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Microwave Measurement on the Degree of Binding of Water in Hevea *Rubber Latex*

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Measurements of the dielectric properties of Hevea *rubber latex at various moisture content and selected temperatures (2, 15, 25, 35 and 50°C) were done using the automated network analyser. The results were interpreted using Weiner's theory for dielectric mixtures to evaluate the degree of binding of water molecules in* Hevea *rubber latex. Changes in the mixing condition η indicated in the degree of binding or polarisation of the water molecules At all frequencies there existed a region of constant η where optimum polarisation occured before decreasing due to an increase in thermal agitation with increasing temperature. This indicated that η was not only dependent on moisture but very much affected by frequency and temperature. These results were further supported by data on the variation of the degree of binding at selected temperatures and frequencies.*

Key words: microwave; binding; water; *Hevea* latex; measurements, dielectric properties; moisture content; polarisation; Weiner's theory; mixing condition; temperature

In every study, knowing the composition of a material is essential for the interpretation of the results obtained. Hence, this is true for *Hevea* rubber latex. Being a biological product, its composition varies from tree to tree and by many other factors such as weather, soil conditions, clones, tapping system, *etc*. Since it is not a man-made product, it is truly impossible to know the exact composition of each sample that is being used. Fresh latex consists of 55%–80% water, 15%–45% rubber hydrocarbon and approximately 2%–4% non-rubber constituents[1,2]. The variation of moisture in fresh latex and latex concentrate is very broad, ranging from 40%–80%. Since latex consists of more than 50% water or moisture, water plays an important role in the dielectric measurements. The moisture content refers to the water that is found in the latex but which is not chemically combined with the latex particles. When water molecules are bound to another molecule or particle, they would no longer be free to participate in the polarisation of the medium, as in free water. Bound water can exist in different forms, with different degrees of binding, which are dependent on the amount of moisture. When bounded the molecules move slower and polarisation involves breaking of bonds. Therefore, the relaxation frequency of bound water is lower than free water. The relaxation frequency of free water is 17 GHz[3]. There are two methods

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for determining the moisture content by using the latexometer[4] or the conventional oven drying method or both. In this study the conventional oven drying method are employed

The dependence of microwave dielectric properties on the moisture content for moist materials have long been used as an indicator for moisture determination This contribution is proportional to the freedom of movement or polarisation of the water molecules and thus to the degree of binding of the water molecules Latex is treated as a biphase liquid consisting a mixture of water and dry rubber. The theory of dielectric mixtures will be used to determine the mixing condition in latex and thus the degree of binding of the water molecules at selected frequencies (0 2 GHz–20 GHz) and temperatures (2°C–50°C)

Dielectric Mixtures

In all the dielectric mixture theories proposed for a biphase mixture, it is assumed that one component forms the matrix or the host material and the other is sparsely dispersed in the form of particles For *Hevea* rubber latex, the rubber particles and non-rubber constituent are considered as a host of dispersive continuum of latex particles of complex permittivity ε^*_r These continuum of latex particles mixes with the inclusion, a high volume fraction of ellipsoidal water molecules with complex permittivity ε^*_w Since water is a polar molecule, when placed in an electric field the dipoles tend to rotate in order to align with the field The orientation of the dipoles mark the ability to be polarised and is a measure of the dielectric constant of the medium ε' and hence the degree of binding of the inclusion in the host material The dielectric loss factor ε'' is a measure of energy loss in the form of heat or dissipation of electromagnetic energy

For this analysis, the most appropriate theory that can be applied is Weiner's theory This theory has been used by Ozawa and Kuroiwa[5] in their analysis of ice, snow and water mixtures, Suresh *et al*[6] in water absorbed in soils, Bhartia[7] in water absorbed in leather and Khalid *et al*[8] in *Hevea* rubber latex at various moisture contents at room temperature In this theory uncertainty in the electric field is taken into consideration by a factor η, known as the mixing condition Weiner's equation for mixed dielectrics[5] for latex may be expressed as

$$\frac{\varepsilon^* - 1}{\varepsilon^* + \eta} = \delta_w \frac{\varepsilon^*_w - 1}{\varepsilon^*_w + \eta} + \delta_w \frac{\varepsilon^*_r - 1}{\varepsilon^*_r + \eta} \qquad 1$$

where ε^*, ε^*_w, ε^*_r are the complex permittivities of the mixture (*Hevea* rubber latex), water and solid continuum, respectively

δ_w, δ_r are the volume fractions of water and the solid continuum, respectively

η is the mixing condition

The maximum limit is achieved when $\eta = \infty$, also called Weiner's upper bound This corresponds to the case where the ellipsoidal water molecules (inclusions) with the major axis are parallel to the direction of the applied electric field, easily polarised and dielectric constant is maximum The water molecules are loosely bound The minimum limit corresponds to $\eta = 0$, known as Weiner's lower bound, where the major axis of the ellipsoidal water molecules are perpendicular to the applied field and the dielectric constant is minimum This also indicated that the water molecules are tightly bound By substituting $\eta = \infty$ and $\eta = 0$ in *Equation 1*, the maximum and minimum limits for a two-component mixture are respectively given by

$$\varepsilon^*_{max} = \delta_w \varepsilon^*_w + \delta_r \varepsilon^*_r \qquad 2$$

$$\frac{1}{\varepsilon^*_{min}} = \frac{\delta_w}{\varepsilon^*_w} + \frac{\delta}{\varepsilon^*_r} \qquad 3$$

The permittivity of the mixture must always be within the limits of Weiner's upper bound and Weiner's lower bound. By determining the permittivity of the mixture and studying the variation of η, interpretations can be made on how the water molecules are aligned. With increasing η, more of the water molecules are aligned parallel to the field. The reverse is true with decreasing η where more of the water molecules are aligned perpendicular to the field. Suresh *et al.*[6] indicate in their study that the loss tangents of the mixtures are independent of the mixing condition. In this study, only the real relative permittivity or the dielectric constant are taken into account.

MATERIALS AND METHOD

Samples used were prepared from latex concentrate and freshly tapped latex diluted with deionised water. Latex concentrate which had been preserved with 0.025% tetramethylthiuram disulphide/zinc oxide and 0.2%–0.3% ammonia[9] was supplied by the Rubber Research Institute of Malaysia (RRIM) while freshly tapped latex was collected daily from the Universiti Research Park of Universiti Putra Malaysia. Over 40 samples were used for this study ranging in moisture content from 40%–98%.

An automated network analyser (HP 8720B) coupled with a computer and open-ended coaxial line probe was used to measure the permittivity of the samples from 0.2 GHz–20 GHz and 2°C to 50°C. The probe is calibrated according to standard calibrating procedures[10,11]. For variation in temperature, two water baths were used to heat or cool the samples as reported[12]. The accuracy of the measurements was about ±5% for ε' and ±3% for ε''. The conventional oven-drying method was used to obtain the moisture content of the samples. Three 1.5 g–2.5 g of the samples were dried at 70°C and the average results recorded.

Figures 1–3 show the moisture dependence of the dielectric constant to illustrate the degree of binding at selected temperature and frequency. *Figure 4* illustrates variation of the degree of binding with respect to temperature at 2.6, 10 and 18 GHz with data (*Table 1*).

RESULTS AND DISCUSSION

As a basis for moisture determination, the dependence of the microwave dielectric properties of

TABLE 1. DATA FOR THE DEGREE OF BINDING η WITH RESPECT TO TEMPERATURE AND FREQUENCY

Frequency	2.6 GHz	10 GHz	18 GHz
Degree of binding Temperature (°C)	$\eta_{2.6}$	η_{10}	η_{18}
2	70	25	–
15	70	35	20
25	70	40	30
35	60	40	40
50	50	30	40

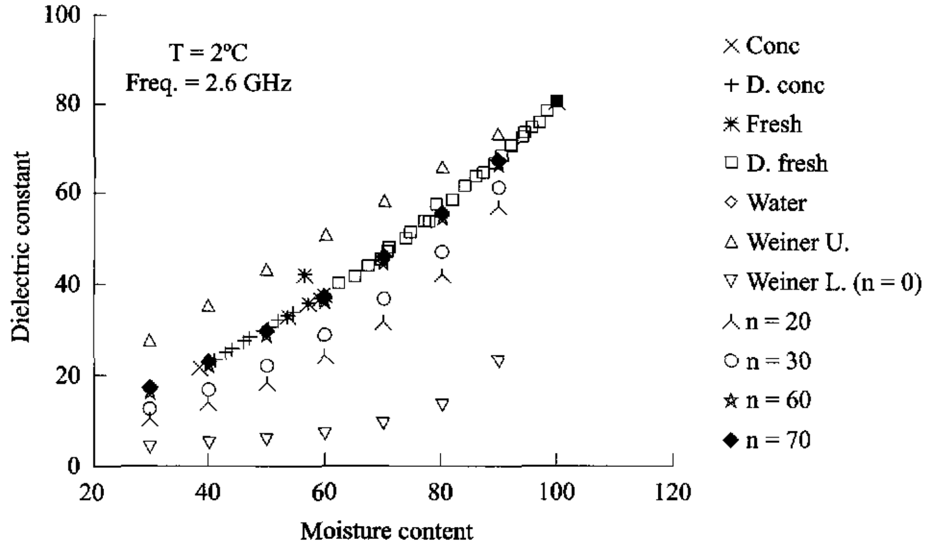

Figure 1a. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 2.6 GHz at 2°C.*

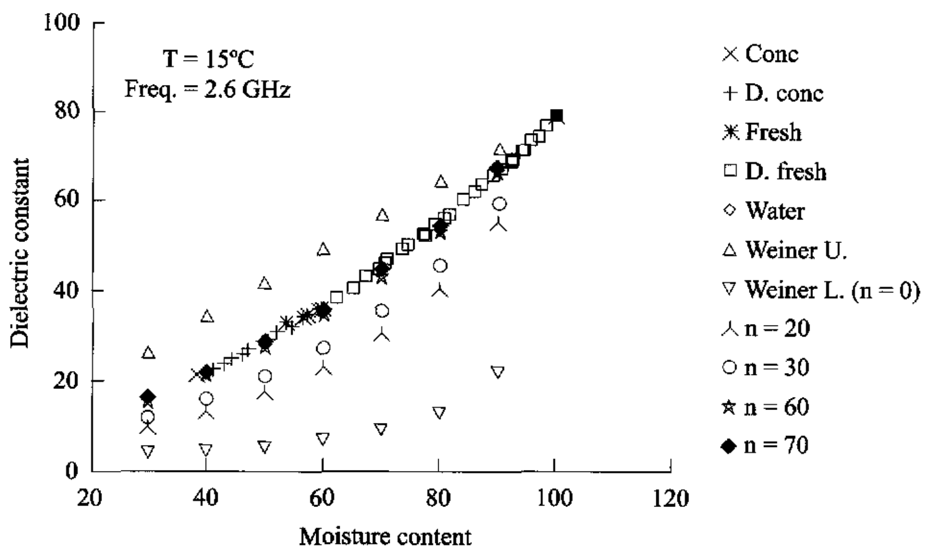

Figure 1b. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 2.6 GHz at 15°C.*

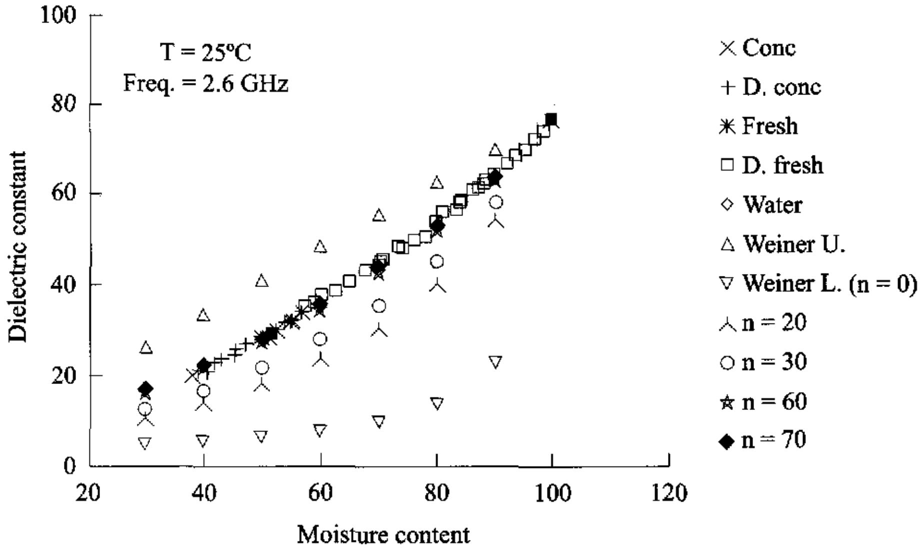

Figure 1c. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 2.6 GHz at 25°C.*

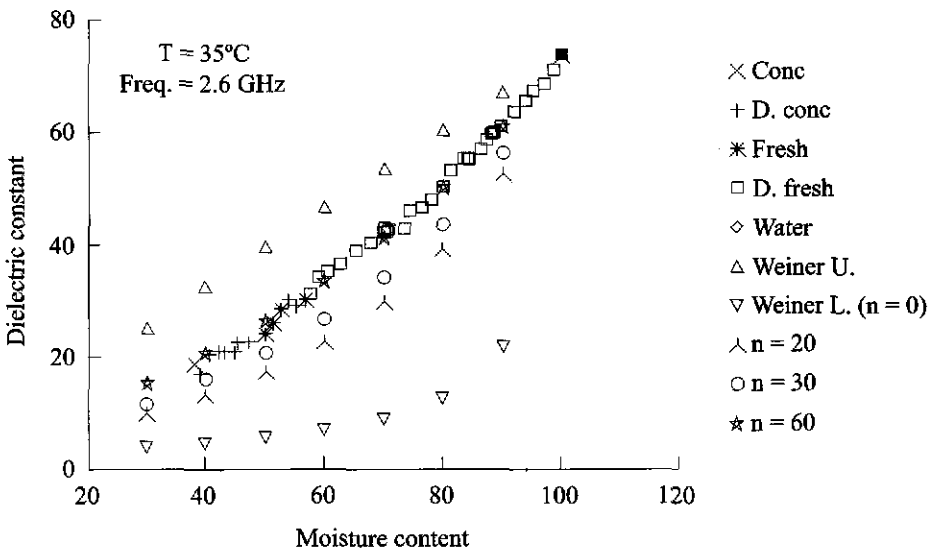

Figure 1d. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 2.6 GHz at 35°C.*

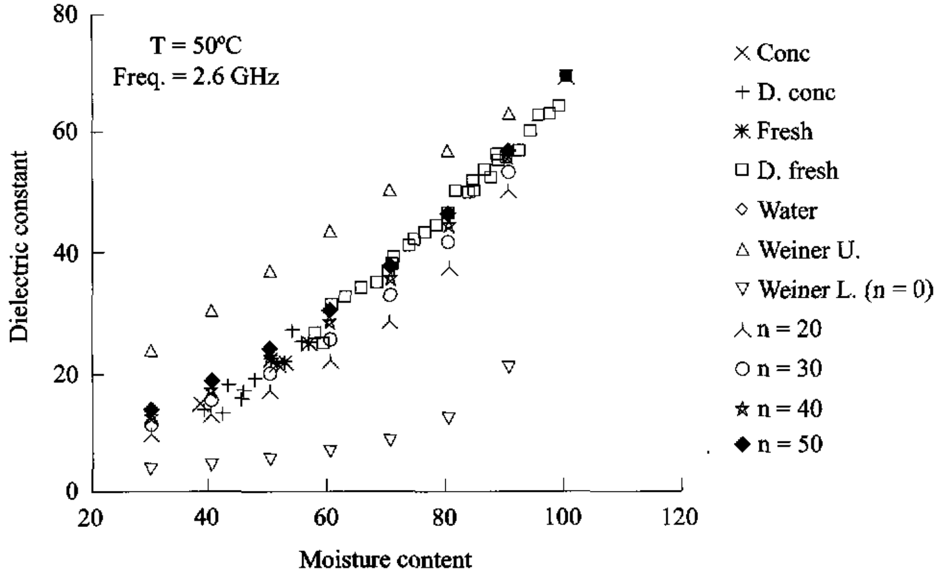

Figure 1e. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 2.6 GHz at 50°C.*

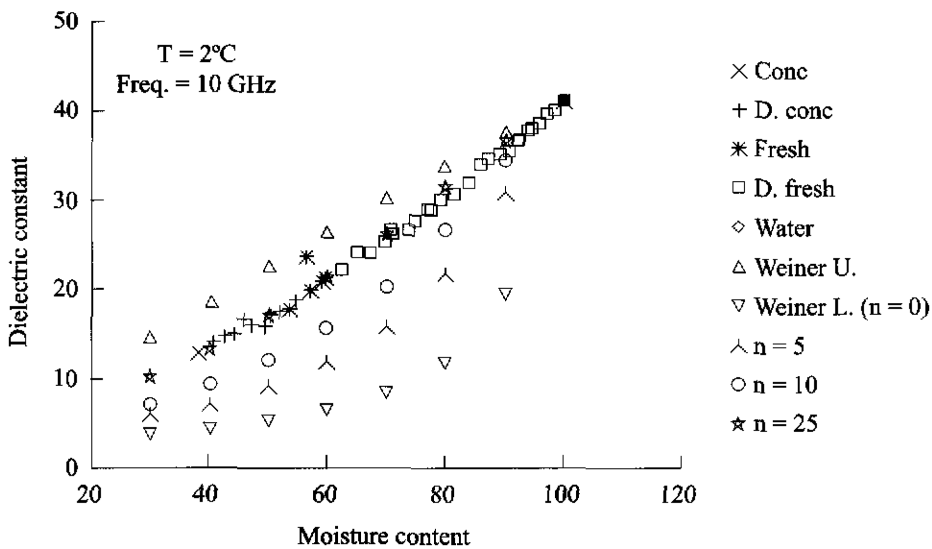

Figure 2a. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 10 GHz at 2°C.*

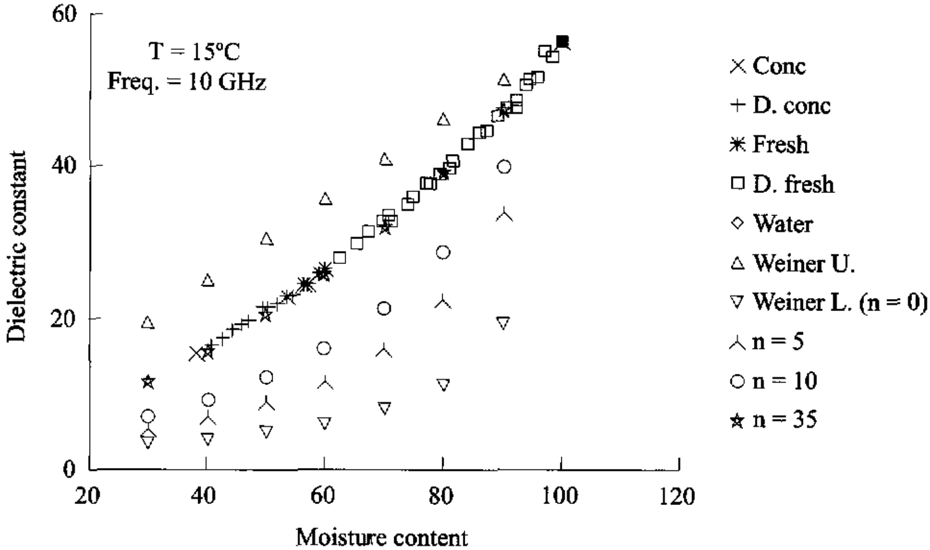

Figure 2b. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 10 GHz at 15°C.*

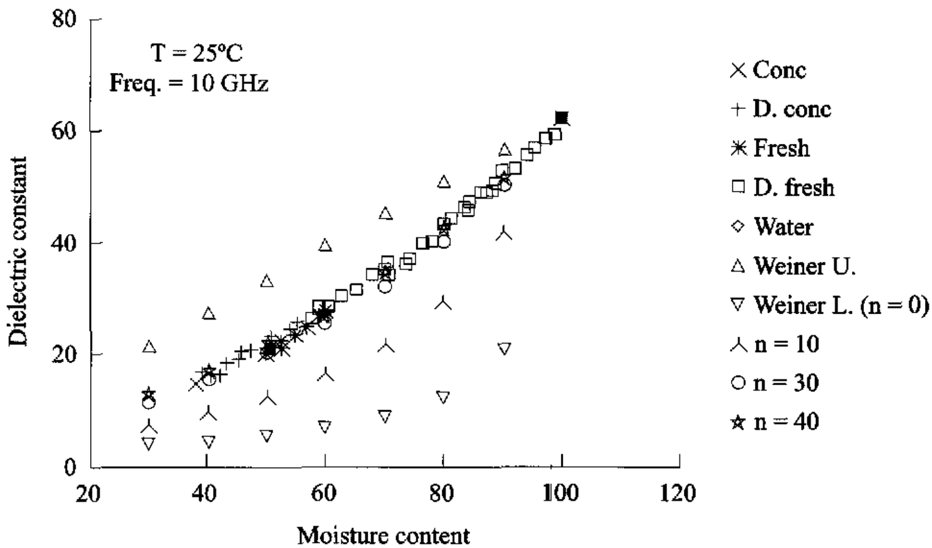

Figure 2c. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 10 GHz at 25°C.*

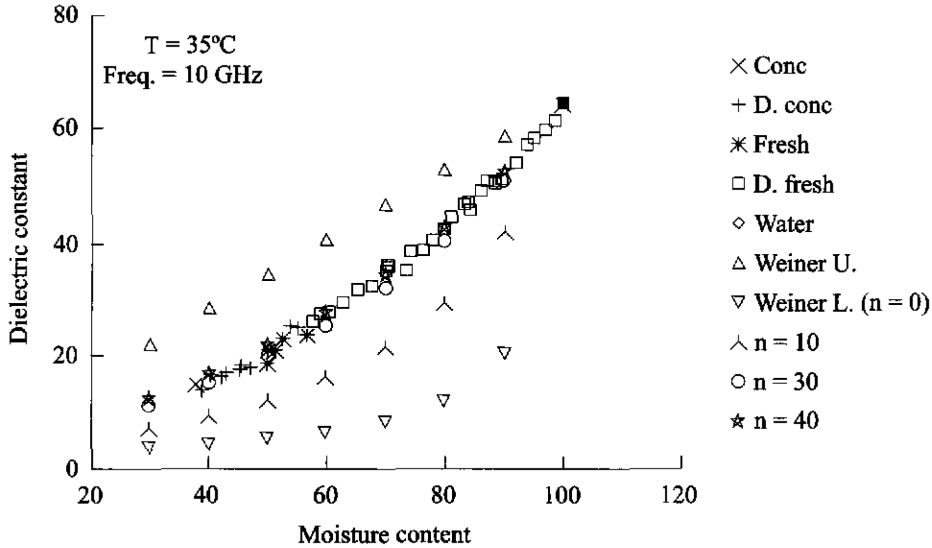

Figure 2d. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 10 GHz at 35°C.*

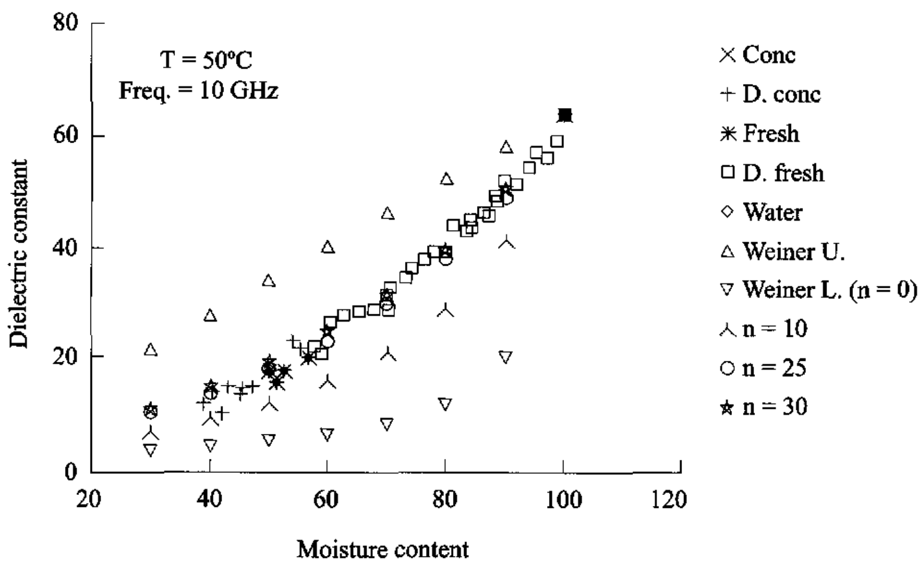

Figure 2e. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 10 GHz at 50°C.*

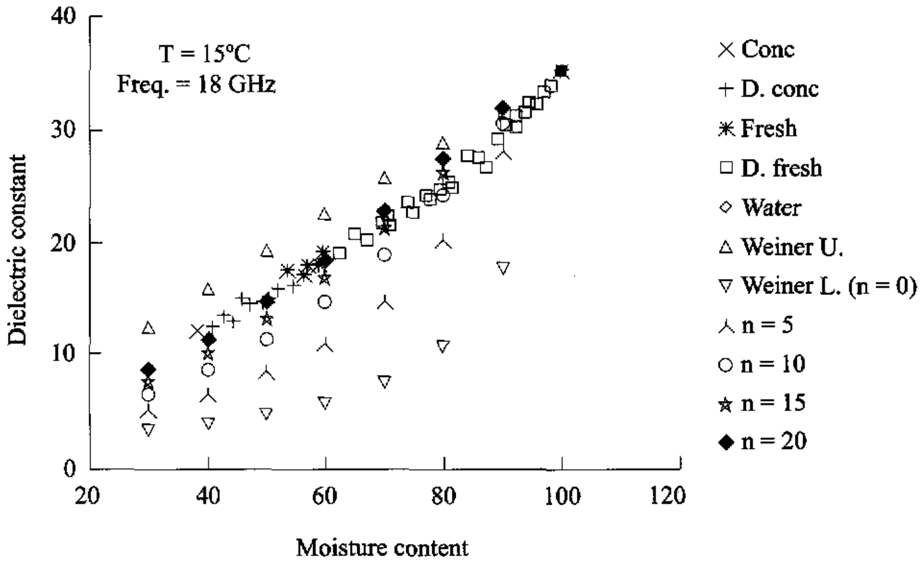

Figure 3a. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 18 GHz at 15°C.*

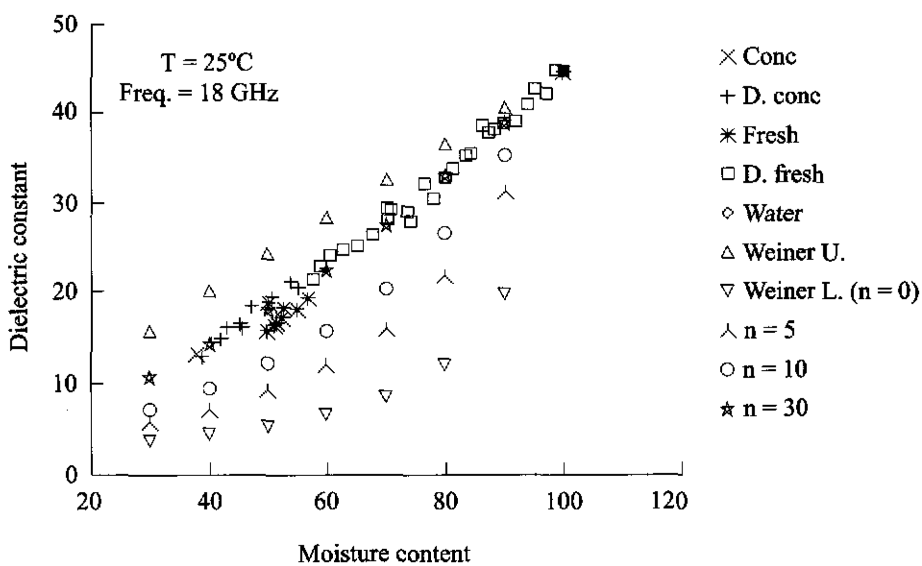

Figure 3b. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 18 GHz at 25°C.*

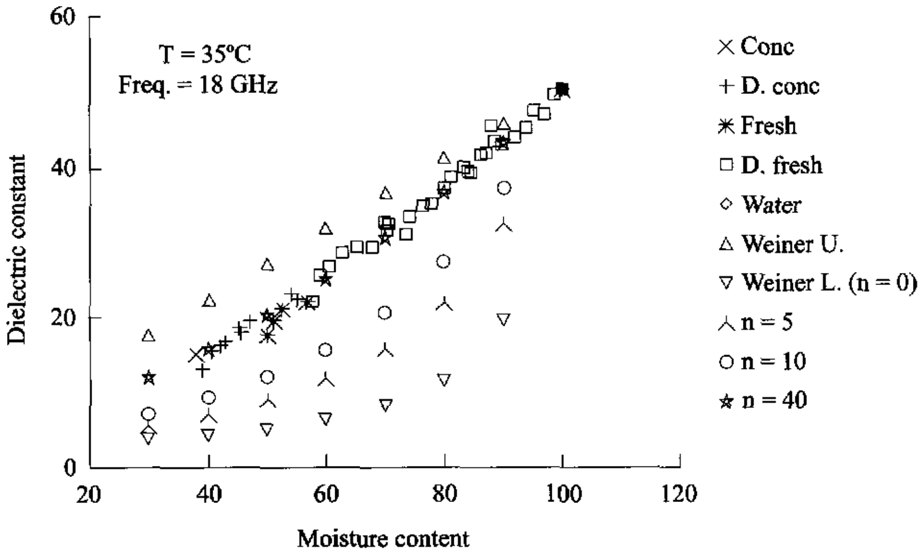

Figure 3c. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 18 GHz at 35°C.*

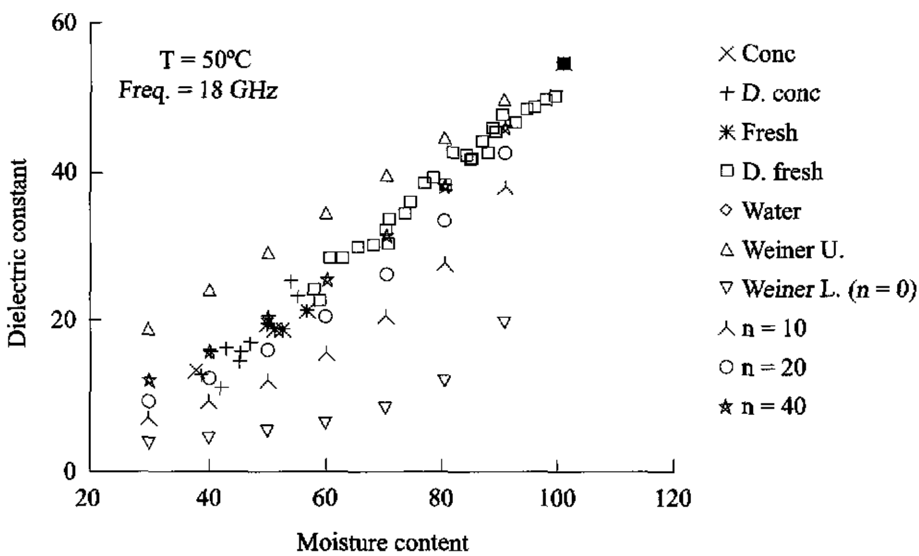

Figure 3d. Moisture dependence of the dielectric constant to illustrate the degree of binding η for Hevea latex *at 18 GHz at 50°C.*

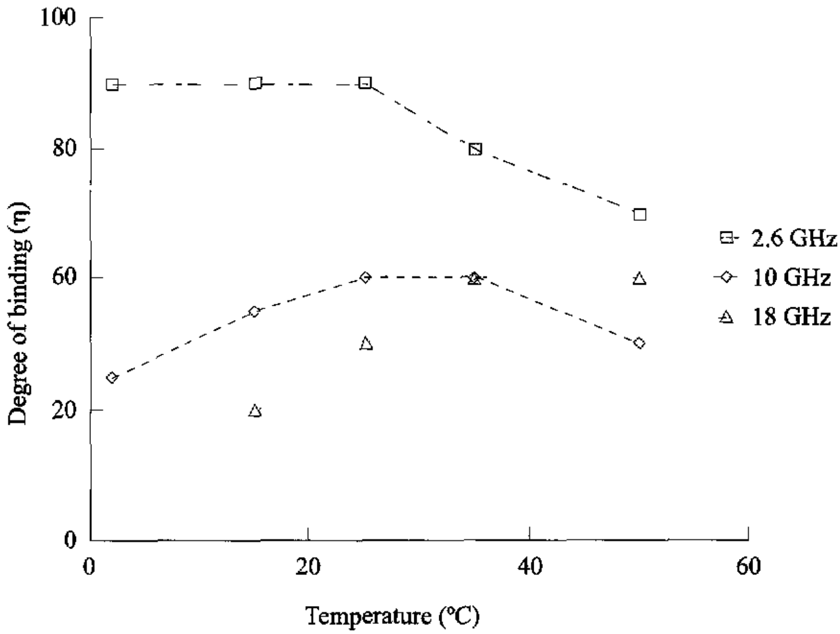

Figure 4. Variation of the degree of binding (η) with respect to temperature at selected frequencies.

common moist materials on the moisture content has been widely used. This dependence was produced by the contribution of the absorbed water molecules to the polarisation in the material. It was common that the magnitude of the contribution is proportional to the freedom of movement of the water molecules and hence to the degree of binding of the water. By applying the Weiner's theory of mixed dielectrics, the mixing condition η could be used to deduce the approximate shape or orientation assumed by the molecules. This is only applicable to the dielectric constant because the orientation of the molecules indicate the degree of polarisation. The shape of the molecules is assumed to be ellipsoidal.

Figures 1–3 summarise the values of ε' for Weiner's upper bound (η=∞) and lower bound (η=0) and degree of binding with respect to moisture content. *Figure 4* shows the variation of the degree of binding η with respect to temperature at selected frequencies with data shown in *Table 1*.

At 2.6 GHz, the degree of binding, η = 70 was constant from 2°C to 25°C but decreased with increasing temperature to 50 at 50°C. This meant that in the beginning, the water molecules are at the optimum level of polarisation before decreasing, resulting in a decrease in ε' where more of the molecules are now aligned parallel to the applied field . This could be due to the increase in thermal agitation with an increase in temperature that resulted in a damping factor inhibiting polarisation of the molecules. A similar pattern was expected at 10 GHz where η = 25 at 2°C, increasing to 40 at 25°C and stayed constant up to 35°C before decreasing to 30 at 50°C At 18 GHz η was much less than 20 at 2°C but reached 20 at 15°C increasing to 40 at 35°C and remained at this value until 50°C A distinct pattern could be observed in all the three curves of the degree of binding (*Figure 4*) where a plateau of constant η independent of temperature or constant polarisation existed before η decreased For 2 6, 10 and 18 GHz, η was constant in the temperature range of 2°C–25°C, 25°C–35°C and 35°C–50°C, respectively At 2 6 GHz η was constant at 70 while at 10 and 18 GHz η was 40 With increasing frequency, the region of constant η where polarisation was constant it shifted towards the higher temperature range This indicated that the degree of binding was not only dependent on moisture but very much affected by frequency and temperature

CONCLUSION

The variation between the dielectric constant of *Hevea* rubber latex and moisture content obtained experimentally were used to evaluate the mixing condition η and hence the degree of binding of the water molecules using Weiner's theory of mixed dielectrics A region of constant η existed, independent of temperature where polarisation was constant and beyond this region, η decreased with temperature η also decreased with frequency This was when more of the water molecules were aligned perpendicular to the field and polarisation decreased Changes in moisture, temperature and frequency might be used to detect changes in the degree of binding of the water molecules in *Hevea* latex This study provided valuable information concerning water-binding processes and process control in latex based industries

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