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Characterizing Microwave Power in a MPCVD System using Gaussian Mixture Modeling

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Abstract: Microwave Plasma Chemical Vapor Deposition (MPCVD) can be used to grow various kinds of diamond films and carbon nanotubes at various temperatures. Issues concerning plasma modeling and control play an important role in MPCVD systems. One crucial factor in controlling the plasma shape and position is the tunable reflected microwave power of the MPCVD system. However, modeling the tunable reflected power is highly complex and remains a poorly developed. In this paper, the microwave power distribution corresponding to the adjustable electromagnetic field is modeled by 2-D Gaussian Mixture Modeling (GMM). The simulations using the model show that microwave power data can be simplified to a linear combination of some Gaussian functions, allowing predictable control for tuning manufacturing parameters and plasma sharp in real-time. The experimental results show that each E-H tuner position can fabricate the Multiwall Carbon Nanotubes (MWCNTs) films with high reproducibility after GMM modeling.

Keywords: MWCNTs, Gaussian mixture model, MPCVD, E-H tuner, ID/IG ratio

1 Introduction

Microwave Plasma Chemical Vapor Deposition (MPCVD) techniques have been widely studied and applied in several fields, especially to growing diamond films and carbon nano-tubes (CNTs) [1,2,3]. In MPCVD methods, many researches have shown that plasma modeling is increasingly needed to optimize manufacturing processing and control. However, dynamic behaviors of plasma are highly stochastic and complex. Various methodologies have been developed for modeling plasma [4,5,6] and trying to formulate useful mathematical equations for plasma dynamics. But, these models are highly nonlinear and sophisticated, and need a proper choice of parameters.

In general, modeling of plasma can be divided into two main approaches: analytical models and statistical models. Analytical models, [5,6] try to derive self-consistent solutions to physical equations which involve momentum, continuity, and energy conservation. However, precisely modeling plasma variables is difficult due to their high complexity. In statistical models, many recent studies have modeled plasma by using neural networks [7,8]. Neural network approaches have a better ability to precisely predict plasma dynamics than others, but have encountered problems in training.

In this paper, we focus on the problem of modeling the power of microwave plasma and I_D/I_G ratio distribution. The microwave power of MPCVD systems is presented in Section II. The quality indexes of MWCNTs are described in Section III. A new modeling method is proposed for optimizing the power of microwave plasma. Then, the performance of the new method is assessed by using model-based clustering analysis. First, the approach for modeling the power of microwave plasma is proposed in Section IV. Section V presents the experimental results. Finally, section VI presents some conclusions and suggestions for future efforts.

2 The Microwave Power of MPCVD systems

The MPCVD system used in this research is shown in Fig. 1. The E-H tuner shown in Fig. 2 is the tuning element. Tuning the microwave power is required because plasma impedance is influenced by various parameters of

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the plasma source and the plasma properties. Generally, it can be said that changes in process conditions require tuning. The E-H tuner includes three adjustable parameters, the x, y, and z axis positions. Tuning the x and y positions adjust the amplitude of the microwaves in the x and y directions. Tuning z adjusts the position of the standing wave. To obtain a high density plasma the microwave power needs to transmit maximum power to the plasma. The reflected power can be thought of as the characteristic of full power transmission of microwave power in MPCVD systems. Fig. 3 shows plasma image frames with different E-H tuner settings. A plasma density can be obtained by optimally tuning the position of the E-H tuner. In this paper, we focus on the modeling of microwave power in MPCVD systems and providing optimal positioning of the E-H tuner for obtaining high density plasma.

Fig. 1: The MPCVD System.

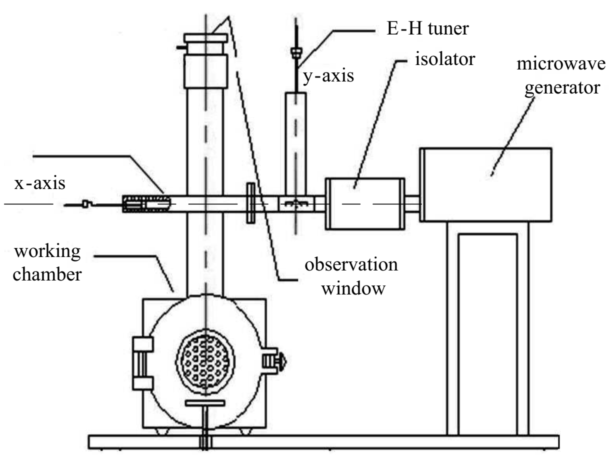


Fig. 2: E-H tuner microwave power.

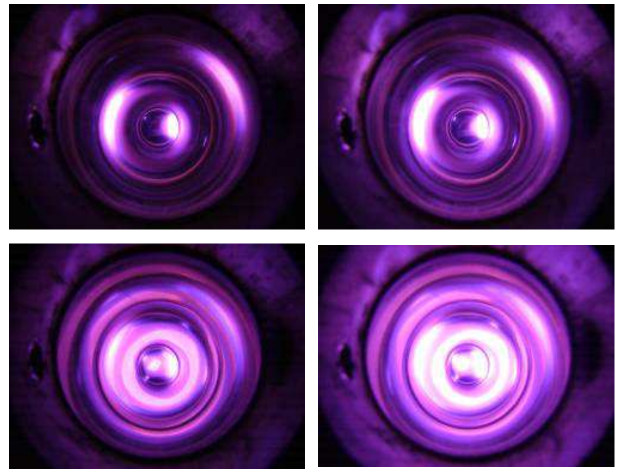


Fig. 3: Plasma image frames with different E-H tuner settings.

3 The Quality Indexes of MWCNTs

3.1 The aspect ratio

One of the novel properties of carbon nanotubes is that they have a high aspect ratio (ratio of length to diameter). Because of this high aspect ratio, they can be used in many applications, such as field emission displays, microwave power amplifiers, hydrogen uptake materials, nano-size probes, etc. The length and diameter can be inspected by scanning electron microscopy (SEM). One image of MWCNTs produced by our MPCVD system is shown in Fig. 4. The length and diameter of the MWCNTs are 19.61μm and 46.67nm, respectively. Therefore, its aspect ratio is about 420.

3.2 ID/IG band intensity ratio

Raman spectroscopy is often used in determining properties of carbon materials. Raman scattering by carbon materials has been analyzed in many studies. Some authors have used principal component analysis to discriminate the Raman spectra of functionalized MWCNTs, and some have discussed methods for determining Raman spectra of diamond films.

There are two important spectral bands in MWCNTs, at $\sim$1575 cm^{-1} (G line) and at $\sim$1338 cm^{-1} (D line). These represent sp^2 and sp^3 structures and have been reported by some groups. The G band corresponds to the E_{2g} modes, which represent the movement in opposite directions of two neighboring carbon atoms in a graphite sheet. The D band is induced by its dispersive disorder presents in the MWCNTs. It is known that the I_D/I_G band intensity ratio increases when covalent bonds are formed due to the formation of sp^3 hybridized carbon defect sites. Some authors not only discussed the intensity of I_D and I_G bands but also the effects of full width at half maxima (FWHM) of I_D and I_G band. They mentioned that larger peak intensity and smaller FWHM are consistent with the higher contents.

An RENISHAW inVia Raman spectrometer for which the excitation source was 514.5nm and the incident power at the sample was 25mW was used to obtain the Raman spectra in our experiment. One Raman spectrum of our MWCNT shows two peaks in the G and D bands. Moreover, we used the I_D/I_G band intensity ratio to define the quality index of MWCNTs.

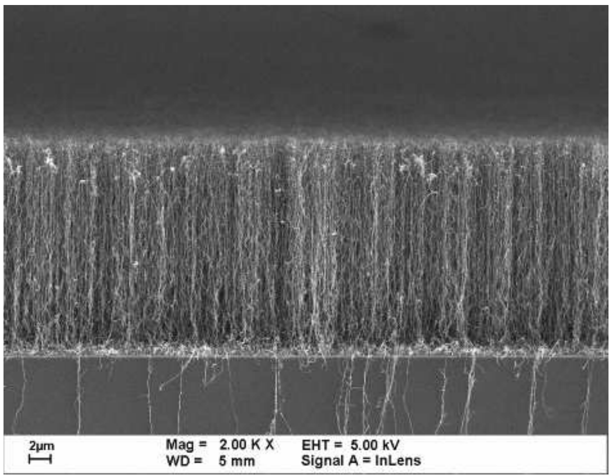


Fig. 4: SEM picture of growing MWCNTs.

4 Modeling Approach

4.1 Model-based clustering

Cluster analysis is a popular and useful technique in pattern recognition for unlabeled data. The goal is to separate the data into groups which have similar characteristics. These groups can be furthered studied and analyzed from the homogenous characteristics in each group. Several clustering techniques from time series analysis have been proposed in the literature [9,10,11]. The five best known approaches for clustering techniques [9] are partitioning methods, hierarchical methods, density-based methods, grid-based methods and model-based methods. There has also been extensive work on classified clustering methods [10] for various time series data. The three major approaches are raw-data-based methods, feature-based methods and model-based methods.

In our experiments, we observed the reflected power data from our MPCVD system. Fig. 5 shows the transmission of microwave power in the MPCVD system as a function of tuning E-H tuner. The x and y axes are the positions of the E-H tuner and z axis is the intensity of microwave power. Fig. 5 shows four peaks (high intensity) which we propose to model using a mixed Gaussian distribution. We will assume a mixed Gaussian model for each cluster and find the best fit for the microwave power data.

4.2 Gaussian Mixture Modeling

In this paper, the GMM is used to approximate the distribution of D/IG band intensity ratio and microwave power. Here, based on the central limit theorem, the overall uncertainty caused by the various factors over of the ID/IG band intensity ratio and microwave power can be well-modeled by one Gaussian distribution. The Gaussian mixture modeling [12,13] is expressed by

$$p(x|\theta) = \sum_{i=1}^{M} w_i p_i(x|\theta_i) \tag{1}$$

$$p_i(x|\theta_i) = \frac{1}{(2\pi)^{n/2}|\Sigma_i|^{1/2}} \exp\left[-\frac{1}{2}(x-\mu_i)^T \Sigma_i^{-1} (x-\mu_i)\right]$$

where x is the vector of random variables, M is the number of mixtures, parameter set $\theta = \{w_i, u_i, \Sigma_i\}$, $i = 1,\ldots,M$. $p_i(x|\theta_i)$ is a normal distribution with mean u_i and variance Σ_i, and w_i is mixture weighting such that

$$\sum_{i=1}^{M} w_i = 1, \;\; w_i \geq 0. \tag{2}$$

The goal is to find the optimal estimate for θ by using maximum likelihood estimation. The performance of GMM depends on haveing sufficient training data. We assume a training set $x = (x_1,\ldots,x_n)$ of n independent and identically distributed samples of random variable x. The maximum likelihood function is

$$\hat{\theta} = \arg\max_{\theta} L(\theta), \quad L(\theta) = \prod_{j=1}^{n} p(x_j)\,. \tag{3}$$

However, the parameters vector θ in GMM can not be estimated because the training data involves hidden parameters. Expectation-Maximization [13] is preferred method for finding the solution of maximum likelihood estimation when hidden parameters are present. Let X be the observed sample data set from some distribution and Y be the hidden variables. At each EM step the algorithm computes the expected value with Y as the random vector quantity

$$Q(\theta|\theta^t) = E_Y[\log p(X,Y|\theta)|X,\theta^t) \tag{4}$$

The E-M algorithm iterates to improve θ

$$w_i^{t+1} = \frac{1}{n}\sum_{j=1}^{n} p(i|x_j),$$
$$\mu_i^{t+1} = \frac{\sum_{j=1}^{n} p(i|x_j)x_j}{\sum_{j=1}^{n} p(i|x_j)}$$
$$\Sigma_i^{t+1} = \frac{\sum_{j=1}^{n} p(i|x_j)(x_j-\mu_i^{t+1})(x_j-\mu_i^{t+1})^T}{\sum_{j=1}^{n} p(i|x_j)} \quad (5)$$

until the log-likelihood increases by less than a threshold from one iteration to the next.

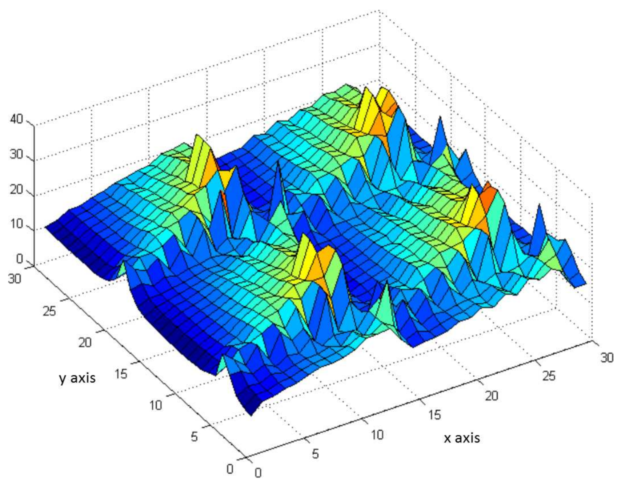


Fig. 5: The transmission of microwave power in the MPCVD system as a function of tuning E-H tuner.

5 Experimental Results

The E-H tuner in the MPVCD system uses the usual X, Y and Z axes. Axis Z represents a stationary wave that produces plasma at the center of the quartz tube when the Z-tuner is set to 5 cm. While the Z-tuner remains fixed, both the X and Y axes need to be changed to facilitate a series of experiments. This study utilizes a reflective power lower than 50% to adjust the E-H tuner position and analyze the micro structure of the MWCNTs which are fabricated at each corresponding position. The fixed parameters used in this study are shown in Table 1. In addition, the distribution of the plasma in the chamber affects the growing MWCNTs. However, the distribution of the plasma in the chamber is not easy to calculate from the equation because of complicated conditions, such as the shape of the chamber, the pressure, and the temperature [16]. Therefore, we set up the reflected power sensor to measure the reflected power near the substrate and the CCD camera in the observation window to observe the image of the plasma in this chamber.

Table 1: Fixed parameters of MWCNTs fabrication

Gas Flow Rate($[N_2]$:$[H_2]$:$[CH_4]$)	10 sccm : 40 sccm : 20 sccm
Microwave Power source	1000 W
Substrate Temperature	1200°C
Chamber Pressure	35 torr
Working distance	10 mm
Deposition Time	40 min
Preprocess	Sol-Gel
Catalyst	Fe_3O_4

5.1 ID/IG band intensity ratio and Microwave Reflected power

Data for this study were collected from the microwave tuning unit in the MPCVD system. The applied microwave power ranged from 500 to 2000 watts. The gaseous pressure in the reaction chamber was 35 torr. The step motion of the E-H tuner in the x and y directions was 6 mm. The total number of steps in x and y directions were 30 steps. Power was measured over 30 by 30 steps. The reflected power distribution corresponding to x-y positions is shown in Fig. 4.

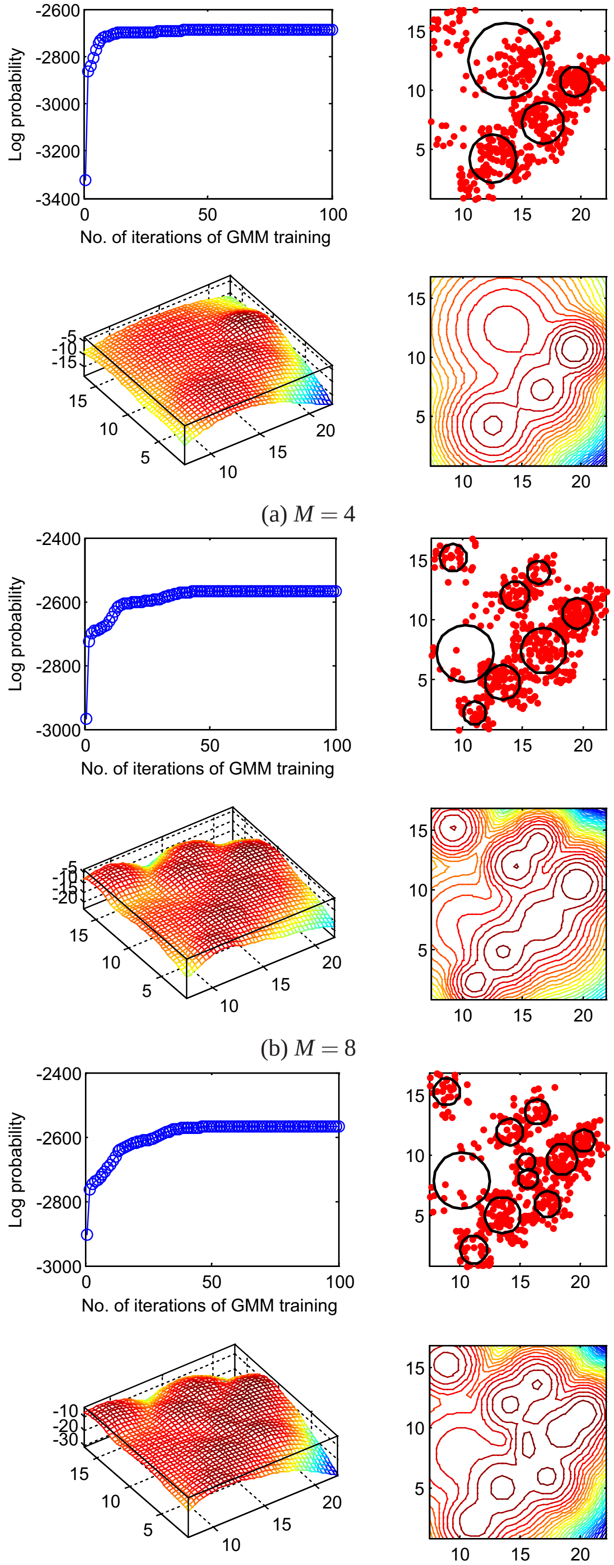
5.2 2-D pattern analysis

The model of microwave power was obtained using the GMM approach described above. Before training GMM, the 3-D microwave power distribution is resampled into 2-D histogram [14]. Fig. 6 shows the results of training the microwave power with GMM. The upper left plot of Fig. 6a presents the log probability of GMM training with 50 times iteration and the upper right plot shows the scatter plot of microwave power. The lower left plot of Fig. 6a presents the result of GMM training and the lower right shows its contour plot. For accurate modeling, we choice different M for the GMM and the result for $M = 4$, 8, and 12 are shown in Fig. 6. To verify the performance of the GMM, the root mean square errors (RMSE) are used as the performance index

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left|\frac{Y_i - Y_i^*}{Y_i}\right|^2} \quad (6)$$

where Y_i are the observation values and Y_i^* the modeling values. The best value M can be obtained by comparing the RMSE for different M.

The RMS errors for various values of M are plotted in Fig. 7. To model the I_D/I_G ratio distribution at each position of the E-H tuner, we also adopt the GMM. The result for $M = 18$ is shown in Fig. 8. The RMS errors for various values of M are shown in Fig. 9 and indicate that the RSME asymptotically converges to about 10% when $M = 18$. The microwave power and I_D/I_G ratio were successfully modeled with a high reproducibility.



(a) $M = 4$

(b) $M = 8$

(c) $M = 12$

Fig. 6: Results of training the microwave power with GMM for $M = 4$, 8, and 12.

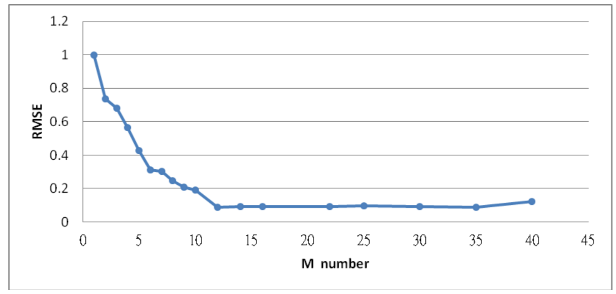


Fig. 7: RMSE for different M.

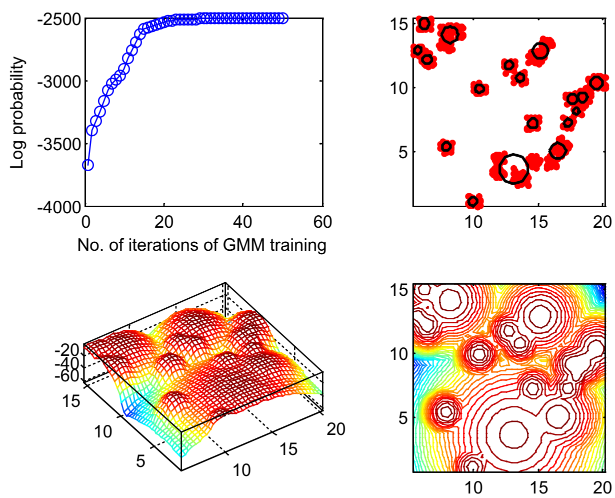


Fig. 8: Result of training the ID/IG ratio distribution with GMM for $M = 18$.

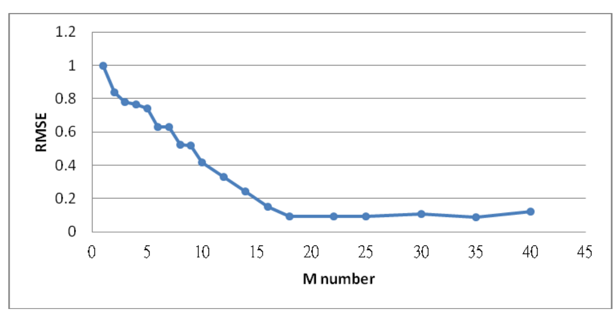


Fig. 9: RMSE for different M.

5.3 Specific properties MWCNT fabrication controlled by GMM

In the MPCVD system, the plasma distribution caused by different E-H tuner positions also affected the I_D/I_G ratio of MWCNTs. Fig. 10 shows the I_D/I_G ratio variation in Raman spectroscopy for 32 different positions in x and y. By the morphology of produced nanotubes shown by SEM, there were four examples that confirm that not only the I_D/I_G ratio varied but also that thickness and diameter varied (as shown in Fig. 11 and Table 2). This indicates the plasma density had a strong effect on the carbon atoms in nanotube production. The experimental results show that when the position for E-H tuner adjustment is at $x = 19.3$ cm, $y = 10$ cm, then the best average I_D/I_G ratio 0.336, the average films thickness is 41.77 μm, and the average tube diameter is 31.26 nm. On the other hand, when (X,Y) position is (7.3 cm, 13.9 cm), the I_D/I_G is 1.32, the average films thickness is 9 μm, and average tube diameter is 57.78 nm. The SEM morphology and Raman spectra of these two samples are shown in figures 12 and 13. Comparing with these the two micro-structures by field emission test, the result shows that the lower the I_D/I_G ratio, the better turn on voltage 0.54 V/μm, but the turn on voltage of higher I_D/I_G ratio is 0.82 V/μm.

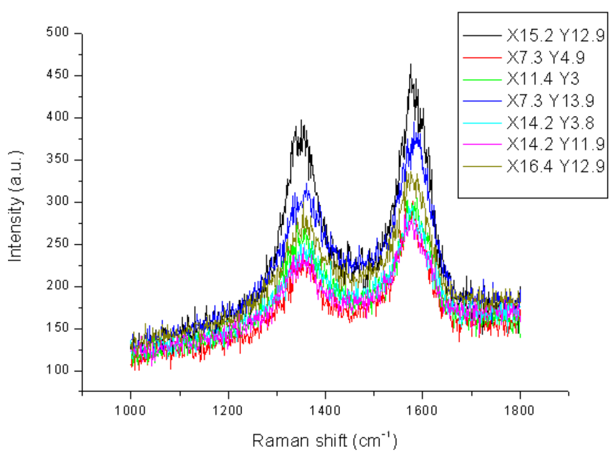


Fig. 10: Raman spectra for different E-H tuner positions.

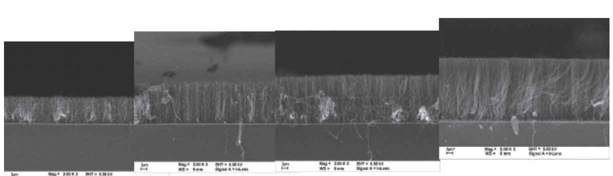

(a) (b) (c) (d)

Fig. 11: SEM morphology for different E-H tuner positions.

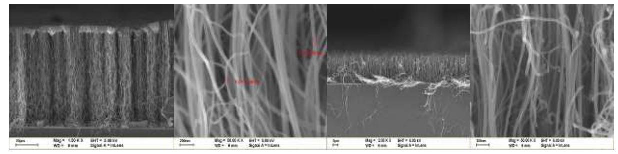

(a) (b) (c) (d)

Fig. 12: SEM morphology for different E-H tuner positions.

Table 2: The positions of figure (a)–(d)

	(a)	(b)	(c)	(d)
Position(X, Y) unit: cm	(8.2, 13.9)	(7.3, 12.9)	(16.4, 4.9)	(17.7, 7.9)
I_D/I_G ratio	0.986	1.1	1.112	0.84
Thickness unit: μm	8.11	12.28	14.56	19.27

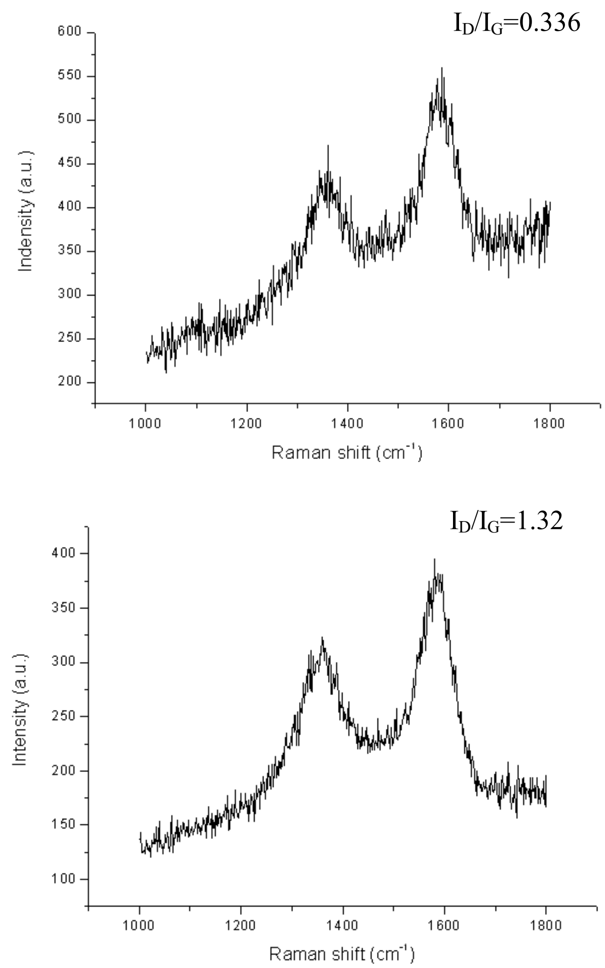


Fig. 13: Raman spectra for different E-H tuner positions.

6 Conclusions and future works

In this paper, we proposed a GMM approach to modeling the microwave power in MPCVD systems. The results of GMM training that the performance index (RMSE) can be reduced to under 10 %. The microwave power distribution corresponding to the adjustable electromagnetic field can be modeled by the 2-D GMM. The results of modeling show that microwave power can be simplified to be a linear combination of some Gaussian functions that provides a predictable and controlled basis for tuning manufacturing parameters and plasma sharp in a real-time control. Each E-H tuner position can fabricate the MWCNTs films with a high reproducibility after GMM modeling. The different micro-structure had the different application properties. This study shows that the system could make more applications by a high reproducibility MWCNTs fabrication.

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