# Characterization of dielectric properties of the enamel filled with carbon nanotubes for the frequency range of 50 Hz - 5 MHz

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**Abstract:**

It has been observed that the addition of nano fillers to the enamel can greatly improve the dielectric properties of the enamel. Carbon nanotubes have been tested as nano filler. Chemical vapour deposition method was used to synthesize carbon nanotubes. Scanning electron microscope has been used to augment the particle size of carbon nanotubes. These carbon nanotubes were mixed with standard (Elmo Luft 1A-FD) enamel with help of ultrasonic vibrator. The basic dielectric properties such as dielectric loss tangent (tanδ), dielectric constant(ε), quality factor, phase angle, dielectric conductivity and dielectric power loss of the enamel filled with carbon nanotubes were analyzed and compared with the properties of the standard enamel. These different dielectric properties were found with the help of the Dielectric Spectroscopy. Dielectric constant, dielectric losses, dielectric conductivity and heat generated under AC fields were analyzed for the frequency range of 50 Hz to 5 MHz. At 50 Hz, the enamel filled with 1 wt% of carbon nanotubes has lower dissipation factor and dielectric losses. At 5 MHz, the enamel filled with 3 wt% of carbon nanotubes has lower dissipation factor and dielectric losses.

**Key words –** carbon nanotubes, dielectric constant, dielectric losses, dielectric conductivity, quality factor, dielectric loss tangent, dielectric power loss.

## 1. INTRODUCTION

In the last few years, a great deal of attention has been given to the applications of nano dielectrics in the field of electrical insulating materials. It has been reported that the use of nano composites in the matrix of polymeric materials can greatly improve the thermal, mechanical and electrical properties of polymeric nano composites [4].

In the last few years, a great deal of attention has been given to the application of nano dielectrics in the field of electrical materials. Nano dielectrics were a class of materials containing at least one phase at the nanometer scale. The basic understanding of electrical breakdown of materials and electrical surface flashover phenomena must be investigated before they can be commercially available [1]. The findings of such studies are essential for the development of nano - dielectric materials.

A driving force of the nano revolution was a continuous progress in the nano dielectrics towards increasing the level of stability, the reduction in the size and weight of the insulating materials [5]. The different dielectric properties such as dielectric loss tangent (tanδ), dielectric constant(ε), quality factor, phase angle, dielectric conductivity and dielectric power loss were important for an insulating material [3].

## 2. EXPERIMENTAL

### 2.1. Sample Preparation

The nanocomposites were prepared by radical initiator curing method. 80% of enamel and 20% of epoxy resin were taken. Diamino Diphenyl Methane (DDM) was used as curing agent. For 1g of resin, 0.27g of DDM was taken. The DDM was melted at 60° - 80°C for 10 minutes. The enamel, resin and melted DDM were mixed in a beaker. The mixture was poured into the die coated by a Teflon sheet. The die was heated at 120° C for 3 hours. Then, the die was taken away from the oven and it was cooled for 1 hour.

### 2.2. Synthesis of carbon nanotubes

The synthesis of carbon nanotubes consists of three stages: Preparation of Catalysts for carbon nanotubes, Chemical vapour deposition (CVD) process and Purification of Carbon Nanotubes.

Catalysts with different Mo: Fe: $Al_2O_3$ molar ratios were prepared by adding calculated amount of Ammonium hexa molybdate tetra hydrate and ferrous sulphate heptahydrate into a suspension of alumina powder in methanol [2]. The different concentrations of catalysts prepared were illustrated in the Table 1.

**Table 1 Different catalyst concentrations**

| S. No | Molar ratio of Mo:Fe:$Al_2O_3$ | Weight of Ammonium hexamolybdate grams | Weight of ferrous sulphate grams | weight of alumina grams | Volume of methanol (ml) |
|---|---|---|---|---|---|
| A | 0.16:1:16 | 2.0995 | 0.8062 | 9.09 | 25 |
| B | 0.32:1:16 | 4.199 | 0.8062 | 9.09 | 25 |
| C | 0.32:2:16 | 4.199 | 1.6124 | 9.09 | 25 |
| D | 0.48:2:16 | 6.2985 | 1.6124 | 9.09 | 50 |

CVD process was done in an experimental set up consisting of a horizontal reaction furnace, Quartz tube, PID controller, Flow meters, Control valves, Gas sources and Thermocouple.

The quartz tube was placed inside the horizontal reaction furnace. A proportional integral differential (PID) controller was provided in the furnace to control the temperature, time and heating rate of the reaction.

Three cylinders of pure Nitrogen ($N_2$), Acetylene ($C_2H_2$) and Argon (Ar) were connected to the quartz tube through control valve and flow meter. The rate of carbon and nitrogen entering the reaction tube was shown by the flow meter. Control valves were used to control the flow rate of gases passing through the furnace. Rubber hoses were used to connect the cylinders with flow meter and valves.

After synthesis, the raw products were treated with a solution of KOH (1M) at 80$^o$C in order to remove the alumina support and molybdenum. The resulting material was washed several times with distilled water and then treated with a solution of HCL (1M) in order to remove the remaining iron particles. It was then washed with distilled water.

Then the particle size of the powder was analyzed by using the SEM characterization techniques. From the results, the particle size was found to be tens of nanometer.

## 2.3. Calculation of relative permittivity, dielectric conductivity and dielectric losses

Dielectric spectroscopy was used to find the loss factor, quality factor and phase angle of the dielectric material. From these factors, dielectric constant, dielectric conductivity, dielectric loss and heat generated under AC fields were found.

Dielectric spectroscopy [LCR HITESTER 3532-50] was also known as Electrochemical Impedance Spectroscopy. The dielectric properties of a medium were measured as a function of frequency. It was based on the interaction of an external field with the electric dipole moment of the sample, often expressed by permittivity.

The set up of dielectric spectroscopy was shown in figure 1.

Figure 1 Dielectric spectroscopy [LCR HITESTER 3532-50]

The real and imaginary part of relative permittivity of the enamel was calculated from the following equations which were used to calculate the dielectric loss [5].

Loss factor
$$\tan\delta = \frac{\varepsilon_r''}{\varepsilon_r'} \quad (1)$$

Real part of relative permittivity
$$\varepsilon_r' = \left(\frac{t * C_p}{A * \varepsilon_0}\right) \quad (2)$$

Imaginary part of relative permittivity
$$\varepsilon_r'' = \left(\frac{t}{\omega * R_p A * \varepsilon_0}\right) \quad (3)$$

Dielectric conductivity $\sigma = \omega * \varepsilon''$ (4)

Dielectric losses $P = 2\pi * f * C * V^2 * \tan\delta$ W (5)

The heat generated under AC field $W_{ac} = E^2 * f * \varepsilon_r * \tan\delta / 1.8 \times 10^{12}$ W/cm$^3$ (6)

Where

f was the frequency in Hz,

$\delta$ was the loss angle of the dielectric material and

E was the applied electric field.

$C_p$ was the equivalent parallel capacitance [F]

$R_p$ was the equivalent parallel Resistance [Ω]

D was the dissipation factor (measured value)

$t_m$ was the average thickness [m]

A was the guarded electrode's surface area [m$^2$]

D was the guarded electrode's diameter[m]

$\varepsilon_o$ was the permittivity of free space =8.854 x 10$^{-12}$ [F/m]

V was the applied voltage [V]

tanδ was the dielectric loss tangent

# 3. RESULTS

## 3.1. SEM analysis of Carbon nanotubes

Figure 2 shows the SEM analyzed image results. These results show that particles were in the form of nano metric range. The sizes of the particles were in the range from 50 to 120 nm size.

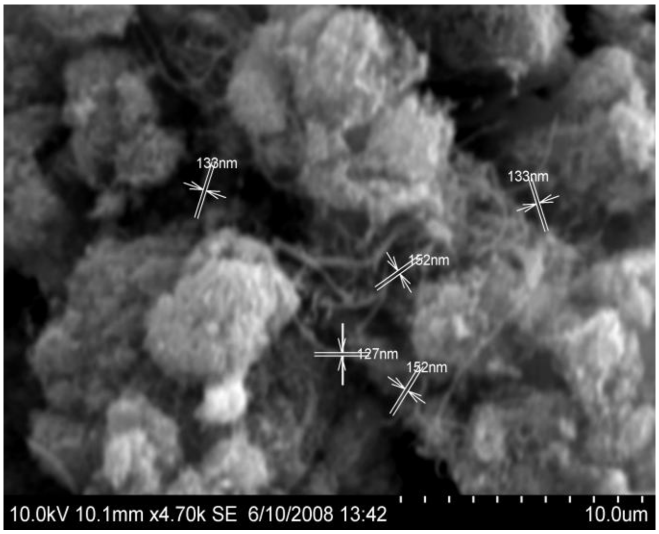


Figure 2 SEM analysis of Carbon nanotubes

## 3.2. Various dielectric properties of the carbon nanotubes filled enamel for the frequency range (50 Hz to 5 MHz) at 90$^o$ C

The different dielectric properties like dissipation factor, quality factor and phase angle were found with the help of the Dielectric Spectroscopy. Dielectric constant, dielectric losses, dielectric conductivity and heat generated under AC fields were analyzed for various frequencies. The loss factor and the quality factor were dependent upon the frequency. The values of various dielectric properties of the carbon nanotubes filled enamel for the frequency range (50 Hz to 5 MHz) at 90$^o$ C were listed in the tables 2 to 8.

**Table 2 Dissipation factor**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | 7.865 | 0.793 | 0.978 | 1.083 |
| 100 Hz | 5.496 | 0.594 | 0.59 | 0.663 |
| 1 kHz | 1.537 | 0.274 | 0.321 | 0.499 |
| 10 kHz | 0.508 | 0.129 | 0.388 | 0.229 |
| 100 kHz | 0.197 | 0.072 | 0.137 | 0.0608 |
| 1 MHz | 0.121 | 0.063 | 0.0877 | 0.0922 |
| 5 MHz | 0.0627 | 0.093 | 0.0663 | 0.1282 |

At 50 Hz, the enamel filled with 1 wt% of carbon nanotubes has lower dissipation factor. At 5 MHz, the enamel filled with 3 wt% of carbon nanotubes has lower dissipation factor.

**Table 3 Quality factor**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | 0.13 | 1.26 | 1.02 | 0.92 |
| 100 Hz | 0.18 | 1.68 | 1.69 | 1.51 |
| 1 kHz | 0.65 | 3.65 | 3.11 | 2 |
| 10 kHz | 1.97 | 7.72 | 2.58 | 4.35 |
| 100 kHz | 5.06 | 13.97 | 7.3 | 16.43 |
| 1 MHz | 8.26 | 15.8 | 11.39 | 10.84 |
| 5 MHz | 15.95 | 10.65 | 15.07 | 7.8 |

The enamel filled with 1 wt% of carbon nanotubes has higher quality factor at 50 Hz. At 5 MHz, the enamel filled with 3 wt% of carbon nanotubes has higher quality factor.

**Table 4 Dielectric constant**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | 220.35 | 57.37 | 104.49 | 103.37 |
| 100 Hz | 161.47 | 51.85 | 97.14 | 95.41 |
| 1 kHz | 76.054 | 35.66 | 72.16 | 58.3 |
| 10 kHz | 44.51 | 28.66 | 38.29 | 28.36 |
| 100 kHz | 33.43 | 25.81 | 30.03 | 29.23 |
| 1 MHz | 28.96 | 24.5 | 26.95 | 28.23 |
| 5 MHz | 26.39 | 20.01 | 23.33 | 26.16 |

The value of dielectric constant was high for the enamel at 50 Hz. The lowest value of dielectric constant was observed for the enamel filled with 1 wt% of carbon nanotubes at 5 MHz.

**Table 5 Dielectric losses (μW)**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | 6.17 | 0.162 | 0.364 | 0.4 |
| 100 Hz | 6.33 | 0.22 | 0.41 | 0.45 |
| 1 kHz | 8.35 | 0.697 | 1.653 | 2.1 |
| 10 kHz | 16.11 | 2.632 | 10.6 | 4.9 |
| 100 kHz | 47.01 | 13.22 | 27.53 | 12.2 |
| 1 MHz | 250.76 | 109.79 | 176.24 | 172.26 |
| 5 MHz | 590.63 | 664.35 | 551.68 | 1197.58 |

The enamel filled with 1 wt% of carbon nanotubes has lowest value of dielectric losses at 50 Hz. At 5 MHz, the enamel filled with 3 wt% of carbon nanotubes has higher value of dielectric losses.

**Table 6 Dielectric conductivity (S)**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | $4.832 \times 10^{-6}$ | $1.567 \times 10^{-12}$ | $9.261 \times 10^{-14}$ | $2.28 \times 10^{-13}$ |
| 100 Hz | $4.95 \times 10^{-9}$ | $4.35 \times 10^{-12}$ | $2.811 \times 10^{-13}$ | $5.80 \times 10^{-3}$ |
| 1 kHz | $6.52 \times 10^{-6}$ | $1.822 \times 10^{-10}$ | $3.63 \times 10^{-11}$ | $1.1 \times 10^{-10}$ |
| 10 kHz | $1.25 \times 10^{-8}$ | $1.338 \times 10^{-8}$ | $8.924 \times 10^{-9}$ | $1.56 \times 10^{-8}$ |
| 100 kHz | $3.68 \times 10^{-8}$ | $1.944 \times 10^{-9}$ | $1.22 \times 10^{-6}$ | $1.17 \times 10^{-6}$ |
| 1 MHz | $1.96 \times 10^{-4}$ | $8.66 \times 10^{-10}$ | $5.86 \times 10^{-4}$ | $9.81 \times 10^{-4}$ |
| 5 MHz | $4.60 \times 10^{-4}$ | $6.2 \times 10^{-11}$ | $57.91 \times 10^{-3}$ | $10.32 \times 10^{-3}$ |

**Table 7 Phase angle**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | -7.25 | -51.55 | -45.62 | -42.71 |
| 100 Hz | -10.31 | -59.29 | -59.44 | -56.46 |
| 1 kHz | -33.04 | -74.66 | -72.18 | -63.45 |
| 10 kHz | -63.06 | -82.62 | -68.78 | -77.05 |
| 100 kHz | -78.82 | -85.91 | -82.2 | -86.52 |
| 1 MHz | -83.1 | -86.38 | -84.98 | -84.73 |
| 5 MHz | -86.41 | -95.36 | -93.8 | -82.69 |

**Table 8 Heat generated ($W/cm^3$)**

| Frequency | Enamel | Enamel filled with 1 wt% of carbon nanotubes | Enamel filled with 3 wt% of carbon nanotubes | Enamel filled with 5 wt% of carbon nanotubes |
|---|---|---|---|---|
| 50 Hz | 32.04 | 0.465 | 1.56 | 1.74 |
| 100 Hz | 32.34 | 0.632 | 1.77 | 2.04 |
| 1 kHz | 42.61 | 2.2 | 7.13 | 8.9 |
| 10 kHz | 82 | 7.64 | 46.24 | 25.77 |
| 100 kHz | 240.3 | 36.9 | 127.76 | 53.38 |
| 1 MHz | 1277.59 | 304.01 | 739.21 | 773.15 |
| 5 MHz | 3027.76 | 1824.05 | 2448.81 | 5185.01 |

The enamel filled with 5 wt% of carbon nanotubes was generating the highest amount of heated at 5 MHz when compared to other samples.

## 4. CONCLUSION

SEM analysis showed that the prepared carbon particles were appearing in the form of nano metric size. The various dielectric properties were analyzed by dielectric spectroscopy instrument at 90°C for the frequency range of 50 – 5 MHz. These results show that the additions of few weight percentages of carbon nanotubes would improve the dielectric behaviour of the enamel.

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