

## Microwave Dielectric Properties of Hevea Rubber Latex in the Temperature Range of -30°C to 50°C

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### ABSTRAK

Sifat dielektrik lateks hevea diukur pada frekuensi mikrogelombang tertentu meliputi julat suhu -30°C hingga 50°C. Pengukuran dibuat pada lateks pekat, lateks segar dan lateks segar yang telah dicairkan dengan menggunakan penduga talian sepaksi terbuka di hujung dan Automated Network Analyzer. Ada terdapat penambahan mendadak di dalam nilai pemalar dielektrik sebanyak sekali ganda dan faktor kehilangan dielektrik sebanyak dua kali ganda apabila fasa lateks berubah daripada pepejal menjadi cecair. Kesan suhu pada kekonduksian ion pada 0.2 GHz dan orientasi dwikutub pada 2.6 GHz dan ke atas dapat dilihat dengan jelas dalam kajian ini. Didapati bahawa frekuensi di sekitar 10 GHz adalah frekuensi operasi yang sesuai untuk latexometer mikrogelombang di mana sifat dielektrik tidak terlalu bersandar pada suhu.

### ABSTRACT

The dielectric properties of hevea rubber latex were measured at selected microwave frequencies over the temperature range of -30°C to 50°C. The measurements were made on latex concentrate, fresh latex and diluted fresh latex by using an open-ended coaxial-line probe and an automated network analyser. There is a steep increase in the dielectric constant of about one order and dielectric loss factor of about two orders as the phase of latex changes from solid to liquid. The effect of temperature on the ionic conductivity at 0.2 GHz and dipole orientation at 2.6 GHz and above can be clearly seen in these studies. It was found that, the frequency around 10 GHz is a suitable operating frequency for microwave latexometer as dielectric properties are fairly independent of the temperature.

**Keywords:** hevea rubber latex, dielectric constant, dielectric loss factor, ionic conductivity, dipole orientation

### INTRODUCTION

The dielectric properties of hevea rubber latex at various moisture contents have been reported over a frequency range of 0.2 GHz-20 GHz (Khalid and Wan Yusoff 1992; Khalid *et al.* 1994). However, all measurements were done only at room temperature and data at various temperatures are desirable.

In the previous work, it was suggested that the dielectric properties of latex are mainly due to the migration of dissolved ions in the latex, especially at frequencies below 2 GHz. These ions originate from the conducting phases in latex which arise from proteins, fatty acid soaps, phospholipids and various types of ohmic species (Chen 1979; Gorton and Pendle 1985). Above 2 GHz, the losses are due to the dipole orientation of the water molecules. Both factors are considerably affected by thermal agitation, and the corresponding dielectric properties of latex are therefore temperature dependent.

The previous studies on food products (Bengtsson and Risman 1971) showed that both ionic conductivity,  $\varepsilon_i''$  and dipole orientation,  $\varepsilon_d''$  are temperature dependent with  $\varepsilon_i''$  increases and  $\varepsilon_d''$  decreases with increasing temperature.

The dielectric properties of water change drastically as its phase changes from solid to liquid (Hasted 1973). As latex consists of 40-60% water, the same behaviour might be expected. Knowledge of the dielectric properties of latex at various temperatures are required to determine the different absorption of microwave energies in latex during microwave heating. These properties are also essential in modelling the electromagnetic wave propagation and interaction in latex, especially in estimating the effect of temperature on the microwave latexometer (Khalid 1994).

This article describes the microwave dielectric properties of latex over a temperature range of -30°C to 50°C. The dielectric properties are measured from a wide range of samples of fresh latex and latex concentrate diluted from 40 to 98% (wet basis) and deionized water for comparison. The detailed studies will emphasize the effects of temperature on the ionic conductivity and dipole orientation in various samples of latex.

## MATERIALS AND METHODS

The samples used included freshly tapped latex, latex concentrate (LA-TZ), diluted fresh latex, diluted latex concentrate and deionized water. Diluted fresh latex has a moisture content (MC) of 55-98% (wet basis), while latex concentrate has 40-55%. In this study, latex concentrate which had been preserved with 0.025% tetramethyl-thiuram disulphide (TMTD)/zinc oxide (ZnO) and 0.2-0.3% ammonia (John *et al.* 1982) was supplied by the Rubber Research Institute of Malaysia.

Dielectric measurements were made using a 4-mm open-ended coaxial line probe with an automated network analyser (HP8720B) and a computer. This technique is based on the determination of the phase of the reflection coefficient, and is nondestructive. The electric field strength is constant throughout this study. For calibration purposes, three dielectric references with well-known dielectric constants were used: air, a metal short and deionized water.

Three separate experiments were performed to obtain a complete data of the dielectric properties of latex for the temperature range of -30-50°C. A wide range of samples within 2-3% difference in moisture content were tested with

a total of about 40 samples. Two waterbaths were used to warm the calibration liquid (deionized water) and latex. Approximately 40 ml of latex and water were poured into their respective waterbaths. Both the latex and water were maintained at  $\pm 1^{\circ}\text{C}$ . A digital thermometer was used to monitor the temperature. The water and latex were stirred for temperature equilibration. Using standard calibration procedures (Krazewski *et al.* 1983) which involve the open circuit (air) and the short circuit (metal short), the probe is then immersed in the deionized water to attain the required temperature. When the temperature reached  $25 \pm 1^{\circ}\text{C}$ , the probe was calibrated, then dried before immersing it in latex at about the same temperature ( $\pm 1^{\circ}\text{C}$ ) before the first measurement was made. The water and latex were again warmed slowly to  $30^{\circ}\text{C}$  and this same procedure is repeated for 35, 40, 45 and  $50 \pm 1^{\circ}\text{C}$ . Care was taken to ensure that there were no air bubbles between the sensor and the water or latex. For best measurement results, it is important to enter the correct calibration constant for water of the required temperature (Hasted 1973) and maintain the same temperature during calibration and measurement (Hewlett Packard 1990). This is especially important for samples containing a lot of moisture or liquid samples.

The second part made use of the same set-up, but two icebaths were used instead of waterbaths. The above procedures were repeated as dielectric measurements were made at 20, 15, 10, 5 and  $2 \pm 2^{\circ}\text{C}$ .

The third set-up used dry ice for an ice bath. The sensor was first calibrated at  $0 \pm 1^{\circ}\text{C}$  in ice water before it was frozen inside the latex. A copper-constantan thermocouple was frozen inside the latex to monitor the temperature with the help of a digital multimeter (HP3478A) to register the voltage. Dielectric measurements were then made from -2.5 to  $-30 \pm 2.5^{\circ}\text{C}$ . The actual moisture content was determined experimentally by drying 1.5-2.5 grams of the samples in an oven at  $70^{\circ}\text{C}$  until the latex became transparent. The difference in the weight of the sample before and after drying was then divided and multiplied by 100 to give the percentage moisture content. Three sets of the same sample were dried each time and the average results recorded.

## RESULTS AND DISCUSSION

The temperature dependence of the dielectric properties of hevea latex at 0.2, 2.6, 10.0 and 20.0 GHz are summarized in Fig. 1-4 respectively with data tabulated in Tables 1(a), (b). Curves shown are the dielectric constant  $\epsilon'$  and dielectric loss factor  $\epsilon''$  for latex concentrate (MC=38%), fresh latex (MC=56%), diluted fresh latex (MC=89%) and deionized water (MC=100%).

Generally, the variation of  $\epsilon'$  and  $\epsilon''$  of hevea latex over the temperature range of -30 to  $50^{\circ}\text{C}$  can be divided into 3 regions as follows

(i)  $-30^{\circ}\text{C}$  to  $3^{\circ}\text{C}$  (solid state region)

In this range of temperature (0.2 GHz)  $\epsilon'$  is almost constant with temperature for fresh latex, diluted fresh latex and water with its value around  $4.1 \pm 0.5$  while at 2.6 GHz  $\approx 3.3$ , 3.6 and  $4.0 \pm 0.5$  respectively. However, at 0.2 GHz for latex

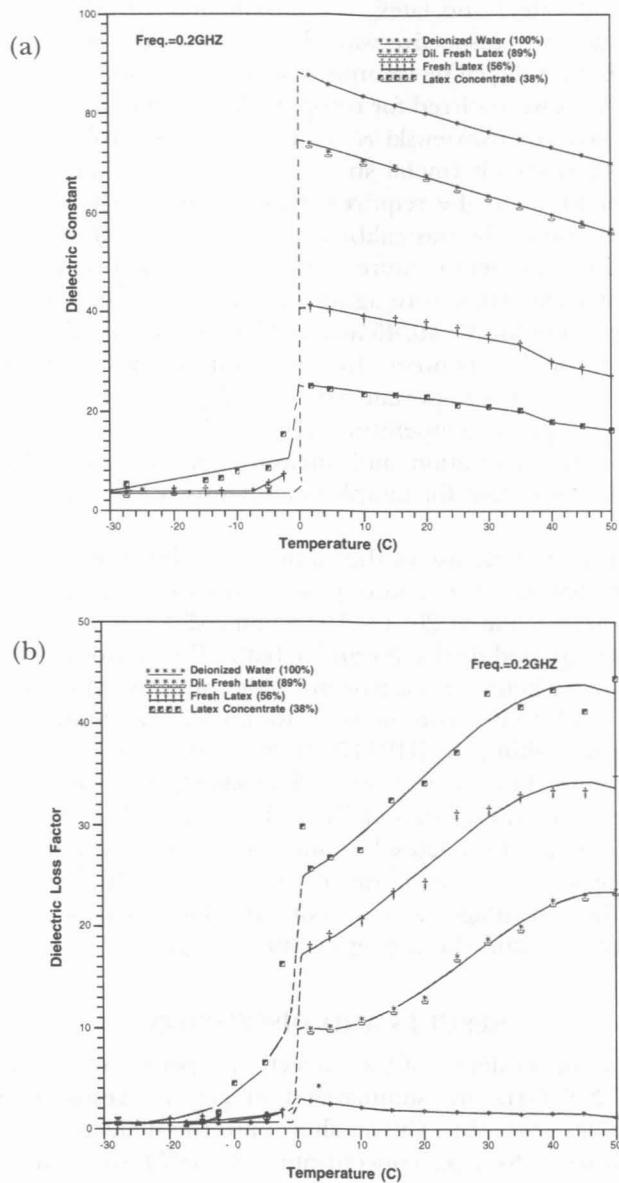


Fig 1. Variation of the dielectric properties for various solutions of Hevea latex with respect to temperature at 0.2 GHz.  
(a) Dielectric constant (b) Dielectric loss factor

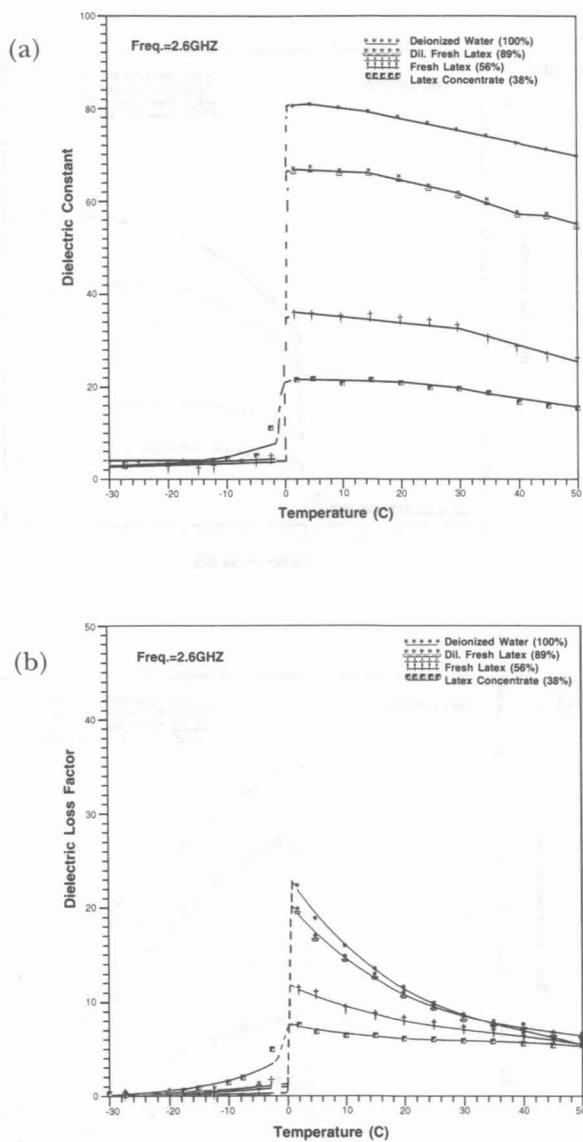


Fig 2. Variation of the dielectric properties for various solutions of Hevea latex with respect to temperature at 2.6 GHz.  
 (a) Dielectric constant (b) Dielectric loss factor

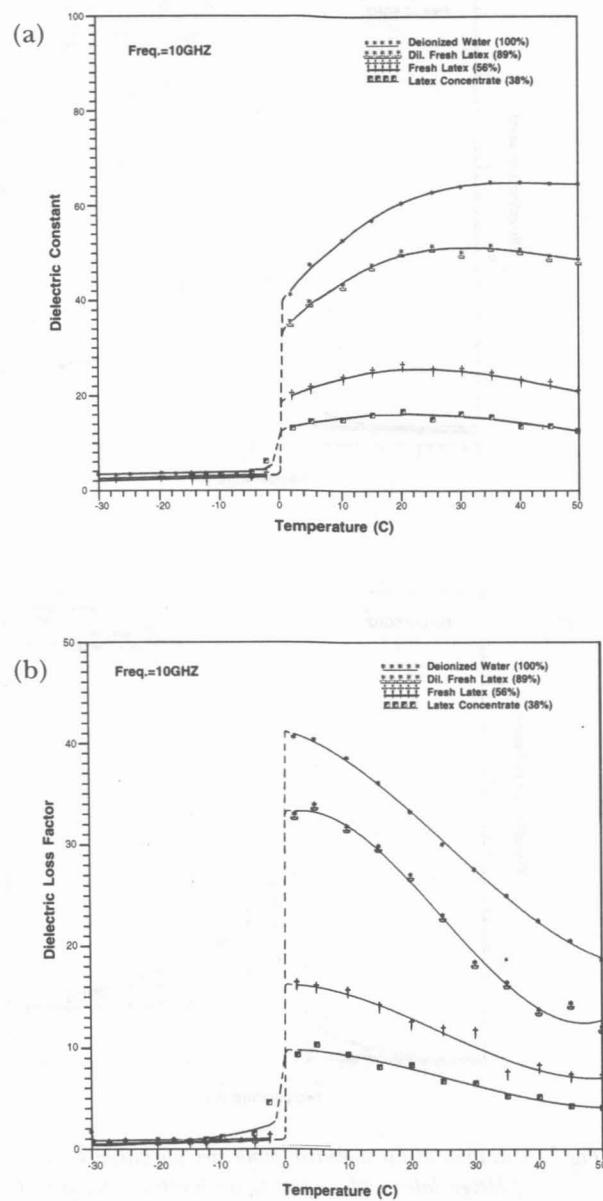


Fig 3. Variation of the dielectric properties for various solutions of Hevea latex with respect to temperature at 10 GHz.  
(a) Dielectric constant (b) Dielectric loss factor

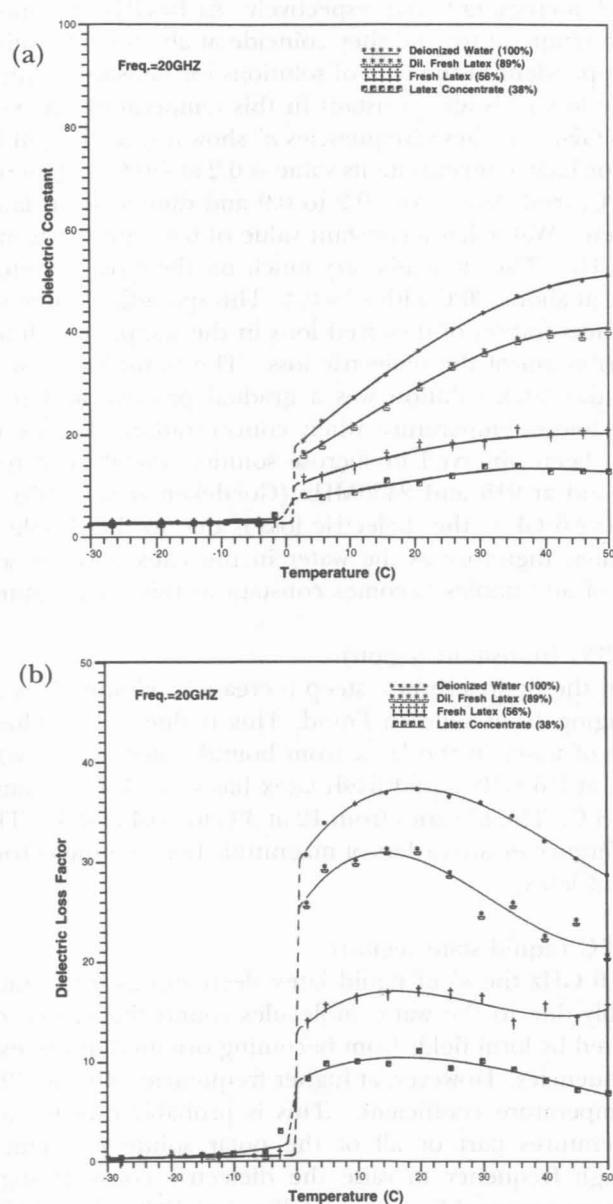


Fig 4. Variation of the dielectric properties for various solutions of Hevea latex with respect to temperature at 20 GHz. (a) Dielectric constant (b) Dielectric loss factor

concentrate its value is 4.1 at -30°C increasing to 8.3 at -10°C while at 2.6 GHz its value is 2.4, increasing to 5.2 respectively. At 10 GHz and above,  $\epsilon'$  is almost constant with temperature and they coincide at about -20°C with  $\epsilon' \approx 3 \pm 0.5$ , and is almost independent of the type of solutions for moisture content above 50%.

Dielectric loss  $\epsilon''$  is also constant in this temperature range, except at 0.2 GHz and 2.6 GHz. At these frequencies  $\epsilon''$  show a spreading in its value where at 0.2 GHz, for latex concentrate its value is 0.2 at -30°C and increases gradually to 3.0 at -10°C, fresh latex from 0.2 to 0.9 and diluted fresh latex from 0.1 to 0.6 respectively. Water has a constant value of 0.4. Similar trends can also be seen at 2.6 GHz. This depends very much on the type of solution used, and they coincide at about -20°C with  $\epsilon'' \approx 0.4$ . This spreading is due to the different degree of concentration of dissolved ions in the sample which are responsible for the contribution of the dielectric loss. The transition of water from solid to liquid in the latex solution was a gradual process, and it occurred at a progressively lower temperature when concentration of latex was increased. This has also been observed in sucrose solution (0–60%) at temperatures of 25 to 20°C and at 915 and 2450MHz (Goedeken *et al.* 1995). Since at high frequencies (>2.6 GHz) the dielectric loss is due to the dipole orientation of water molecules, therefore as the water in the latex become solid at around -10°C the  $\epsilon''$  of all samples becomes constant at this temperature.

#### (ii) -3°C to 3°C (transition region)

In this region the figures show a steep increase in  $\epsilon'$  and  $\epsilon''$  as a phase of the latex is changing from solid to liquid. This is due to the changing in the physical state of water in the latex from bound water to free water.

For example, at 2.6 GHz liquid fresh latex has  $\epsilon' = 35$  at 3°C and frozen latex has  $\epsilon' = 3$  at -3°C. The  $\epsilon''$  varies from 12 at 3°C to  $\approx 0.4$  at -3°C. This means that there is a difference of two orders of magnitude between dielectric loss in liquid latex and solid latex.

#### (iii) above 3°C (liquid state region)

At 0.2 and 2.6 GHz the  $\epsilon'$  of liquid latex decreases as temperature increases. This is possibly due to the water molecules round the ion or organic group being prevented by local fields from becoming oriented in the external field at these low frequencies. However, at higher frequencies, 10 and 20 GHz  $\epsilon'$  shows a positive temperature coefficient. This is probably due to the fact that at higher temperatures part or all of the polar solute molecule relaxes at a sufficiently high frequency to raise the dielectric constant slightly. Similar results have been reported for water (Collie *et al.* 1948; Cook 1952; Hasted and El Sabeh 1953). Since latex follows the behaviour of water the same interpretation of the results can be applied.

As previously reported, at 0.2 GHz the dielectric loss factor is mainly ionic due to the conducting phases in the latex. Elevations in temperature raise the mobility of ions in the solution, resulting in  $\epsilon''_i$  increases, but a decrease in the concentration of latex decreases the availability of the ionic species.

TABLE 1(a)

Fitted dielectric constant and dielectric loss factor data from -30 to 50°C at 0.2 and 2.6 GHz

Temperature (°C)	Moisture Content, % (wet basis)															
	0.2 GHz								2.6 GHz							
	38		56		89		100		38		56		89		100	
	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$
-30	4.1	0.2	3.4	0.2	3.3	0.1	4.1	0.4	2.4	0.2	3.0	0.1	3.2	0.1	4.0	0.2
-20	5.8	0.8	4.1	0.5	3.9	0.2	4.1	0.4	3.5	0.6	3.3	0.3	3.6	0.4	4.0	0.2
-10	8.3	3.0	4.9	0.9	4.7	0.6	4.2	0.4	5.2	1.6	3.6	0.5	3.9	0.7	4.1	0.2
2	24.9	24.9	40.5	16.8	73.3	9.1	87.5	1.9	21.7	7.3	35.3	11.2	66.8	19.3	80.8	21.8
10	24.1	28.3	39.5	20.0	70.7	9.7	83.9	1.3	21.6	6.5	35.6	9.4	66.9	14.6	80.2	16.0
20	22.7	34.1	37.5	25.1	67.0	13.0	79.9	0.9	20.7	5.9	34.5	7.8	64.8	10.6	78.2	11.2
30	20.8	39.4	34.6	29.9	63.1	17.5	76.4	0.8	19.1	5.6	32.0	6.8	61.5	8.3	78.4	8.4
40	18.6	42.7	31.0	33.0	59.4	21.3	73.1	0.7	17.1	5.4	28.5	6.0	57.9	7.0	72.3	6.6
50	16.0	42.1	26.5	32.9	56.4	22.5	69.8	0.4	14.6	4.9	24.4	5.2	55.2	6.1	69.4	5.1

TABLE 1(b)

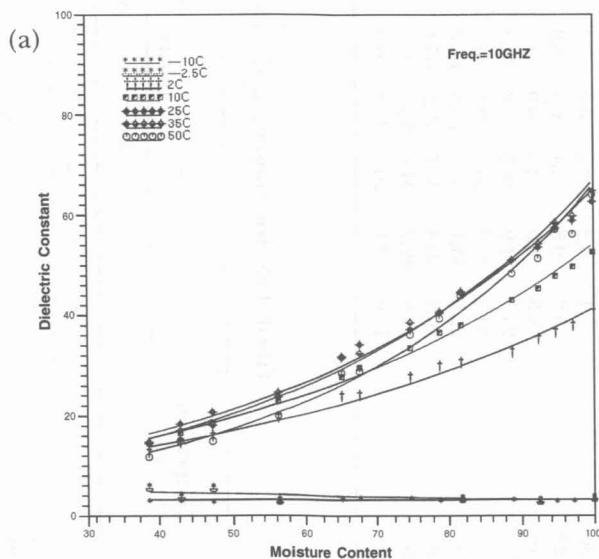
Fitted dielectric constant and dielectric loss factor data from -30 to 50°C at 10 and 20 GHz

Temperature (°C)	Moisture Content, % (wet basis)															
	10 GHz								20 GHz							
	38		56		89		100		38		56		89		100	
	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$	$\epsilon'$	$\epsilon''$
-30	2.3	0.2	2.6	0.3	2.8	0.3	3.3	0.7	2.5	0.1	2.7	0.2	2.8	0.2	3.0	0.3
-20	2.9	0.4	2.7	0.3	2.9	0.5	3.3	0.7	2.8	0.3	2.7	0.2	2.9	0.3	3.0	0.3
-10	3.6	0.8	2.9	0.5	3.1	0.6	3.4	0.7	3.2	0.7	2.8	0.3	3.0	0.4	3.1	0.3
2	13.1	9.5	19.6	15.9	35.1	33.0	41.6	40.7	7.9	8.4	10.9	13.7	16.0	26.2	19.1	31.0
10	15.0	9.0	23.3	15.0	43.5	31.6	52.3	38.0	9.4	9.8	14.1	16.3	22.1	30.2	27.3	35.6
20	15.8	7.6	25.2	12.6	49.1	26.0	60.2	33.0	11.4	10.2	17.1	16.8	29.6	30.0	36.5	37.1
30	15.2	5.9	24.7	9.7	50.7	24.7	63.6	27.3	13.1	9.4	19.2	15.6	35.9	26.7	43.8	35.5
40	13.8	4.4	22.8	7.4	49.8	13.4	64.3	22.0	13.7	8.0	20.3	14.5	44.0	22.9	49.0	32.1
50	12.0	3.7	20.3	6.6	48.0	12.3	63.8	18.5	12.4	6.4	20.4	15.1	49.3	21.4	51.6	28.4

This can be seen in the dielectric loss for water where it does not only have a negative temperature dependence but the loss factor has a value  $<1$  at 25°C. Water does not have any accountable ionic species, but only polar molecules (dipoles). The gradient of the curve is dependent on the degree of concentration of dissolved ions in the sample, i.e. the less moisture a sample has, or the more concentrated a sample is, the more ionic species will be found in the samples. At 2.6 GHz and above  $\epsilon''$  decreases as temperature increases, which is similar to the trend of the deionized water. In this region the losses are mainly governed by the orientation of the dipoles,  $\epsilon_d''$ . The shape of the latex curve shows a depression compared with deionized water, which is due to water binding by dissolved ions. The above phenomenon is almost similar to the temperature dependence for a few food products at 2.8 GHz and from -20 to 60°C reported earlier (Bengtsson and Risman, 1971).

At 10 GHz,  $\epsilon'$  is fairly independent of the temperature in the range of 25-35°C, which makes this frequency suitable for water content measurement of hevea latex. This frequency has been utilized in microwave latexometer described earlier.

The variation of  $\epsilon'$  and  $\epsilon''$  with moisture content at various temperatures and at 10 GHz are shown in Fig. 5. It is clearly shown that in the temperature range of 25-35°C (normal measurement temperatures) the variations in  $\epsilon'$  and  $\epsilon''$  are 0.5-1.0 and 2-4 respectively. However, this does not have much effect on the accuracy of the latexometer since it uses the reflection method (Khalid 1994).



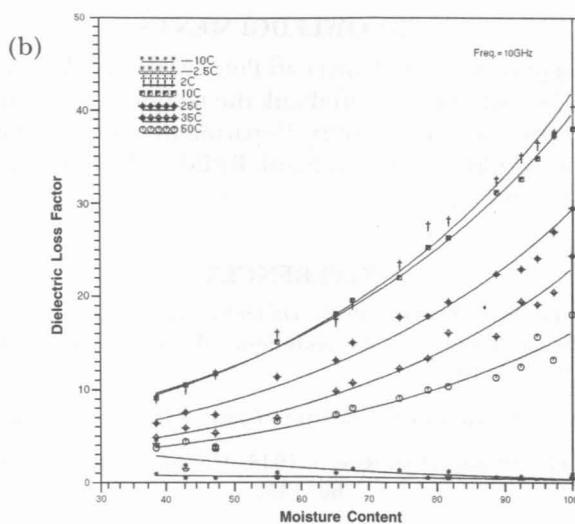


Fig 5. Variation of the dielectric properties at various temperatures with respect to moisture content at 10 GHz. (a) Dielectric constant (b) Dielectric loss factor

### CONCLUSION

The microwave dielectric properties of hevea latex at temperatures of -30-50°C can be divided into three regions, i.e. solid state region, transition region and liquid state region. There is a steep increase of  $\epsilon'$  of about one order and  $\epsilon''$  of about two orders as a phase of latex changes from solid to liquid.

In these studies, the effects of temperature on the ionic conductivity at 0.2 GHz and dipole orientation at 2.6 GHz and above are clearly seen.

In the solid state region at 0.2 and 2.6 GHz,  $\epsilon'$  and  $\epsilon''$  show a spreading according to the strength of dissolved ions in the sample and coincide at about -30°C. However, at higher frequencies (10 GHz and above),  $\epsilon'$  and  $\epsilon''$  coincide at -10°C where the water molecules become bounded and  $\epsilon'$  and  $\epsilon''$  of all samples become constant.

In the transition region, there is a steep increase in  $\epsilon'$  and  $\epsilon''$  as the physical state of water in the latex undergoes changes from bound water to free water.

In the liquid state region at 0.2 and 2.6 GHz,  $\epsilon'$  decreases with increasing temperature but at 10 and 20 GHz  $\epsilon'$  increases with increasing temperature. However, the elevation in temperatures raises the mobility of ions in liquid latex resulting in  $\epsilon''$  due to ionic conductivity ( $\epsilon_i''$ ) increases at 0.2 GHz. But at 2.6 GHz and above  $\epsilon''$  due to dipole orientation ( $\epsilon_d''$ ) decreases as temperature increases.

It was found that 10 GHz is a suitable operating frequency for microwave latexometer as  $\epsilon'$  and  $\epsilon''$  are fairly independent of the temperature.

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