Dielectric Loss Model of Humidity Sensor Based on Multi-Wall Carbon Nanotubes

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Abstract: To study the dielectric loss of carbon nanotubes (CNTs) humidity sensor, a humidity sensor based on multi-wall CNTs was fabricated. The frequency properties were analyzed by dielectric loss theories, and the dielectric loss model was built by universal law. Fitting the model with experimental data, the R-square is 0.994, which verifies that the universal law can be used to describe the dielectric loss of the sensor. As the change in sensor resistance caused by test frequency can affect sensor linearity, the linearity of the sensor under different test frequencies was also analyzed. It is found that when the frequency is 10 kHz, the sensor presents the best linearity (1.52%), and the corresponding sensitivity is −7.83 Ω/%RH.

Keywords: carbon nanotubes (CNTs); humidity sensor; frequency properties; dielectric loss

多壁碳纳米管湿度传感器介电损耗模型

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摘 要:为揭示碳纳米管湿度传感器的介电损耗,制作了一种电阻型碳纳米管湿度传感器. 使用介电损耗相关理论对其频率特性进行分析,并利用普适方程建立了传感器介电损耗模型.对实验数据与所建立模型进行拟合,得到拟合决定系数为0.994,表明可以使用普适方程对传感器介电损耗进行描述. 由于测试频率引起的传感器电阻变化会改变传感器线性度,分析了不同测试频率下传感器的线性度,发现当测试频率为10 kHz时传感器的线性度最佳(1.52%),此时传感器灵敏度为−7.83 Ω/%RH.

关键词:碳纳米管(CNTs); 湿度传感器; 频率特性; 介电损耗
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Recently, humidity sensors were widely applied in industry, agriculture, national defense, meteorology, and so forth[1]. Different sensing materials, including macromolecule compounds[2], semiconducting materials[3], porous silicon[4], carbon nitride film[5] and porous ceramics[6], were explored to detect the humidity.

Since the discovery of carbon nanotubes (CNTs) by Iijima in 1991[7], CNTs have been expected as a novel sensing material, due to their high surface area to volume ratio and good electric properties. Over the last decade, researchers have been exploring the potential of CNTs in humidity sensing applications[8]. Nevertheless, little work focuses on the dielectric loss of CNTs humidity sensor, despite its essential effect on the per-

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formance of the sensor.

In this paper, a resistance type of humidity sensor based on multi-wall carbon nanotubes (MWCNTs) was presented. The frequency properties of sensor resistance were researched, and the results indicate that sensor resistance largely depends on test frequency. The mechanism of the frequency properties were explained using dielectric loss and the dielectric loss model was built. Moreover, the linearity and sensibility of the sensor to relative humidity (RH) were studied.

1 Experiment

1.1 Structure and fabrication of MWCNTs humidity sensor

Fig.1 shows the sensor structure, which consists of a pair of Al interdigital electrodes and an MWCNTs-SiO_2 film coated on the Al electrodes.

Before the fabrication of the sensor, the MWCNTs (purity > 95% in mass, diameter 10—20 nm, length about 1—2 μm) were functionalized with H_2SO_4 and HNO_3 mixture (3:1 in volume) at 100 ℃ for 2 h. Through the acid treatment, MWCNTs can be purified and functionalized with carboxylic (—COOH) groups, which can make the MWCNTs hydrophilic.

Then, the Al layer was evaporated on the glass substrate and made into interdigital electrodes pattern by photolithography. Next, the layer of MWCNTs-SiO_2 mixture was coated on the Al electrodes by silk screen printing. The thickness of the MWCNTs-SiO_2 layer was about 15 μm. At last, the sensor was heated at 380 ℃ in vacuum.

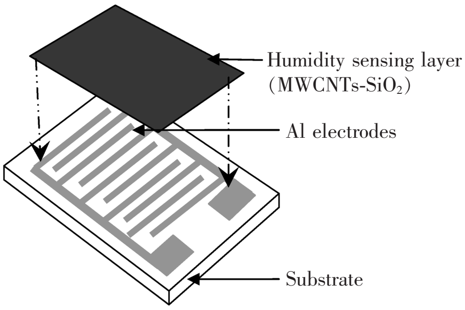

Fig. 1 Schematic drawing of the MWCNTs humidity sensor

1.2 Test instrument

A series of humidity atmospheres with the RH of 11%, 33%, 43%, 52%, 67%, 75.5%, 86% and 97% were achieved respectively by the saturated solutions of LiCl, $MgCl_2$, K_2CO_3, $Mg(NO_3)_2$, $CuCl_2$, NaCl, KCl and K_2SO_4 in airtight glass vessels at room temperature[9]. An automatic RCL meter-PM6306 (shown in Fig. 2) with the test frequencies from 50 Hz to 1 MHz was adopted for the measurement of sensor resistance.

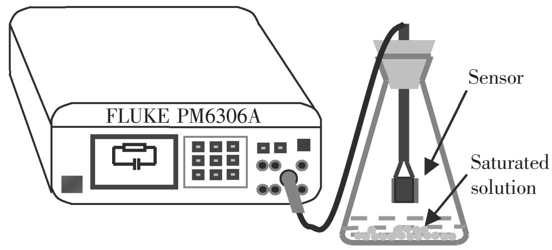

Fig. 2 Schematic diagram of the testing system

2 Frequency properties

According to Ref. [10], the circuit model of the sensor consists of a body resistance R in parallel with a body capacitor C, as shown in Fig.3, where G is body conductance and $1/G$ represents the body resistance R.

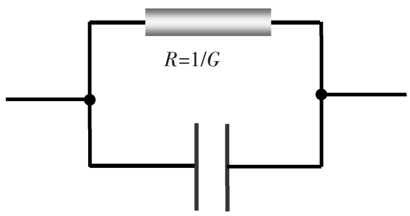

Fig. 3 Circuit model of the sensor

The complex permittivity of the sensing material can be described by

$$\varepsilon=\varepsilon'-\mathrm{i}\varepsilon'' \tag{1}$$

where ε is complex permittivity, and the real part (ε') and imaginary part (ε'') correspond to dielectric constant and the dielectric loss, respectively.

Under AC testing, current density through the medium in MWCNTs film is described by

$$j(\varepsilon)=\varepsilon\times\frac{\partial E}{\partial t}=(\mathrm{i}\omega\varepsilon'+\omega\varepsilon'')\times E \tag{2}$$

where ω is angular frequency, and E is electric field.

Eq. (2) demonstrates that the current density through the medium is proportional to ε''. Thus dielectric loss can produce a frequency-dependent conductance $G(\omega)$ in parallel with the body conductance $G(0)$, as shown in Fig.4.

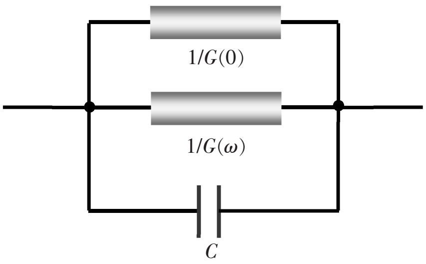

Fig. 4　Circuit model of the sensor considering frequency-dependent resistance

The sensing film was composed of SiO_2 (insulative) and MWCNTs (conductive or semiconductive), thus the polarization of the film could be described by space charge polarization, and the dielectric loss of such polarization could be represented by universal law ($\varepsilon'' \propto \omega^{n-1}$), with $n<1$ [11]. The conductance caused by the dielectric loss of sensing film is

$$G(\omega)=K\omega\varepsilon''=K\omega^n \quad (3)$$

where K is a constant.

When the RH is very low, the conductance of the sensor, G, is mainly composed of $G(0)$ and $G(\omega)$, i.e.,

$$G=G(0)+G(\omega)=G(0)+K\omega^n \quad (4)$$

In order to verify Eq. (4), sensor conductance was measured at the frequencies between 0.1 kHz and 50 kHz in 11%RH circumstance. The experimental data were fitted with Eq.(4). The conductance-frequency characteristics indicate that sensor conductance depends on test frequency, as shown in Fig.5.

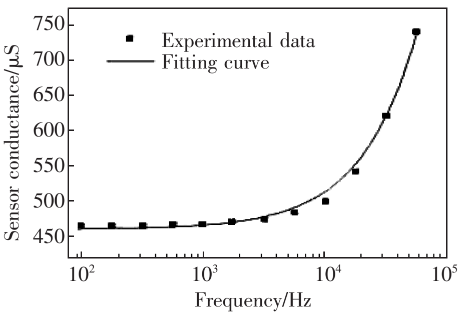

Fig. 5　Fitting curve of sensor conductance by the dielectric loss model

By fitting Eq.(4) with the experimental data, the parameters $G(0)$, K and n were obtained, as shown in Tab.1. R^2 is 0.994, which verifies that the universal law can be used to describe the dielectric loss of the sensor.

Tab. 1 Values of parameters in Eq. (4)

$G(0)$/μS	K	n	R^2
460	0.007	0.951	0.994

3　Linearity and sensitivity

As the change in sensor resistance caused by test frequency can affect the sensor linearity, the variation of sensor resistance with RH at different test frequencies was tested, as shown in Fig.6.

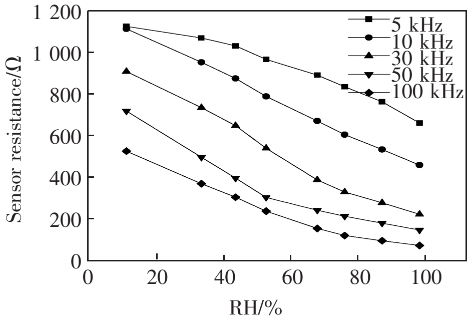

Fig. 6　Sensor resistance *vs* RH at different testing frequencies

It can be seen from Fig.6 that when the frequency is 10 kHz, the sensor presents the best linearity. The sensor resistance changes from 1 115 Ω to 462 Ω and the curve of resistance as a function of RH is quite linear(R=1 202−7.83%RH), as shown in Fig.7, the R^2 of the linear fitting process is 0.996. Thus the sensitivity to RH of the sensor is −7.83 Ω/%RH.

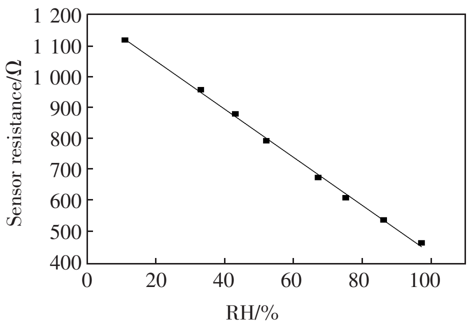

Fig. 7　Sensor resistance *vs* RH at the frequency of 10 kHz

The sensor linearity is defined as

$$\delta=\frac{\Delta R_{max}}{\Delta R_{total}}\times 100\% \quad (5)$$

where ΔR_{max} is the maximum between the experimental data and the fitting curve, and R_{total} is the full scale of sensor resistance. According to the results in Fig.7, sensor linearity to RH is about 1.52%.

4 Conclusion

A humidity sensor based on MWCNTs was fabricated, and the frequency properties of the sensor were studied. Experiment shows that sensor conductance changes with test frequency, which can be explained by the theory of dielectric loss. The dielectric loss of sensing film is represented by universal law, and the model of sensor dielectric loss was verified by experimental results.

The sensor linearity at different test frequencies was studied by experiment, and the results indicate that when the frequency is 10 kHz, the sensor presents the best linearity (1.52%). The sensitivity of the sensor is −7.83 Ω/%RH with the test frequency of 10 kHz.

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