Jurnal Teknologi, 42(A) Jun. 2005: 59–74

MICROWAVE AND REFLECTION PROPERTIES OF PALM SHELL CARBON-POLYESTER CONDUCTIVE COMPOSITE ABSORBER

AHMAD ANAS YUSOF[1], WAN KHAIRUDDIN WAN ALI[2], THAREK ABD RAHMAN[3] & FARID NASIR ANI[4]

Abstract. The microwave properties of permittivity, ε and loss tangent, tan δ of conductive composite utilizing palm shell carbon mixed with unsaturated polyester resin were studied in the 8 to 12 GHz frequency range. The measurement of ε and tan δ emphasize on the influence of carbon concentration (mass %) of palm shell pyrolysed at 600, 700 and 800°C. It was observed that the increase of carbon concentration inside each measured composite influenced the increase of ε and tan δ condition. A microwave reflectivity test was also conducted by using 30% palm shell carbon in a 450 × 450 mm polyester composite panel. The result indicated that the composite thickness had influenced the reflection loss curve throughout the same microwave frequencies. The reflection loss up to –20 dB was possible in specific effective frequency ranges. This indicated the possibility of using carbon derived from palm shell residue in providing significant loss that contributed to microwave absorption, as well as an alternative in managing the increased of oil palm shell residues throughout the country.

Keywords: Conductive composite, microwave absorber, carbon, waste utilization

Abstrak. Sifat gelombang mikro seperti kebertelusan ε dan tangen kehilangan tan δ bagi suatu komposit bersifat pengalir menggunakan campuran karbon kelapa sawit dengan resin poliyester tak tepu dikaji pada julat frekuensi 8 GHz hingga 12 GHz. Pengukuran pada ε and tan δ menekankan kepada pengaruh kandungan karbon (jisim %) menggunakan kelapa sawit yang dipirolisis pada 600°C, 700°C and 800°C. Peningkatan kandungan karbon di dalam setiap komposit dilihat mempengaruhi peningkatan ε and tan δ. Ujian kebolehpantulan gelombang mikro turut dilakukan dengan menggunakan 30% karbon kelapa sawit di dalam panel komposit poliyester bersaiz 450 mm × 450 mm. Keputusan menunjukkan ketebalan komposit telah mempengaruhi kehilangan pantulan pada frequensi gelombang mikro yang sama. Kehilangan pantulan sehingga –20 dB dapat dikesan pada julat frekuensi tertentu. Ini menunjukkan kemungkinan penggunaan karbon daripada sisa kelapa sawit dalam menghasilkan kehilangan yang berkesan kepada penyerapan gelombang mikro, selain menjadi alternatif dalam menghadapi peningkatan sisa kelapa sawit di dalam negara.

Kata kunci: Komposit bersifat pengalir, penyerap gelombang mikro, karbon, penggunaan sisa

[1] Faculty of Mechanical Engineering, Kolej Universiti Teknikal Kebangsaan Malaysia. Locked Bag 1200, Ayer Keroh, 75450 Melaka.
[2&4] Faculty of Mechanical Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor.
[3] Faculty of Electrical Engineering, Universiti Teknologi Malaysia. 81310 Skudai, Johor.

1.0 INTRODUCTION

Malaysian oil palm plantation has seen unprecedented growth in the last four decades to emerge as the leading agricultural industry in the country. Besides oil production for domestic and industrial use, biomass has produced a wide range of utilization in residue and value added by-products. The need to reduce oil palm residues become increasingly important throughout the years [1]. Recent studies have shown that oil palm residuals such as trunks, shells and fibers could be recycled into useful material for other practical applications. Studies have proved that palm fibers can be used to produce medium density fiberboard [2]. Others have produced paper from chopped palm trunk [3]. The shells, which are finally turned into leftover slag from mill boilers, have been studied for a possible element in tarring roads. Besides being cracked into oil and resins, the final product of recycled palm shell residuals is in the form of charcoal or carbon [4-6].

The carbon from palm shell residue had been used as filler in producing conductive composite using polyester resin. In general, conductive composites are produced by filling non-conductive material with conducting particles such as carbon, metal flake, fibers and powder [7-9]. The conductivity of the composite usually provided by the carbon, while the function of the non-conductive material is to hold the material together in one piece [10, 11].

One of the potential applications of conductive composite studied is the application in microwave absorption [12, 13]. Microwave absorber had implemented the use of carbon as a dissipative element, which transform incoming wave into heat. This principle requires the material to be in a lossy or high loss condition, which can be represented by the high value of tan δ. Although the potential utilisation of conductive composite in microwave absorber has been rigorously studied using carbon of various origin integrated with non-conductive material such as rubber and polyurethane, only few researches were focusing on using carbon derived from waste and residues. The application of the conductive composite studied can also be associated in electrical insulation, radar absorber and electromagnetic interference reduction [14-18].

2.0 SAMPLE PREPARATION

The oil palm shells, which were the raw material in producing carbon, were obtained from Federal Land Development Authority (FELDA) Palm Oil Mills in Kulai, Johor. The palm shells were later grinded and sieved into fine particles. Particle size was set at maximum size of 75 μm, using BS 410 stainless steel mesh wire. The use of fine particles can cause better mixture in the composite concentration and increase the particle contact inside the host in promoting optimum conductivity.

This was followed by drying palm shell residues in the Memmert 200 oven for about 24 hours to eliminate moisture content that might influence the carbon yields in the process. Initial weight for the palm shell was set at 120 grams before placed inside a fluidised bed combustor, in which the transformation of palm shells into carbon powder was carried out. Nitrogen was used as the inert gas during pyrolysis. The pyrolysis or carbonisation process started with constant nitrogen gas flow of 1.5 liter/min and let alone for one hour. The ranges of pyrolysis (carbonisation) temperature for palm shell residues in N2 were carried out at 600, 700, and 800°C, with heating rate of 10°C per minute. The surface area of the carbon was later characterised by nitrogen adsorption analysis from Micromeritic ASAP 2010. The results showed that the pyrolysed carbon at 600, 700 and 800°C had resulted in a surface area of 159.4, 184.5 and 195.53 m^2/g respectively.

The palm shell carbons were later mixed with unsaturated polyester resin as the matrix. They were prepared by using different levels of concentrations and pyrolysis temperatures as shown in Table 1, in which the carbon and resin were well mixed before dispensed into selected mould. The process involved adding up of 3% peroxide catalyst known as MEKP (Methyl Ethyl Ketone Peroxide) in order to accelerate the

Table 1 Samples preparation based on pyrolysis temperature (°C) and carbon concentration (%)

Pyrolysis temperature (°C) of carbon	**Concentration (mass %)**	
	Carbon particles	**Unsaturated polyester resin**
600	5.0	95
	10	90
	15	85
	20	80
	25	75
	30	70
700	5.0	95
	10	90
	15	85
	20	80
	25	75
	30	70
800	5.0	95
	10	90
	15	85
	20	80
	25	75
	30	70

curing process. This created possible condition for the mixture to be hardened at room temperature for about 3 hours. The composites were dried in a furnace with temperature up to 150°C for complete hardening. They were later mechanically shaped and precisely cut into blocks of $22.86 \times 10.16 \times 5$ mm in dimension, according to the required test.

3.0 EXPERIMENTAL

3.1 Microwave Properties

The permittivity and loss tangent of the samples were calculated using waveguide transmission line measurement by applying short circuit method during the measurement. The complex propagation function of the material in waveguide, γ_2, was related to the complex permittivity by using the following equation [19]:

$$\gamma_2 = \frac{2\pi}{\lambda_o}\sqrt{\left(\frac{\lambda_o}{\lambda_c}\right)^2 - \varepsilon_r{}' + j\varepsilon_r{}''} \tag{1}$$

Therefore:

$$\varepsilon_r{}' - j\varepsilon_r{}'' = \left(\frac{\lambda_o}{\lambda_c}\right)^2 - \left(\frac{\gamma_2\lambda_o}{2\pi}\right)^2 \tag{2}$$

where λ_c is the cut-off wavelength for the wave-guide used. For propagation in rectangular waveguide, the cut-off wavelength can be related with waveguide wavelength, λ_1 and free space wavelength, λ_o:

$$\lambda_1 = \frac{\lambda_o}{\sqrt{1 - \left(\frac{\lambda_o}{\lambda_c}\right)^2}} \tag{3}$$

or simply:

$$\frac{1}{\lambda_1^2} = \frac{1}{\lambda_o^2} - \frac{1}{\lambda_c^2} \tag{4}$$

From Equations (2) and (4), the complex permittivity value can be represented by:

$$\varepsilon_r{}' - j\varepsilon_r{}'' = 1 - \left(\frac{\lambda_o}{\lambda_1}\right)^2 - \left(\frac{\gamma_2\lambda_o}{2\pi}\right)^2 \tag{5}$$

or

$$\varepsilon_r = 1 - \left(\frac{\lambda_o}{\lambda_1}\right)^2 - \left(\frac{\gamma_2 \lambda_o}{2\pi}\right)^2 \tag{6}$$

which γ_2 is the complex propagation function in form of:

$$\gamma_2 = \alpha + j\beta \tag{7}$$

where α is the attenuation constant and β is the phase constant of the propagation function. The value of γ_2 can be calculated by using the formula given by [20]:

$$\frac{\tan\text{h}\ \gamma_2 d}{\gamma_2 d} = \left(\frac{-j\lambda_1}{2\pi d}\right) \cdot \left(\frac{\dfrac{E_{\min}}{E_{\max}} - j\tan\dfrac{2\pi x_o}{\lambda_1}}{1 - j\dfrac{E_{\min}}{E_{\max}} - \tan\dfrac{2\pi x_o}{\lambda_1}} \right) \tag{8}$$

where x_o is the distance of first minimun position from the sample, $E_{\min}/E_{\max}$ is the voltage standing wave ratio and d is the sample thickness. In most general form, the permittivity of a medium is written as:

$$\varepsilon_r = \varepsilon_r{}' - j\varepsilon_r{}'' \tag{9}$$

where $\varepsilon_r{}'$ and $\varepsilon_r{}''$ is the real and imaginary part of the complex relative permittivity. The loss tangent of the sample is calculated by:

$$\tan\delta = \frac{\sigma + \omega\varepsilon_r{}''}{\omega\varepsilon_r{}'} \tag{10}$$

where ω is the angular frequency and σ represent the conductivity of the sample. The measurement of the conductive composites were performed by using IFR Microwave System Analyzer and a rectangular WR-90 waveguide operating from 8 to 12 GHz frequency range (X-band) as shown in Figure 1. The signal source output from the integral spectrum analyzer had been used to provide signals in the same frequency during the measurement. The reflected signals was analyzed by the spectrum analyser input, where the measured parameters was later calculated to acquire the value of ε and tan δ. Calibration procedure was executed by using two sets of measurement at a time, one with and without the sample inside the waveguide to determine the microwave properties in the measurement. A sample comparison using measurement of pure unsaturated resin was also conducted. The conductivity of the sample was measured by using a digital multimeter.

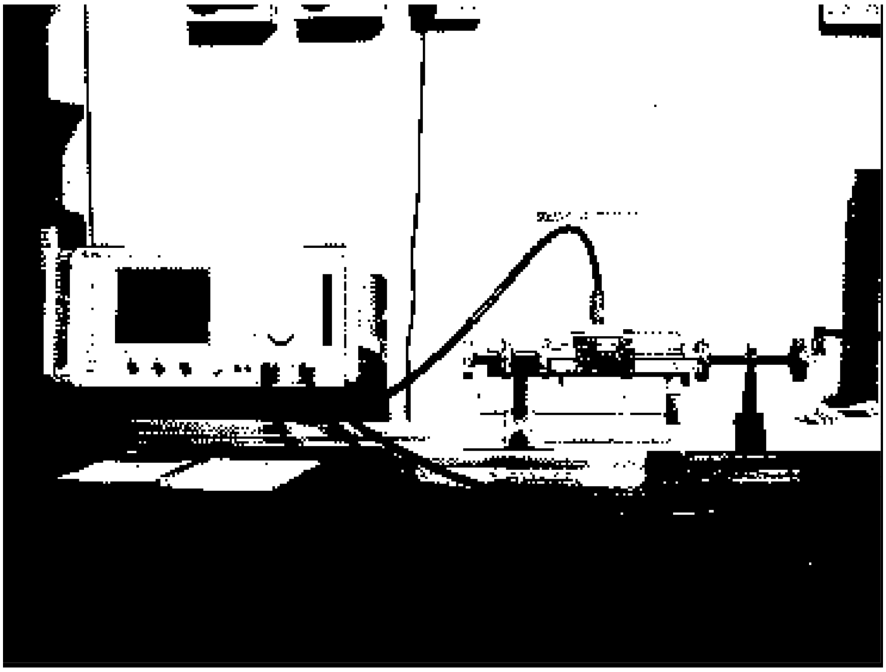

Figure 1 Microwave characterization using waveguide transmission line set-up

3.2 Reflection Properties

The measurement in microwave reflectivity involves the preparation of composite of 450 × 450 mm in cross-sectional area with various thickness. The same preparation method was used as shown in Figure 2, where the composites were mixed using palm shell carbon and polyester resin. The palm shell carbon was obtained from Pacific Activated Carbon Factory from Pasir Gudang, Johor in order to produce bigger composites panels for the test.

The test was executed in a semi-anechoic chamber backed with pyramidal absorber by using free space reflectivity technique, were the amplitude of the reflection loss was relatively measured to a metal plate reference, as referred in Figure 3. Such procedure of initially measuring the reflection of the metal plate was essential in calibrating the system. The signal was generated in the same microwave region (X-band) through a transmitting horn antenna using Marconi Microwave System Analyser series 6204. The reflected signals from 10° incidence angle were received by another horn antenna at which it was analyzed by using ADVANTEST model R3271A spectrum analyser.

The reflection of both set up was compared to evaluate its reflection properties. The difference in signal level between these two conditions indicates the reflection loss of the sample. In this case, the power reflection coefficient data from the measured reflection from the panels is always referred to the metal panel [18]:

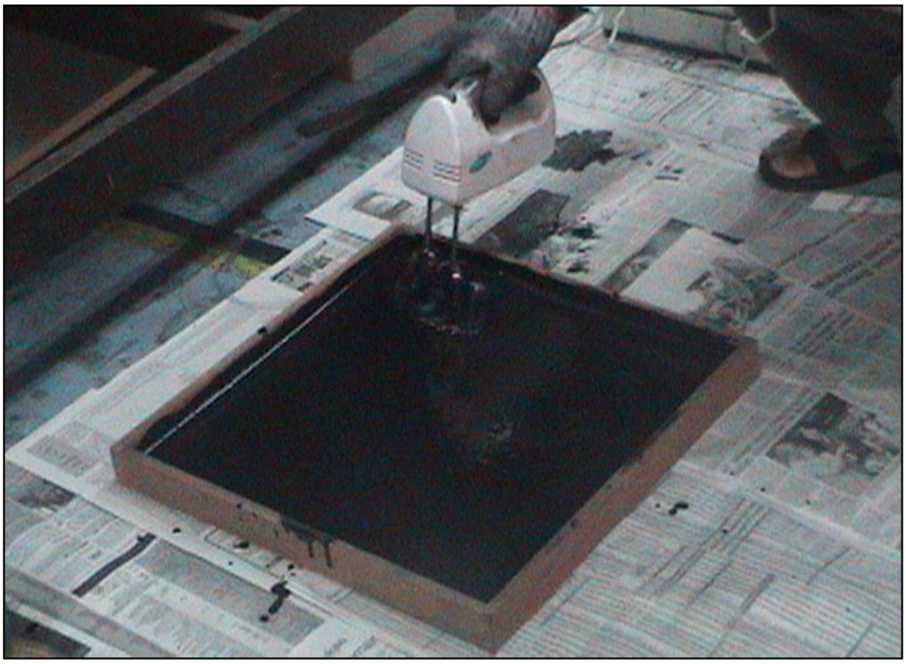

Figure 2 Samples preparation using 450 × 450 mm wooden mould

Figure 3 Free space reflectivity measurement set-up

$$R(dB) = 10 \log \frac{\Gamma_{absorber}}{\Gamma_{reference}} \tag{11}$$

4.0 RESULTS AND DISCUSSION

4.1 Microwave Properties

The effect of mixing pyrolysed palm shell carbon at 600°C with unsaturated polyester resin with different concentrations (%) was investigated. Figure 4 shows an increase in ε'_r with respect to the increasing of carbon concentration (%) inside each composite throughout the X-band frequencies. The dependence of ε'_r with respect to the carbon loading of the composite showed that the pyrolysed carbon at 600°C influenced a small increase in the microwave properties of the material.

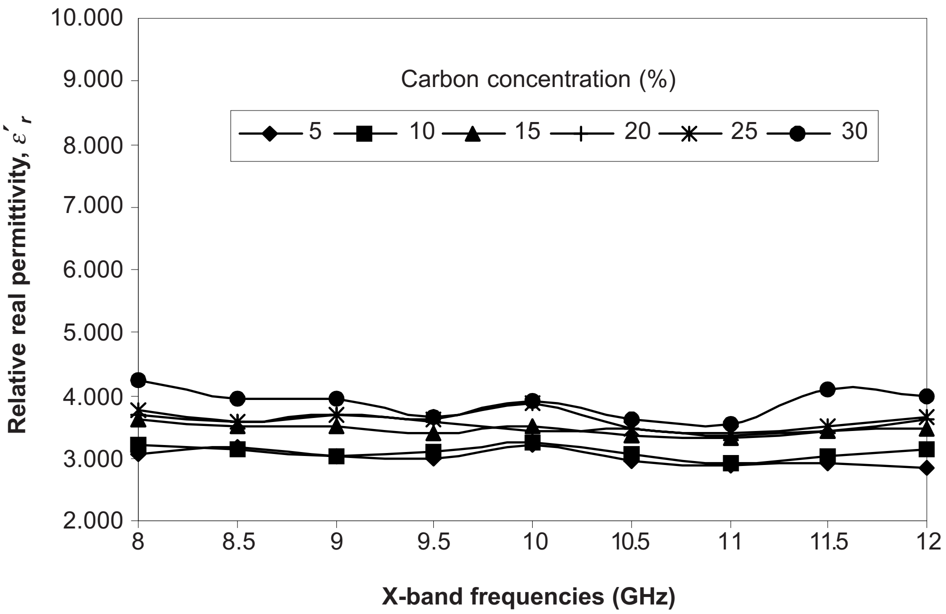

Figure 4 Relative real permittivity, ε'_r, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 600°C)

The lossy condition could be observed in Figure 5, where increase tan δ could be observed with the increase of carbon concentrations. A similar pattern of tan δ can be observed in every concentration, with maximum loss obtained at 10 GHz in nearly all concentration. The small difference in each concentration, however, suggested that the carbon produced at 600°C has a limitation effect over the lossy condition of the mixture.

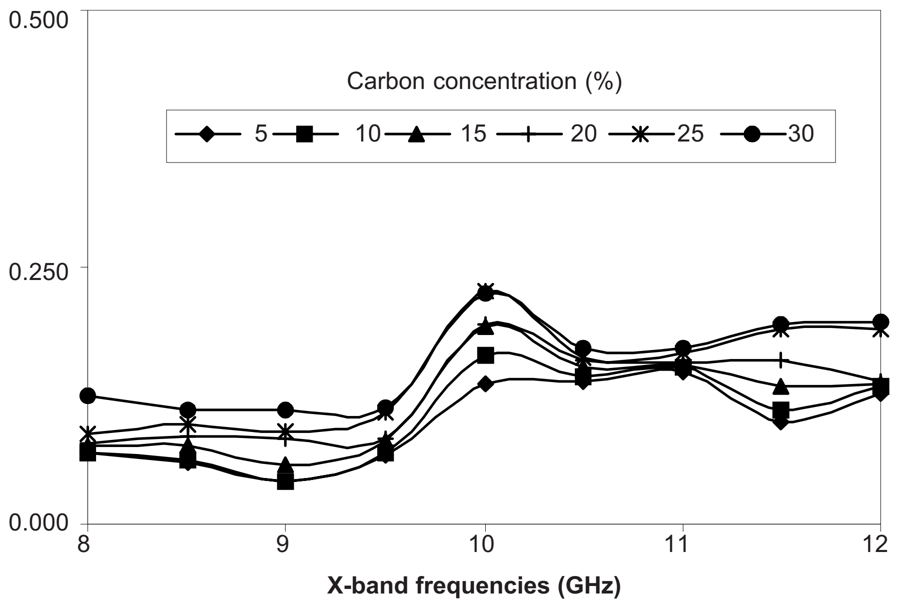

Figure 5 Loss tangent, tan δ, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 600°C)

Microwave properties of polyester resin mixed with pyrolysed palm shell carbon at 700°C had showed significant increase of ε'_r and tan δ with respect to the increase of carbon concentrations (%). Figures 6 and 7 show the results of ε'_r and tan δ based on composites by using pyrolysed palm shell carbon at 700°C.

The result obtained had showed higher values of ε'_r and tan δ with respect to the composites prepared at 600°C. A better increase in the microwave properties can be observed by the utilisation of pyrolysed carbon at 700°C.

Significant tan δ could be observed in Figure 7 to increase from 5 to 30% concentration across the chart. At 8 GHz, carbon loading of 5% concentration showed minimum tan δ while maximum tan δ was observed at 10 GHz in 30% concentration. Figure 7 also showed that an abrupt increase in tan δ at 30% concentration with respect to the other concentration, suggesting the influence of conductivity in promoting the loss.

Figures 8 and 9 illustrate a higher increase of ε'_r and tan δ with respect to the increase of carbon concentrations (%) using carbon pyrolysed at 800°C. The results also substantiated better increments of ε'_r and tan δ over other samples which was mixed with carbon pyrolysed at 600°C and 700°C.

Figure 9 shows the increasing trends of losses from tan δ of 5% concentration, up to a maximum losses condition at 30% carbon concentration. It showed that the maximum tan δ occurs at 10 GHz in polyesters resin mixed with 30% carbon concentration while minimum tan δ is observed with 5 % carbon concentration at 8.5

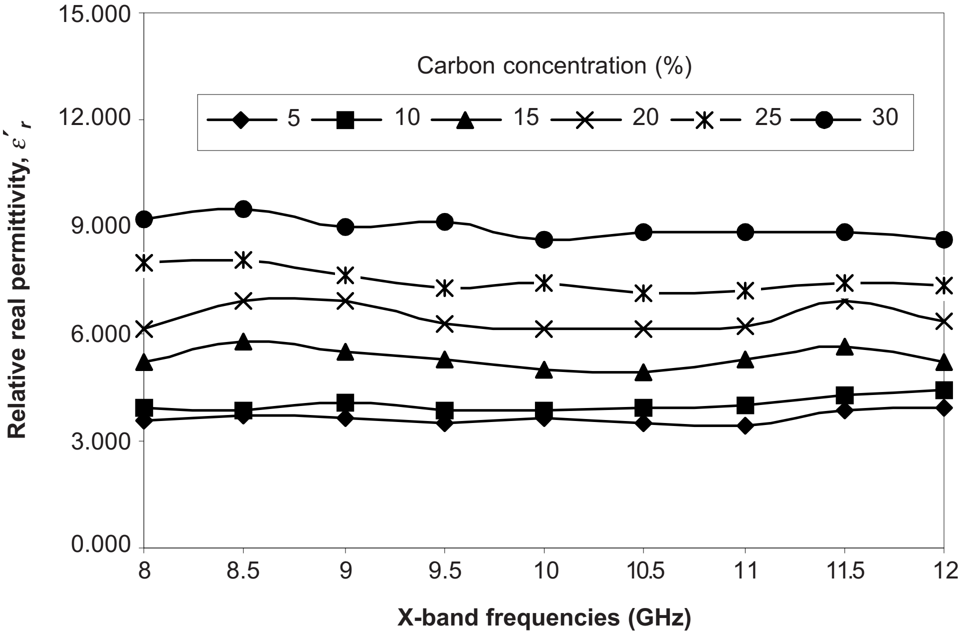

Figure 6 Relative real permittivity, ε'_r, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 700°C)

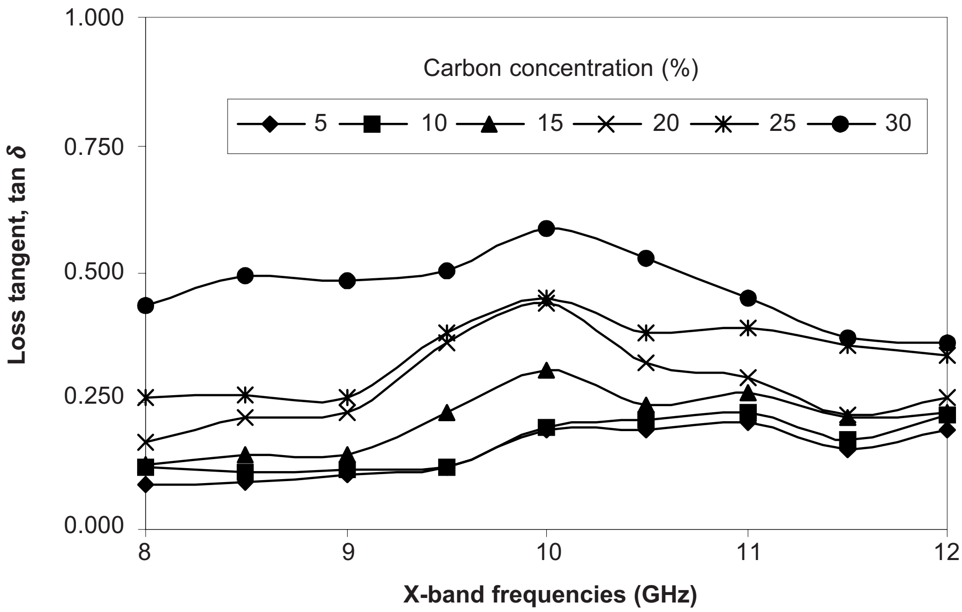

Figure 7 Loss tangent, tan δ, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 700°C)

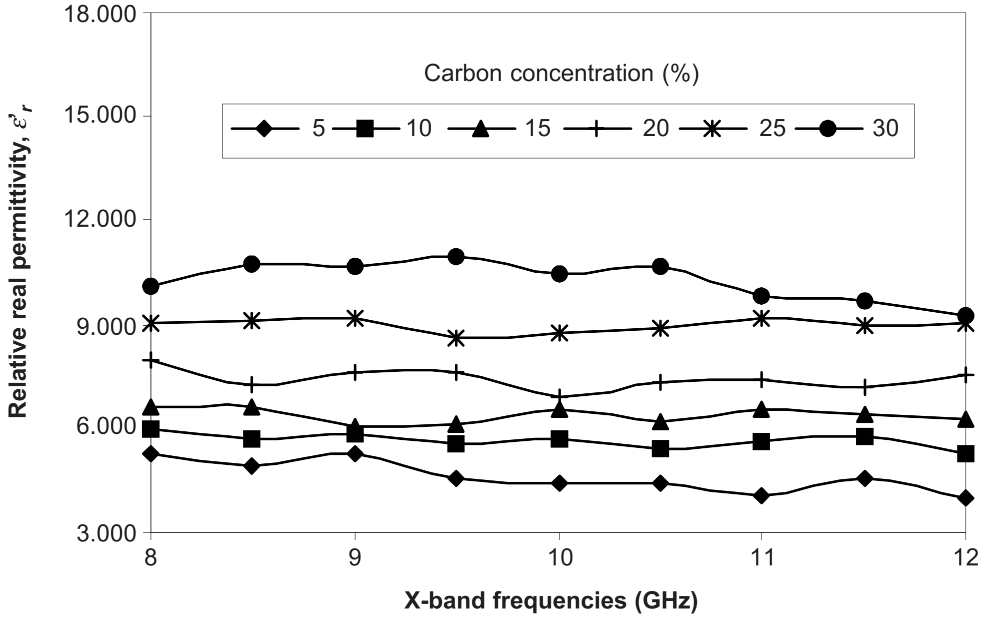

Figure 8 Relative real permittivity, ε'_r, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 800°C)

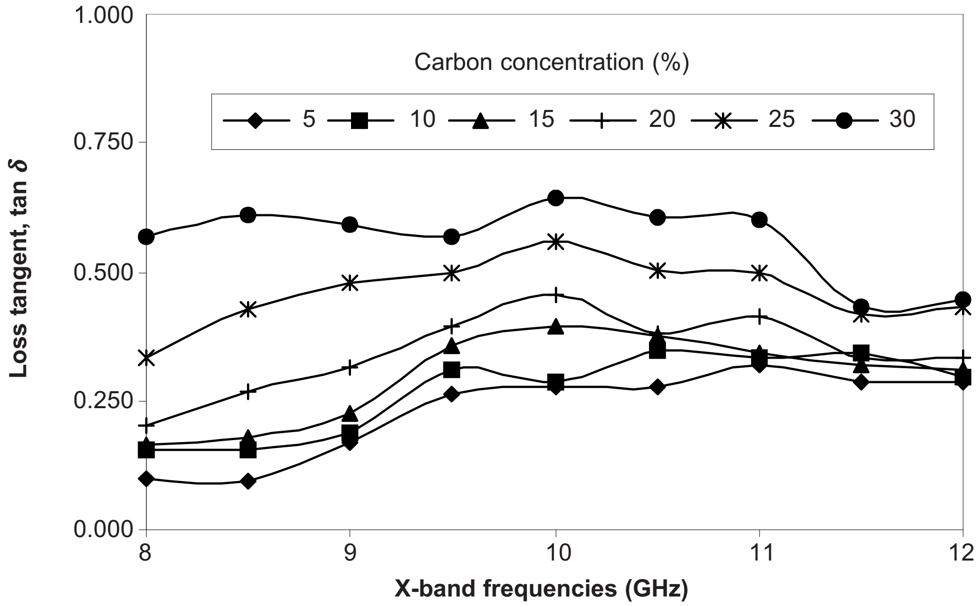

Figure 9 Loss tangent, tan δ, at different carbon concentrations (%) in X-band frequencies (Pyrolysed carbon of 800°C)

GHz. It also showed a decreasing trend of tan δ starting from 11.5 GHz onwards. The same trend can also be seen in mixed resin with pyrolysed carbon at 700°C.

The highest tan δ of the composite with 30% carbon loading concluded that the increase of carbon loading helps in accumulating better loss in microwave absorber. The results also suggested that the high frequency range used in the measurement had influenced the measured tan δ. In this case, the imaginary permittivity, which contributed to the lossy condition of the composite, started to decrease.

4.2 Reflection Properties

Figure 10 illustrates the reflective behavior in terms of thickness and frequency. The microwave absorption of carbon-loaded absorber is basically related to the dielectric loss. Carbon can be used in microwave absorber most effectively in the frequency range where the imaginary component becomes larger than the real component of complex permittivity, thus introducing a high loss condition of tan d. However, in this measurement, large imaginary component of complex permittivity does not necessarily guarantee an excellent microwave absorber.

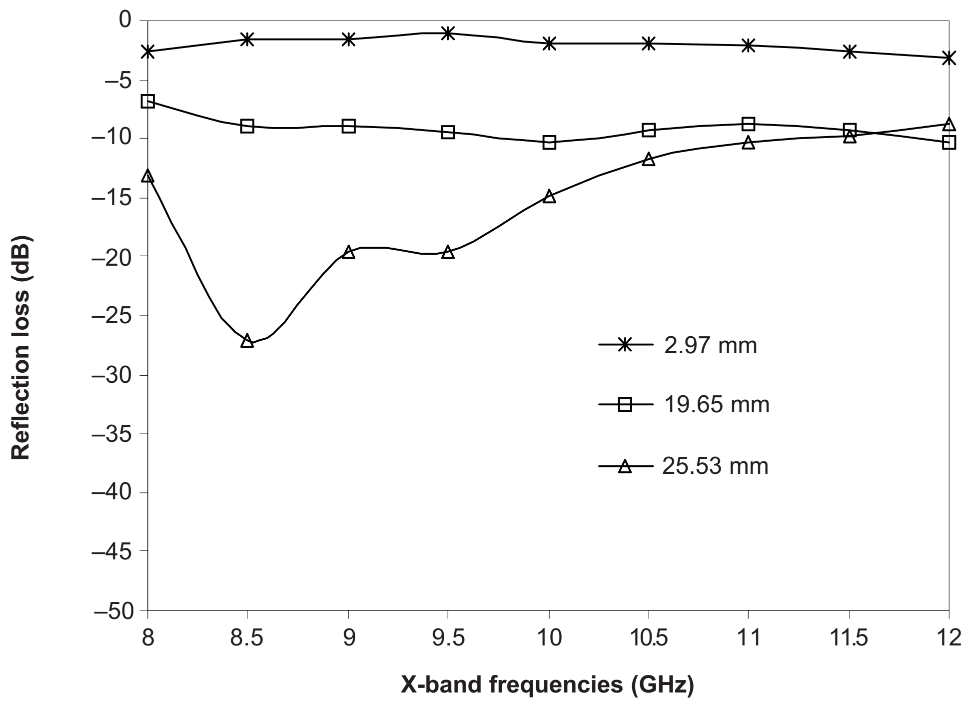

Figure 10 Reflection loss of composites with different thickness of 2.97, 19.65 and 25.53 mm

The figure shows a reflection loss in X-band frequencies, with reference to composite thickness of 2.97, 19.65 and 25.53 mm. Moderate reflection loss around

–3 dB can be achieved by using composite thickness of 2.97 mm in the whole microwave region. Reflection loss of maximum at –10.31 dB can be obtained for 19.65 mm thickness in the same bandwidth.

It was observed that at greater thickness of 25.53 mm, the performance of the composite in the X-band frequency improved with reflection loss up to –27.09 dB could be obtained at 8.5 GHz. The results showed that better performance occurred at –19.57 and –19.62 dB for frequency of 9 and 9.5 GHz respectively. The optimum performance could be observed below –20 dB at frequency ranged from 8.2 to 8.9 GHz. Moderate performance can be observed below –10 dB in the X-band frequencies ranged from 8 to 11.2 GHz in the same thickness.

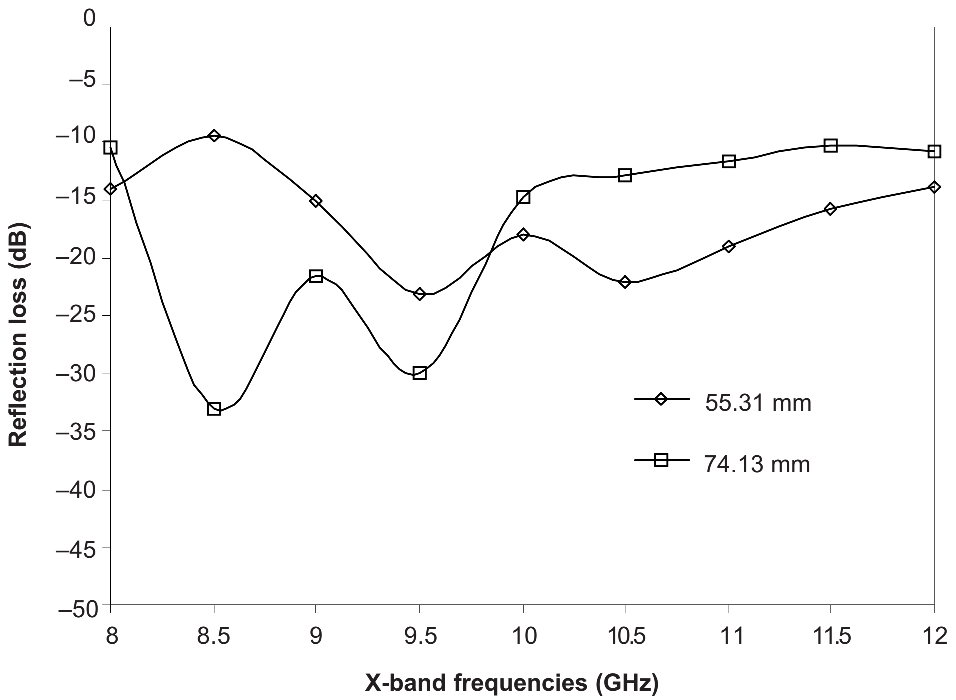

Figure 11 Reflection loss of composites in different thickness of 55.31 and 74.13 mm

Figure 11 illustrates the reflection loss characteristic of two composites at 55.31 and 74.13 mm thickness. Reflection loss of up to –20 dB can be observed for composites with 55.31 mm thickness. The shifting in thickness resulted in optimum reflection loss of –23.16 and –22.13 dB at 9.5 and 10.5 GHz respectively. Decay in reflection loss was observed beyond 10.5 GHz, but still showed better loss from previous thickness.

Maximum thickness of 74.13 mm displayed an improved performance in reflection loss. In this case, significant loss of up to –20 dB can be observed in the frequency ranged from 8.2 to 9.8 GHz, where maximum performance was obtained above –20 dB. The figure also illustrates narrowband reflection loss up to a maximum of –32.97 and –30.07 dB for frequency of 8.5 and 9.5 GHz respectively. Good reflection loss of –21.63 dB was also observed at 9 GHz. The reflection loss of the composite had showed moderated loss at broadband frequency ranged around –10 dB beyond 9.5 GHz in the same X-band frequency.

5.0 CONCLUSION

Reflection and microwave properties in the X-band region using conductive composites loaded with palm shell carbon were studied. Three types of pyrolysed carbon, each at 600, 700 and 800°C were mixed with various mass concentrations of carbon ranging from 5% up to 30%. The change in the conductivity is caused by the formation of continuous conducting network throughout the polymer matrix. The critical concentration in the composite at which a sudden increase in conductivity level was observed at 20 and 15% concentration using carbon pyrolysed at 700 and 800°C respectively. The increase in pyrolysis temperature had promoted a better-structured carbon, which can be observed by the increase amount of carbon surface area. Such increase also produced conductivity of the carbon, and thus promoting the increase of lossy condition. This could be observed by the higher value of microwave properties in the same concentration in each composite. There was no conductivity detected in the composites loaded with pyrolysed carbon at 600°C, suggesting that higher concentration of carbon is needed in the composite to promote an increase in conductivity level.

In the microwave reflectivity measurement, the performance of microwave absorber was influenced by the physical thickness of the absorber, each loaded with the same 30% carbon concentration. In the measurement, the reflection loss curve was observed to move towards effective frequency ranges at the optimum thickness of the composite. Such alteration in composite thickness had shifted the performance of the absorber accordingly. In this case, at constant carbon concentration, optimum thickness of the composites could be considered as the main parameter that influenced the reflection loss. This condition could be satisfied if the absorbers had impedance value similar to that of free space. As impedance values also depend on dielectric constant and thickness of the absorbers, the existence of an optimum thickness would satisfy the best performance of absorption.

ACKNOWLEDGEMENTS

The authors wish to thank the Ministry of Science, Technology and Environment, Malaysia under the IRPA Research Program for the research grants awarded and Universiti Teknologi Malaysia for the support in carrying out the research work.

REFERENCES

[1] Ma, A. N., T. S. Toh, and N. S. Chua. 1999. Renewable Energy From Oil Palm Industry. In Gurmit, S. *Oil Palm and The Environment - A Malaysian Perspective*. Kuala Lumpur: Malaysian Oil Palm Growers' Council. 253-259.

[2] Rahim, and Khozirah. 1991. Effect of Wood/ Gypsum Ratio and Density on Strength Properties of Gypsum Bonded Particleboard from Oil Palm Stems. *Journal of Tropical Science*. 4(1): 80-86.

[3] Kamarudin, H, D. Ariffin, S. Jalani, and M. Y. Mohd Nor. 1999. Pulp and Paper from Oil Palm Biomass. In Gurmit, S. *Oil Palm and The Environment - A Malaysian Perspective*. Kuala Lumpur: Malaysian Oil Palm Growers' Council. 213-225.

[4] Husin, M., R. Ramli, A. Mokhtar, and A. Abdul Aziz. 2002. Research and Development of Oil Palm Biomass Utilization in Wood-based Industries. *Palm Oil Development No 36*. Malaysian Palm Oil Board. 1-5.

[5] Islam, M. N., N. A. Farid, and Z. Ramlan. 1999. Pyrolytic Oil From Fluidised Bed Pyrolysis of Oil Palm Shell and its Characterisation. *Renewable Energy*. 17: 73-84.

[6] Hussain, A. 1993. Kesan Teknik Penyediaan Karbon Teraktif Daripada Tempurung Kelapa Sawit Terhadap Keliangan dan Keupayaan Penjerapan. *Jurnal Teknologi*. 22: 51-70.

[7] Kaynak, A., A. Plat, and U. Yilmazer. 1996. Some Microwave and Mechanical Properties of Carbon Fiber Polypropylene and Carbon-Black Polypropylene Composites. *Material Research Bulletin*. 31(10): 1195-1206.

[8] Yamamoto, K, T. Matsuura, M. Mondo, and S. Tsukawali. 2001. Development of Electromagnetic wave Absorber Composed of Charcoal Powder and Unsaturated Polyester Resin. Technical Report. Tobu Industrial Research Centre. Japan.

[9] Naidasham, K., and P. K. Kadaba 1991. Measurement of the Microwave Conductivity of a Polymeric Material with Potential Application in Absorbers and Shielding. *IEEE Transaction on Microwave Theory and Techniques*. 39(7): 1158-1164.

[10] Raghavendra, S. C., I. A. Khan, P. M. Hadalgi, and A. B. Kulkarni. 2003. Microwave Reflection and Radiation Properties of Polyester-fly Ash Composites. Abstract. International Ash Symposium University. University of Kentucky.

[11] Jachym, B. 1982. Conduction in Carbon Black-Doped Polymers. In Sichel E. K. *Carbon Black - Polymer Composites: The Physics Of Electrically Conducting Composites*. New York: Marcel Dekker Incorporated. 103-132.

[12] Neelakanta, P. S., and J. C. Park. 1995. Microwave Absorption by Conductor Loaded Dielectric. *IEEE Trans on Microwave Theory and Technique*. 43(6): 1381-1383.

[13] Srivastana, G. P., P. P. Singh, and J. Nath. 1992. Microwave Absorber Composed of Rubber, Carbon and Ferrite. Asia Pacific Microwave Conference. Adelaide. 239-242.

[14] Chambers, B. 1995. Symmetrical Radar Absorbing Structures. *Electronics Letters*. 31(5): 404-405.

[15] Kumar. 1987. Acetylene Black: A Single Layer Microwave Absorber. *Electronic Letters*. 23(5): 184-185.

[16] McCauley, J. W., B. M. Halpin Jr., T. Hynes, and S. D. Eitelman. 1980. Radar Absorptive Ferrite/Resin Composites From Industrial Effluent. *Ceramic Engineering and Science Procurement*. 1:356-369.

[17] Pitman, K. C., M. W. Lindley, D. Simkin, and J. F. Cooper. 1991. Radar Absorbers: Better by Design. *IEEE Proceedings-F*. 138(3): 223-228.

[18] Knott, E. F, J. F. Shaeffer, and M. T. Tulley. 1993. *Radar Cross-section*. 2nd Ed. London: Artech House.

[19] Torgovnikov, G. I. 1993. *Dielectric Properties of Wood and Wood-based Materials*. Germany: Springer Verlag.

[20] Robers, S., and A. R. Von Hippel. 1946. A New Method for Measuring Dielectric Constant and Loss in the Range of Centimeter Waves. *Journal of Applied Physics*. 17: 610-616.