Journal of Bangladesh Academy of Sciences, Vol. 38, No. 1, 39-47, 2014

EFFECTS OF ADDITIONAL AMOUNT OF CARBON MICRO-COILS ON THE DIELECTRIC PROPERTIES OF ITS COMPOSITE SHEET

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ABSTRACT

The morphology and dielectric properties of carbon micro-coils (CMC) composite sheet were examined. Frequency dependence of dielectric parameters; dielectric loss angle (θ), dielectric loss tangent ($tan\delta$) and relative dielectric constant (ε_r) were measured on single and double-helix composite sheet. It was found that these parameters of CMC/polysilicone composites extensively changed with the additional amount of CMC with polysilicone resine. It was also observed that the higher additional amount (wt%) of CMC is appropriate for high response.

Key words: Carbon micro-coils, Dielectric parameters

INTRODUCTION

Carbon micro-coils (CMC) with 3D-helical/spiral structure with micrometer to nanometer order dimensions have drawn attention in biotechnology and nanotechnology. But materials with 3D-helical/spiral structure with micrometer to nanometer order dimensions have not been commercially available. Motojima *et al.* (2003) have prepared regularly coiled carbon fibers (carbon coils) with 3D-helical/spiral structures with high coil yield (Motojima and Chen 2004). The CMCs are expected to have many unique characteristics, such as good chiral conductivity, high surface area (Chen *et al.* 1999), very high super-elastic property (Chen *et al.* 2002), very high interaction ability with electromagnetic waves and many potential applications such as tunable micro-devices/sensors, electromagnetic absorbers, energy changing materials, hydrogen absorbers, chiral catalysts, activators of organisms, etc. (Fujii *et al.* 2002, Furuta *et al.* 2004, Kato *et al.* 2003, Varadan *et al.* 1994, Guerin *et al.* 1992, Motojima *et al.* 2003). These excellent characteristics are mainly affected by the special chiral/coiling morphology and the micro- to nano-meter order dimensions. Many researchers have been focused on the preparation of carbon micro-coils and found regularly micro-coiled carbon fibers with single and double helix and chiral conformation (referred to as “carbon micro-coils” or “CMC” hereafter) could be obtained by the Ni-catalyzed pyrolysis of acetylene containing a small amount of sulfur or phosphorus impurity, and also reported the morphologies, growth mechanism and some properties of the products since 1990. The present author studied the electric and dielectric properties of single and double helix composite carbon micro-coils and observed that electric and dielectric properties depend on the frequency and the thickness of the composite sheet (Nessa and Motojima 2006).

Though a lot of work have been conducted on CMC/polysilicone composite sheets, but how CMC addition show high sensitivity is still controversial.

In this paper, efforts were made to discuss the effects of different amount of CMC addition on the frequency dependence of dielectric properties.

MATERIALS AND METHODS

The chemical vapour deposition (CVD) process was used for the preparation of carbon micro-coils (Fig. 1). Catalyst (powder or thin films)-supported graphite plate was used as the substrate, and was set in the horizontal reaction tube (quartz, 1000 mm length and 100 mm i.d.). Acetylene was used as a carbon source and Ni or stainless steel (SUS 304) fine powder was used as a catalyst. The reaction tube was heated at 700 - 780°C for Ni catalyst and 740°C for SUS 304 catalyst from outside by electric heater. A source gas mixture of acetylene, hydrogen, hydrogen sulfide and nitrogen was vertically introduced into the reaction tube through the upper gas inlet and exhausted through the lower gas outlet. The gas flow rates of C_2H_2, H_2, H_2S and N_2 were fixed at 840, 3500, 150, 600 sccm for Ni catalyst and 300, 1000, 120 and 500 sccm for SUS 304 catalyst, respectively.

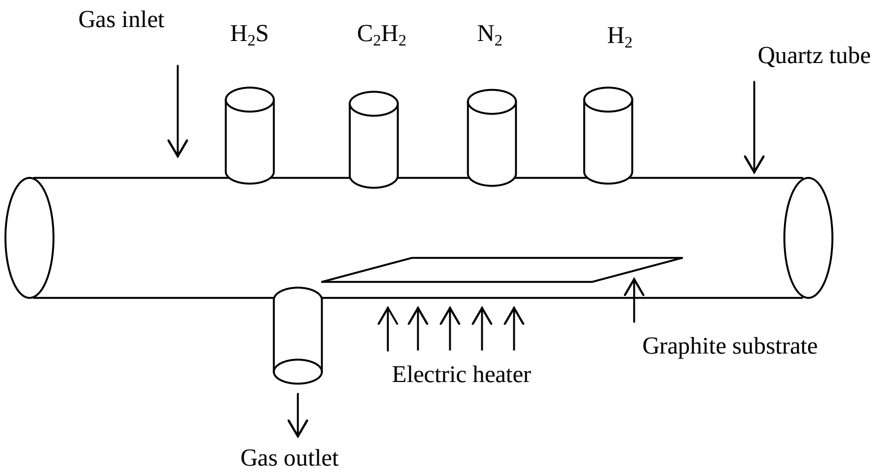

Fig. 1. Schemetic diagram of the chemical vapor deposition apparatus.

The pressure within the reaction tube was atmospheric. High-resolution FE-SEM (TOPCON, ABT-150F) was employed to examine their morphologies and microstructures. The CMC was pulverized and sieved to obtain the composite sample of a coil length below 90 µm.

To obtain a CMC/polysilicone composite sheet, the pulverized CMC was uniformly mixed with polysilicone resine (Shinetsu Chemicals, KE-103) by different wt% addition (0.1, 0.5, 1, 5 and 10) using a centrifugal conditioning mixer (Shinky, AR-100) and molded in Al mould of 10 × 10 × 0.1 mm^3. The CMC sheet sample with thickness of 0.1 mm was prepared.

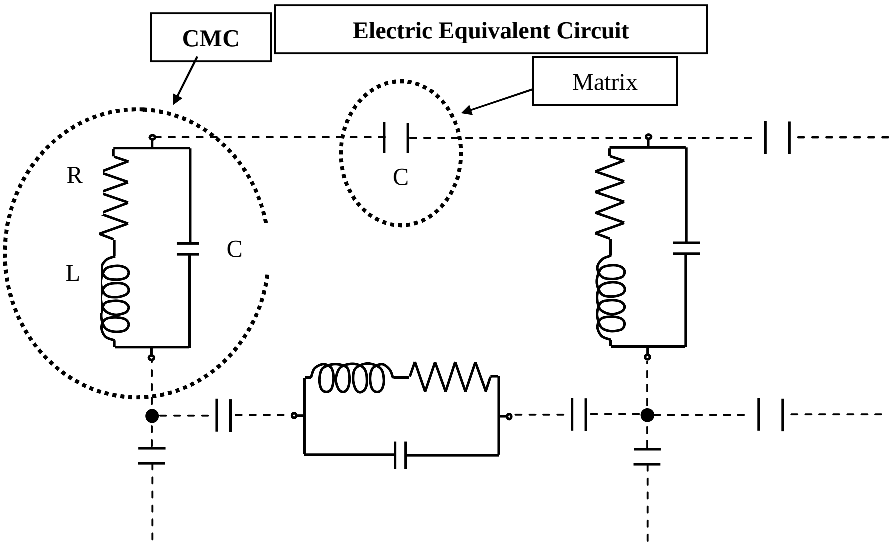

Fig. 2. Equivalent circuit used in the measurement.

The dielectric parameters (θ, tanδ, ε_r) of the CMC/polysilicone composite sheet was measured using an impedance analyzer (Agilent, LCR meter 4294A) at the frequency range from 40 Hz to 25 MHz where the applied voltage was 0.5 volt. The sheet was placed between two vertical parallel electrodes. The used equivalent circuit is shown in Fig. 2

RESULTS AND DISCUSSION

Under the standard reaction conditions, the obtained CMCs have generally an interesting 3D-helical/spiral structure with a constant coil diameter of 1 - 10 μm as shown in Fig. 3. Using Ni fine powder, the obtained CMC was generally very densely coiled double-helix forms as shown in Figs 3c,d. The double-helix CMC have no coil gap through the coil length, and very thin pyrolytic carbon layers of 2 - 20 nm thick was deposited on the surface of the fiber, from which coil is formed. Accordingly, the double helix coils have a thin tube-like pore with a diameter of 0.2 – 1 μm in the central part of the coils through the coil axis. On the other hand, using SUS 304 catalyst, the obtained CMC have generally single-helix forms with large coil gap of 0.2 – 5 μm and have higher elasticity than that of double helix CMC (Figs 3a,b).

The frequency dependence of dielectric properties under the effect of CMC addition with polysilicone composite for single and double-helix CMC sheet have been discussed. Fig. 4 shows the effect of additional amount of CMC with polysilicone on the frequency dependent of dielectric loss angle (θ). For both single and double helix, the CMC addition 0.1, 0.5, and 1 wt%, the value of lose angle (θ), do not show any measurable change with frequency. But for double-helix, the CMC addition 5 and 10 wt%, initially dielectric loss angle increases with frequency until 1 KHz range and decreases with frequency after 1 KHz, and around 1 MHz, the value of dielectric loss angles are attained to a constant value irrespective of the CMC addition with the polysilicone.

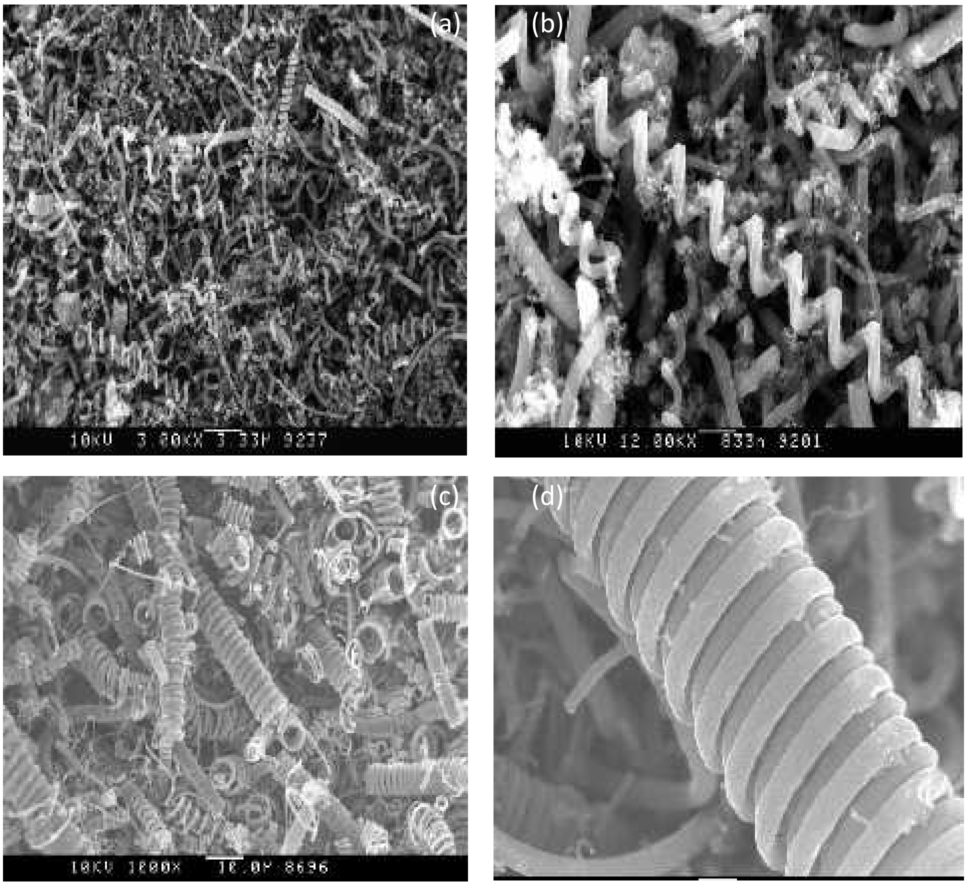

Fig. 3. SEM images of representative carbon micro-coils of (a, b) single-helix and (c, d) double-helix.

On the other hand, for single-helix, the CMC addition of 10 wt%, initially dielectric loss angle decreases with frequency at the frequency range 10^1-10^3Hz then increases at 10^3-10^4Hz and again decreases with frequency and attained constant value at MHz region. Higher the CMC addition dielectric loss angles are high and so on. Although strong noises are initially observed for both cases.

When an electric field acts on any matter a certain quantity of electric energy dissipates that transforms into heat energy. This phenomenon is commonly known as “the expense” or “loss” of power. The loss of power is directly proportional to the square of the applied electric voltage to the specimen. Under a given voltage, the dissipation of power in these dielectrics depends on voltage frequency (Saxena 1996).

A phasor diagram of currents and voltages in a capacitor energized by an alternating voltage is shown in fig.4c.

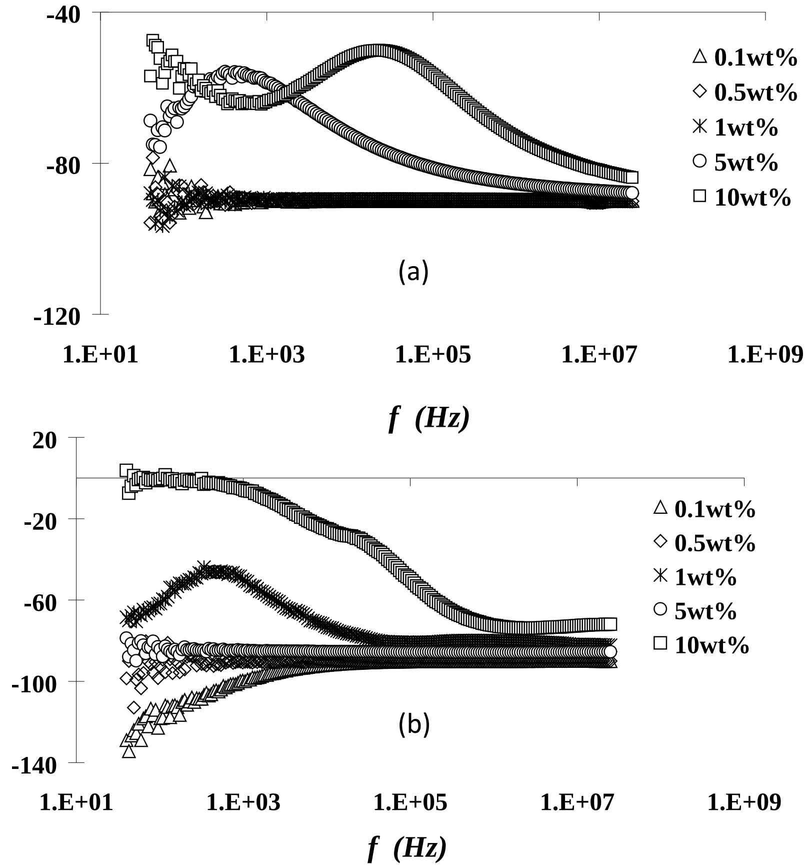

Fig. 4a,b. Variation of dielectric loss angle (θ) with frequency for the composite sheets of (a) single-helix CMC and (b) double-helix CMC. Additional amount of CMC: (Δ) 0.1 wt%; (◊) 0.5 wt%; (Ж) 1 wt%; (O) 5 wt%; (□) 10 wt%.

If the power were not dissipated at all in the dielectric of the capacitor, the phase of the current I through the capacitor would be ahead of voltage precisely by 90° and the current would be purely reactive. In actual fact the phase angle φ is slightly less than 90°. The total current I through the capacitor can be resolved into two components, active I_a and reactive I_r currents. Thus the phase angle describes a capacitor from the view point of losses in a dielectric. The dielectric loss angle $\theta = 90° - \varphi$.

The dielectric loss angle may be due to the induced electric dipole moment which is present only when the electric field is present. The induced dipole moment producing partial alignment with the electric field. Thermal agitation prevents the complete alignment of the dipole (Devid Halliday and Robert Resnick 1993). In higher additional amount of CMC, the number of induced dipole moment might be higher and the partial alignment is increased resulting the dielectric loss angle θ is larger compare to the lower additional amount of CMC.

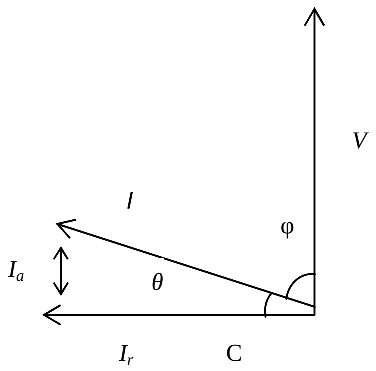

Fig. 4c. Simplified diagram of currents in a lossy dielectric.

The dielectric loss tangent (*tanδ*) is equal to the ratio between the active and reactive currents (Fig. 4c),

$$\tan\delta = I_a/I_r$$

Fig. 5 shows the dielectric loss tangent (*tanδ*) as a function of frequency in relation with the CMC addition in polisilicone composite for single and double helix carbon micro-coils. In single and double-helix CMC sheets, the *tanδ* decreased up to 10^6 Hz regions and then attained constant with increasing frequency. The lower addition of CMC (0.1 and 0.5 wt%), the value of *tanδ* randomly decreased up to 10^6 Hz for single helix but no results for double helix. It again appeared after 10^6 and suddenly rose in both cases at 10^7 Hz region, then sharply decreased. In CMC addition, the higher amount of CMC the higher was *tanδ* for both single and double helix composite sheet.

It was found that the dielectric constant was dominated by the capacitance of CMC. The relative dielectric constant can be expressed as

$$\varepsilon_r = C_c/C_v,$$

where, C_c is the capacitance of dielectric CMC sheets set between two parallel electrodes and C_v is the capacitance of the same thickness of silicone rubber without addition of CMC.

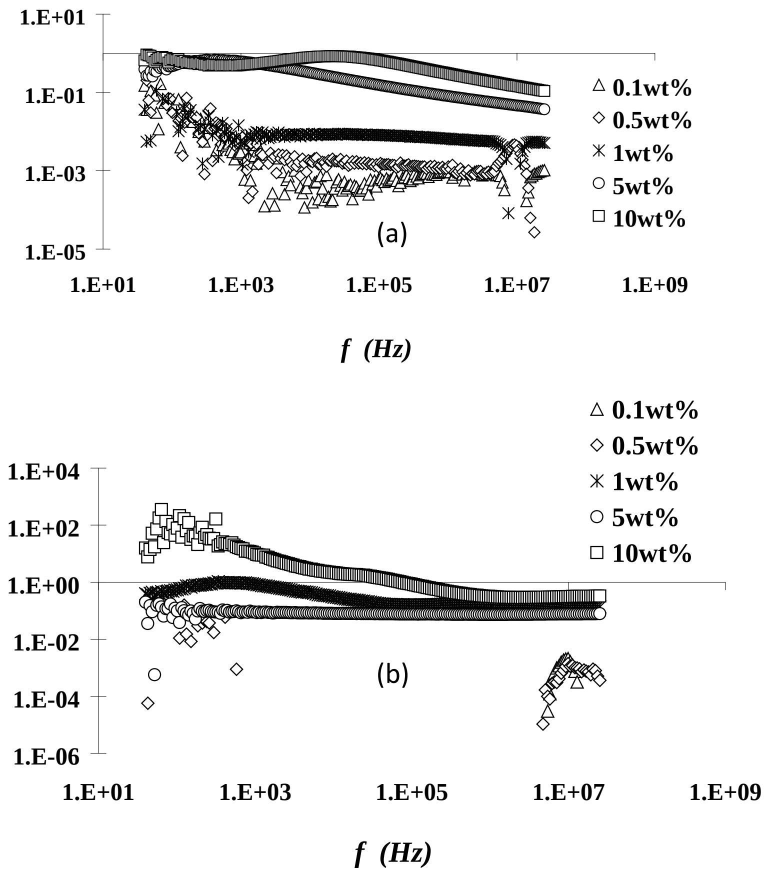

Fig. 5. Variation of dielectric loss tangent (*tanδ*) with frequency for the composite sheets of (a) single-helix CMC and (b) double-helix CMC. Description of symbols is the same as in Fig. 4.

Fig. 6 shows the effect of frequency on the relative dielectric constant (ε_r) in relation to the addition of CMC in the polysilicone composite sheet. In both single and double-helix CMC sheets, dielectric constant decreased with increasing frequency, although initially some noise was observed. Initially the decreasing rate for lower CMC addition was very slow up to 10^3 Hz and then attained constant. At lower additional amount of CMC, the ε_r was smaller, and higher additional amount of CMC the ε_r was larger in both cases.

CONCLUSION

The author studied the effects of different amount of CMC addition on the frequency dependence of dielectric properties of a single-and double-helix CMC. The observed characteristics of the dielectric properties under the additional amount of CMC for single- and double-helix were nearly same. The frequency dependence of dielectric parameters are strongly influenced by the additional amount of CMC in the composite sheet. The higher addition of CMC was better than lower addition for detecting high responsibility of dielectric parameters.

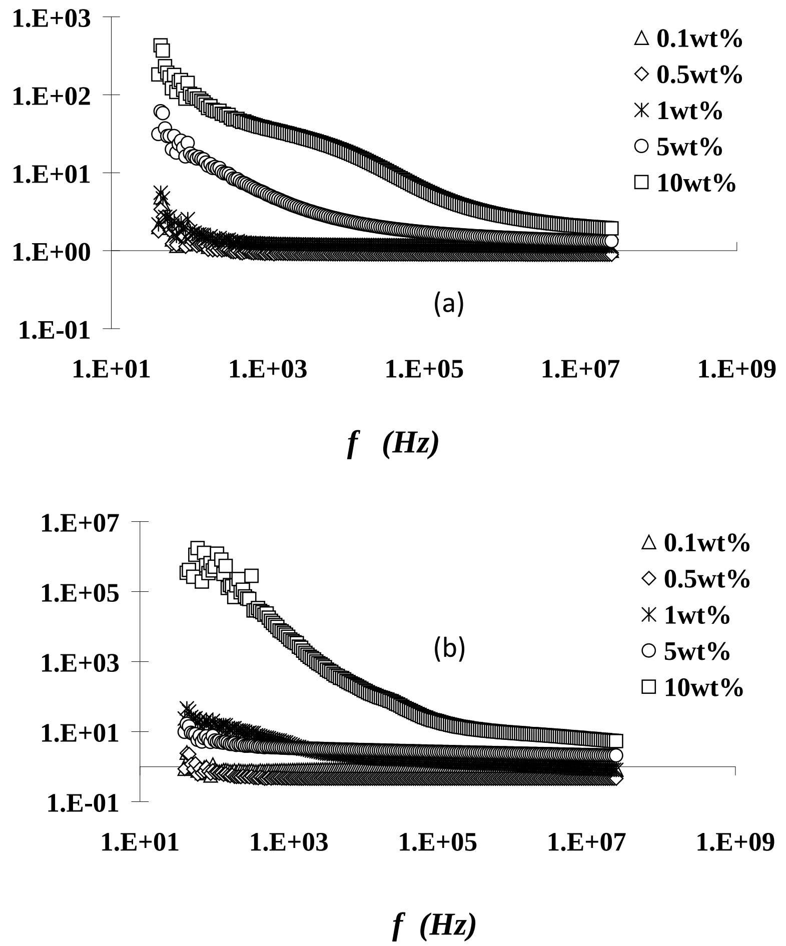

Fig. 6. Variation of relative dielectric constant (ε_r) with a frequency for the composite sheets of (a) single-helix CMC and (b) double-helix CMC. Description of symbols is the same as in Fig. 4.

ACKNOWLEDGEMENTS

The author is grateful to Professor S. Motojima, Department of Applied Chemistry, faculty of Engineering, Gifu University, Gifu, Japan for suggesting this research and valuable comments in preparing this manuscript. She is also thankful to the authority of Virtual System Laboratory (VSL) and Gifu University for funding her postdoctoral research.

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(*Received revised manuscript on 14 August, 2013*)