration Depth Modelling

The penetration depth of such microwave heating systems inside the milk is an effective key of developing new pasteurization solutions. It is mainly dependent on dielectric properties of the materials as shown in Equation (13). Hence, the penetration depth, D, determines the maximum thickness of food specimen which industrially affects the production rate. Table 4 shows the calculated penetration depth at the three operating frequencies during batch pasteurization.

$$D = \frac{\lambda_o \sqrt{\varepsilon_r'}}{2\pi\varepsilon_r''} \tag{13}$$

Table 4. The calculated values of microwave penetration depth for different frequencies.

T (°C)	ρ (g/cm^3)	ε_r'			ε_r''			D (m)		
		915 (MHz)	2.45 (GHz)	5.8 (GHz)	915 (MHz)	2.45 (GHz)	5.8 (GHz)	915 (MHz)	2.45 (GHz)	5.8 (GHz)
25	1.0248	66.63	64.69	59.86	14.20	13.36	20.76	0.0300	0.0117	0.0031
30	1.0220	65.76	63.82	59.62	14.78	13.42	20.66	0.0286	0.0116	0.0031
35	1.0188	65.13	63.34	59.27	14.49	12.72	19.30	0.0290	0.0122	0.0033
40	1.0151	64.21	62.42	58.82	15.98	12.86	18.54	0.0261	0.0120	0.0034
45	1.0108	63.66	61.87	58.27	16.43	12.70	17.67	0.0253	0.0121	0.0036
50	1.0058	63.12	61.26	57.88	17.28	12.36	16.64	0.0240	0.0123	0.0038
55	1.0002	62.62	60.69	57.70	17.63	12.31	16.25	0.0234	0.0123	0.0038
60	0.9937	62.08	60.15	57.15	17.99	12.26	15.86	0.0228	0.0123	0.0039
65	0.9863	61.45	59.67	56.80	18.34	12.18	15.15	0.0223	0.0123	0.0041
70	0.9780	60.53	58.75	56.35	18.40	11.32	13.29	0.0220	0.0132	0.0046
75	0.9687	59.66	57.88	56.11	18.85	11.16	12.42	0.0214	0.0133	0.0050

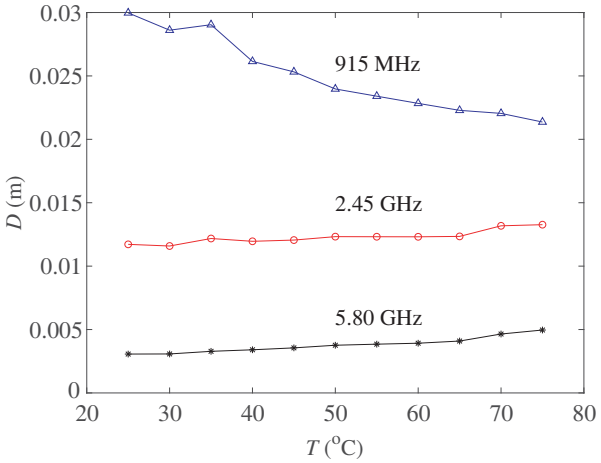

Figure 7. The penetration depth, D at 915 MHz, 2.45 GHz, and 5.8 MHz.

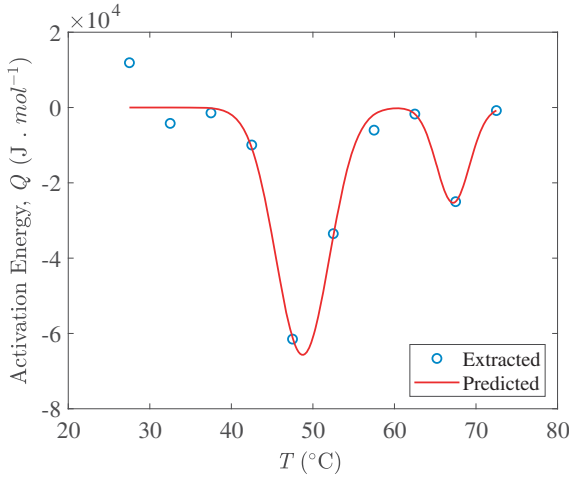

Figure 8. Variations in conductivity along side with variations the temperature and corresponding model.

Figure 7 shows the penetration depth D versus temperature T at 915 MHz, 2.45 GHz, and 5.8 GHz, respectively. Overall, D at 915 MHz shows a high value compared to 2.45 GHz and 5.8 GHz. However, the value of D at 915 MHz decreases with T because the ion conductivity (ion conductivity dominated) of milk decreases with T at low frequency (< 2 GHz). At low frequencies, loss factor ε_r'' has been dominated by ion conductivity σ, and the effect of the conductivity will be lost when the operating frequency exceeds 2 GHz. Thus, at 2.45 GHz and 5.8 GHz, the value of D increases with T caused by ε_r'' (Polar dispersion dominated) increase with T. The corresponding polynomial models for the penetration depths D at three operating frequencies are shown in Equations (14), (15), and (16).

$$D_{915} = 2.405594406 \times 10^{-6}T^2 - 4.187412587 \times 10^{-4}T + 3.931258741 \times 10^{-2};\ R^2 = 0.973 \quad (14)$$

$$D_{2.45} = \left\{ \begin{array}{c} -1.487179505\times 10^{-10}\,T^5 + 3.778554824\times 10^{-8}T^4 - 3.6802448\times 10^{-6}T^3 \\ +1.712803051\times 10^{-4}T^2 - 3.779398657\times 10^{-3}T + 4.331468605\times 10^{-2} \end{array} \right\};\ R^2 = 0.91 \quad (15)$$

$$D_{5.8} = 5.547785548 \times 10^{-7}T^2 - 2.075058275 \times 10^{-5}x + 3.302797203 \times 10^{-3};\ R^2 = 0.964 \quad (16)$$

3.4. Macroscopic Modelling

Studying the effective pasteurization temperature T using high frequency heating requires efficient modelling of process activation energy. Hence, the model of activation energy Q is a tool for validating the time temperature integrator of future pasteurization [21]. Hence, the activation energy Q has been modelled based on applying a set of linear Arrhenius equations on the adjacent and consecutive temperature, T degrees. It has been effectively modelled using two Gaussian distributions as shown in Figure 8 and expressed in Equation (17), while the model extracted parameters are listed in Table 5.

$$Q = k_1 \cdot \exp\left\{-\left(\frac{T-\mu_1}{c_1}\right)^2\right\} + k_2 \cdot \exp\left\{-\left(\frac{T-\mu_2}{c_2}\right)^2\right\} \tag{17}$$

The parameters of the modelled activation energy are shown in Table 5. All determined bound ranges give $R^2 = 0.9541$ as a regression confidence.

Table 5. The approximated values of model's parameters.

Parameters	Mean	Lower Bound	Upper Bound
k_1 (Mol/J)	-6.57×10^4	-8.78×10^4	-4.36×10^4
k_2 (Mol/J)	-2.50×10^4	-5.22×10^4	1631
μ_1(°C)	48.75	47.43	50.07
μ_2(°C)	67.19	57.53	76.84
c_1	4.643	2.781	6.505
c_2	2.86	−3.121	8.84

4. CONCLUSION

The measurements of dielectric properties of cow's raw milk from 1.9 GHz to 6 GHz have been reported in this paper. The wide range of temperature measurements provides a clear dispersion that exists in milk through pasteurization process which could not be monitored by the literature mentioned. The triple-dispersion has been effectively modelled using modified combination of Cole-Davison and Debye equations. The developed model gives good estimation. The dielectric constants, electrical conductivity based on loss factor, relaxation time, and activation energy are effectively modelled with corresponding verifications by the extracted parameters. However, the dielectric modelling of milk through certain pasteurization technique is an important tool which demonstrates the relationship between pasteurization parameters and electromagnetic properties of milk which is an important factor in future of the microwave-based pasteurization development.

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