Neural network based modeling of PL intensity in PLD-grown ZnO thin films

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Abstract

The process modeling of ZnO thin films grown by pulsed laser deposition (PLD) was investigated using neural networks based on radial basis function networks (RBFN) and multi-layer perceptron (MLP). Two input factors were examined with respect to the response factor, photoluminescence (PL), which is one of the main factors to determine the optical characteristic of the structure. In order to minimize the joint confidence region of fabrication process with varying the conditions, D-optimal experimental design technique was performed and PL intensity was characterized by neural networks. The statistical results were then used to verify the fitness of the nonlinear process model. Based on the results, this modeling methodology can optimize the process conditions for semiconductor manufacturing.

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1. Introduction

The semiconductor manufacturing process exhibits the nonlinear characteristics in general because of the nature of the process and the unavoidable random variations of the process. One of the most important issues for semiconductor manufacturing process is process optimization, which is difficult to characterize the process from the relationship between process input factors and the performance metrics with the process fluctuations. In order to characterize the nonlinear process model, neural networks (NNets) developed. However, the accurate explanation by the process model can significantly impact the values and the range of process input factors. Therefore, the statistical variations of process input factors must be carefully examined to build the process model by considering the ranges and the values of process variables.

The efforts to develop the methodology to characterize the nonlinear process have been pursued by several researchers. Gianchandani and Selden Crazy [1] used I- and D- optimal design to optimize the parametric modeling of a micromachinerometer. Rafiq et al. [2] represented three types of NNets, MLP, RBFN and normalized radial basis function (NRRB), for engineering applications. Byungwhan Kim et al. [3] investigated the statistical experimental design and process model for plasma etching using NNets. May [4] performed modeling and optimization of integral capacitor fabrication using NNets.

In this paper, NNets based on the RBFN and MLP were applied to the PLD process modeling. PL intensity, which is one of the main characteristics of ZnO thin film, was measured to predict the model of PLD process. Prior to experiment, the D-optimal design was used to make design matrix in this experiment efficiently. Considering the ranges and the values of input factors of the process, RBFN and MLP based NNets was carried out to build the process model. The fitness of the model between simple linear regression model and NNet models were analyzed by analysis of variance (ANOVA). The hypothesis testing and the assumption of the model were also verified. Based on the results, this modeling methodology can be extended to allow device engineers to predict the process characteristics.

2. Experiments

The initial substrate of n-type InP has 3 × 10^{18} cm^{-3} doping concentration. The PLD technique has been used...
for the deposition of n-type ZnO layer. The chamber was evacuated by a turbomolecular pump to base pressure $1 \times 10^{-6}$ Torr. Pulsed Nd:YAG laser was operated at a wavelength of 355 nm and repetition rate of 2 Hz. The laser energy density was $2.5 \text{ J/cm}^2$. The ZnO films were deposited with designated ranges, substrate temperature was $350$–$450 \text{ }\circ\text{C}$ and oxygen pressure was $250$–$450 \text{mTorr}$. A substrate holder was placed at 5 cm from the target [5]. After ZnO thin film deposition by PLD, the diffusion process was performed. The Zinc diffusion process maintaining temperature at about $495 \text{ }\circ\text{C}$ in flowing $\text{N}_2$ gases with varying the process conditions [6]. The ampoule in a quartz boat was loaded in the pre-heating zone and waited for 10 min before diffusion. After diffusion process was performed for a 60 min, the ampoule was pulled out of diffusion furnace and cooled rapidly by dropping it into water. After diffusion process was performed for a 60 min, the ampoule was pulled out of diffusion furnace and cooled rapidly by dropping it into water. After diffusion process was performed for a 60 min, the ampoule was pulled out of diffusion furnace and cooled rapidly by dropping it into water.

### 3. Modeling scheme

In order to characterize the PLD process, the variation of process variables, which are considered as input factors of interest, are summarized in Table 1. Substrate temperature ($T$) and oxygen pressure ($P$) were selected to investigate the characterization of PLD process. These input factors were explored via D-optimal experimental design with 12 runs to minimize the volume of the joint confidence region on the vector of regression coefficients [7]. Table 2 shows the experimental design matrix of PLD process factors used in each run. The run order has been randomized to avoid statistically the effect of irrelevant process factors used in each run. The run order has been randomized to avoid statistically the effect of irrelevant process factors used in each run. The designated input variables by D-optimal design were used in input layer. Each hidden layer has seven and two nodes with activation function, designated ranges, substrate temperature was $350$–$450 \text{ }\circ\text{C}$ and oxygen pressure was $250$–$450 \text{mTorr}$. A substrate holder was placed at 5 cm from the target [5]. After ZnO thin film deposition by PLD, the diffusion process was performed. The Zinc diffusion process maintaining temperature at about $495 \text{ }\circ\text{C}$ in flowing $\text{N}_2$ gases with varying the process conditions [6]. The ampoule in a quartz boat was loaded in the pre-heating zone and waited for 10 min before diffusion. After diffusion process was performed for a 60 min, the ampoule was pulled out of diffusion furnace and cooled rapidly by dropping it into water. After diffusion process was performed for a 60 min, the ampoule was pulled out of diffusion furnace and cooled rapidly by dropping it into water.

### 4. Results and discussion

The total deviation consists of the sum between the deviation of the fitted value, $\hat{Y}$, around the mean, $\bar{Y}$, and the deviation of the observation, $Y$, around fitted regression line. The measure of total variation, denoted by total sum of square (SSTO), is the sum of the squared deviations. Regression sum of square (SSR) and sum of square error (SSE) represent the sum of square of the regression and error, respectively. The relationship among the each sum of square error

### Table 1. Summary of process parameters

<table>
<thead>
<tr>
<th>Factor</th>
<th>Symbol</th>
<th>Unit</th>
<th>Value</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>Substrate temperature</td>
<td>$T$</td>
<td>°C</td>
<td>350–450</td>
<td>Controllable</td>
</tr>
<tr>
<td>Oxygen pressure</td>
<td>$P$</td>
<td>mTorr</td>
<td>250–450</td>
<td>Controllable</td>
</tr>
<tr>
<td>Laser energy density</td>
<td>–</td>
<td>J/cm²</td>
<td>2.5</td>
<td>Fixed</td>
</tr>
<tr>
<td>Distance</td>
<td>–</td>
<td>cm</td>
<td>5</td>
<td>Fixed</td>
</tr>
</tbody>
</table>

### Table 2. D-optimal design

<table>
<thead>
<tr>
<th>Run</th>
<th>$T$ (°C)</th>
<th>$P$ [mTorr]</th>
<th>Remark</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>400</td>
<td>450</td>
<td>TR</td>
</tr>
<tr>
<td>2</td>
<td>400</td>
<td>300</td>
<td>TR</td>
</tr>
<tr>
<td>3</td>
<td>375</td>
<td>550</td>
<td>TR</td>
</tr>
<tr>
<td>4</td>
<td>350</td>
<td>250</td>
<td>TR</td>
</tr>
<tr>
<td>5</td>
<td>425</td>
<td>400</td>
<td>TR</td>
</tr>
<tr>
<td>6</td>
<td>425</td>
<td>300</td>
<td>TR</td>
</tr>
<tr>
<td>7</td>
<td>400</td>
<td>400</td>
<td>TR</td>
</tr>
<tr>
<td>8</td>
<td>450</td>
<td>350</td>
<td>TR</td>
</tr>
<tr>
<td>9</td>
<td>400</td>
<td>350</td>
<td>TR</td>
</tr>
<tr>
<td>10</td>
<td>425</td>
<td>350</td>
<td>TR</td>
</tr>
<tr>
<td>11</td>
<td>375</td>
<td>400</td>
<td>TR</td>
</tr>
<tr>
<td>12</td>
<td>450</td>
<td>450</td>
<td>TR</td>
</tr>
<tr>
<td>13</td>
<td>400</td>
<td>250</td>
<td>TR</td>
</tr>
<tr>
<td>14</td>
<td>350</td>
<td>450</td>
<td>TR</td>
</tr>
<tr>
<td>15</td>
<td>350</td>
<td>350</td>
<td>TE</td>
</tr>
<tr>
<td>16</td>
<td>375</td>
<td>800</td>
<td>TE</td>
</tr>
<tr>
<td>17</td>
<td>450</td>
<td>250</td>
<td>VA</td>
</tr>
</tbody>
</table>

TR: training data; TE: testing data; VA: validation data.

A typical radial basis function (RBF) is defined as:

$$ f(x) = \sum_{i=1}^{N_R} w_i \phi_i(x) $$  \hspace{1cm} (1)

where $x$ is the input factor, $f(v)$ is the response of the output node, $w_i$ are the output linear combining weights. $\phi_i(x)$ is RBF and $N_R$ is the number of RBFs [8,9].

The most frequently used RBF is the Gaussian basis function that is defined as:

$$ G(x, c, r) = \exp\left(\frac{|x - c|^2}{2r^2}\right) $$  \hspace{1cm} (2)

where $|x - c|$ is the Euclidian distance between an input vector $x$ and a center $c$ and $r$ is the radius. Other types in this study were used for RBFs that are sigmoid function and tangent hyperbolic at each hidden layer [10].

NNets has a learning rate of 0.7 and a momentum coefficient of 0.4. These NNets were trained on 14 experimental runs. The two trials are used in testing data and the one trial is used in validation data. Network training was completed when root mean square (RMS) training error and validation error of 0.1% were achieved.
and the notations are in the following form:

\[
SSTO = SSR + SSE
\]  

(3)

\[
\sum(Y_i - \bar{Y})^2 = \sum(Y_i - \hat{Y})^2 + \sum(\hat{Y}_i - \bar{Y})^2
\]  

(4)

where \(Y_i\) is the observations, \(\hat{Y}_i\) is the predicted value, and \(\bar{Y}\) is the mean of the fitted values. The \(R^2\) value represents the proportion of variation in the response explained by the model. Therefore, the \(R^2\) value can be expressed as the reduction in variation \((SSTO - SSE = SSR)\), as a proportion of the total variation:

\[
R^2 = \frac{SSR}{SSTO} = \frac{SSTO - SSE}{SSTO}
\]  

(5)

The portion of the model explanation in NNets is better than the portion of the regression model. These regression models have the following form:

\[
Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_1 X_2 + \varepsilon
\]  

(6)

where \(Y\) is a dependent variable, \(X_3\) are the independent variables, \(\beta_s\) are regression coefficients estimated using the least squared method, and \(\varepsilon\) is a model error. The root mean square error (RMSE) is defined as:

\[
RMSE = \sqrt{\frac{\sum SSE}{n - 1}}
\]  

(7)

where \(n\) is the size of the sample. The \(F\)-values are calculated, analysis of variance (ANOVA). The \(F\)-values are the following form:

\[
F = \frac{MSR}{MSE}
\]  

(8)

where MSR is the mean sum of square of the regression, MSE is the mean sum of square error. The test for significance of regression is to determine if there is a linear relationship between the response variable and a subset of the regression variables. The appropriate hypotheses are \(H_0: \beta_3 = \beta_4 = \cdots = \beta_k = 0\) and \(H_1: \beta_j \neq 0\) for at least one \(j\) [11]. The \(F\)-value in simple linear regression model, 0.1, was less than \(F\) statistics. It means that \(H_0\), the null hypotheses, was accepted in hypothesis testing under \(\alpha = 0.05\). There was no linear relationship between the response variable and the regression variable. On the contrary, the \(F\)-value in NNets, 2856.8, is larger than \(F\) statistics. Based on the results, it is verified that NNets is superior to simple linear regression model in order to optimize the characteristics of the nonlinear process. The \(R^2\) value represents the proportion of variation in the response explained by the model. In case of linear model, the \(R^2\) value is 0.021. NNets is 0.996. It shows that NNets provides better explanation of the variation in the PL spectrum [12]. The RMSE and mean absolute (MA) error were 12.71 and 11.56, respectively. MA is 4.10%. The plots of residual versus fitted value are shown in Fig. 1. One of the assumptions of this analysis is that the residuals are both normally and randomly distributed [13]. It is observed that the residuals should be scattered evenly about zero and there is no special features or patterns in residuals. Linearity of predicted value versus measured value was illustrated in Fig. 2. It is verified that the fitted line between predicted value and measured value is the linear relationship. From these verifications, the sensitivity of the modeling results is investigated. Fig. 3 shows the sensitivity at pressure is 350 mTorr and temperature is 400 °C. This fact revealed that this condition is the optimal point for PLD process to optimize the PL intensity. These results agreed well with experimental results. Fig. 4 shows the sensitivity at pressure was 400 mTorr and temperature was 425 °C. As the temperature increased from 400 to 425 °C, the MLP result of the modeling decreases by 83.71%, from 686.2 to 111.7. Fig. 5 shows the contour plot of response surface model. It shows that the optimum condition of PL intensity for PLD-grown ZnO thin films is observed at pressure is 350 mTorr and temperature is 400 °C. It is confirmed that the second optimum point exists at pressure is 400 mTorr and temperature is 425 °C. Fig. 6 shows the contour plot of response surface model. Although this model has the local minimum between the first optimum point and the second optimum point, it is represented that the status of the comprehensive response...
model by NNets based on the RBFN and MLP. These NNet models are in good agreement with experimental results.

5. Conclusion

In order to characterize the response factor of PLD process, PL, the D-optimal design and NNets based on the RBFN and MLP have been performed. The D-optimal design was selected in order to minimize the joint confidence
region on the vector of regression coefficients and NNets was performed to characterize the nonlinear PLD process. Based on the modeling results, NNets exhibited the good agreement between the measured and the predicted performance metrics. The modeling can be finding the optimal point of PLD process that has a different experiment condition. Although the manufacturing process has nonlinear properties, it can control process through the predicted value obtained from design of experiment and NNets used modeling methodology. Therefore, this methodology can be used to improve the manufacturability of the semiconductor manufacturing.

Acknowledgements

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References