Hybrid Metamodel—NSGA-III—EDAS Based Optimal Design of Thin Film Coatings

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Abstract: In this work, diamond-like carbon (DLC) thin film coatings are deposited on silicon substrates by using plasma-enhanced chemical vapour deposition (PECVD) technique. By varying the hydrogen (H2) flow rate, CH4−Argon (Ar) flow rate and deposition temperature (Td) as per a Box-Behnken experimental design (BBD), 15 DLC deposition experiments are carried out. The Young’s modulus (E) and the coefficient of friction (COF) for the DLCs are measured. By using a second-order polynomial regression approach, two metamodels are built for E and COF, that establish them as functions of H2 flow rate, CH4−Ar flow rate and Td. A non-dominated sorting genetic algorithm (NSGA-III) is used to obtain a set of Pareto solutions for the multi-objective optimization of E maximization and COF minimization. According to various practical scenarios, evaluation based on distance from average solution (EDAS) approach is used to identify the most feasible solutions out of the Pareto solution set. Confirmation experiments are conducted which shows the efficacy of the polynomial regression—NSGA-III—EDAS hybrid approach. The surface morphology of the DLCs deposited as per the optimal predictions is also studied by using atomic force microscopy.

Keywords: Multi-objective optimization; regression analysis; thin-film coating

1 Introduction

The excellent mechanical, tribological and optical properties of diamond-like carbon (DLC) coatings offer a wide application in the automotive and electronic industries. DLC coating is a mixture of both graphite-like sp2 bond and diamond-like sp3 bonds which shows that the properties of DLCs depend on the number of bonds present within the coatings. DLCs are considered to be the hybrid form of carbon which holds both graphite-like sp2 bond and diamond-like sp3 bonds [1]. It is well known that graphite...
(100% sp2) is having a zero-band gap whereas diamond (100% sp3 bond) has a bandgap of 5.5 eV and thus, by the synthesis of DLCs by using different methods the amount of sp3 and sp2 bonds can be altered [2]. Due to the mixture of sp2 and sp3 bonds within the DLC films, it possesses characteristics of both graphite and diamond. The good electrical and electronic properties of DLC are due to the sp2 hybridized carbon whereas, the tribological and mechanical properties are due to the sp3 hybridized carbon [3,4]. DLC films are also used for making surgical equipments, automotive engine parts, magnetic storage discs, micro-electromechanical devices (MEMS), etc.

Various chemical vapour deposition (CVD) techniques are used for the deposition of thin-film coatings and out of all the CVD techniques, plasma-enhanced chemical vapour deposition (PECVD) technique is most widely used for the DLC coating synthesis because of its quality coating at low temperature [5–7]. The different PECVD deposition parameters like gas flow rate, duty cycle, gas composition, deposition temperature, power supply etc. influence the properties of DLC thin films. In recent years, the selection of deposition parameters is a matter of prime concern for the researchers to get the desired properties of the films [8,9]. Singh et al. [10] used a Taguchi technique to find the combination of PECVD deposition parameters like bias voltage (V), bias frequency (f), gas composition, deposition pressure (P) of DLC coatings with PECVD technique to get optimum response parameters, i.e., roughness and hardness. Ghadai et al. [11] used particle swarm optimization (PSO) techniques to optimize the PECVD process parameters to get high hardness of the DLC coating. In an extension to the previous work, Ghadai et al. [12] found that symbolic regression metamodels are superior to traditional polynomial regression metamodels. The symbolic regression metamodels were form-free and thus, were better at modelling the inherent non-linearity in the deposition process. Ghadai et al. [13] also used a genetic algorithm to fine-tune the DLC deposition parameters in APCVD process.

Despite the considerable amount of work done on DLC thin film coatings, only a handful of works are seen on the implementation of advance computational intelligence techniques like multi-objective optimization based on metamodels. This work attempts to address this lacuna by building metamodels that express two different DLC performance parameters (Young’s modulus and coefficient of friction) as functions of three DLC deposition process parameters (hydrogen flow rate, CH4-Argon flow rate and deposition temperature). The metamodels are then deployed in conjunction with the non-dominated sorting genetic algorithm (NSGA-III) for carrying out Pareto optimization. Finally, based on certain scenarios, a multi-criteria decision-making method called EDAS is used to identify the desirable solutions from the Pareto set. Surface morphologies of these optimal designs are studied by using atomic force microscopy.

2 Materials and Methods
2.1 Experimental Procedure

In the work, the synthesis of DLC coatings over silicon (Si) was done by using PECVD deposition technique. To remove the oxide layer, the substrates were dipped into 2% HF solution for 4 min followed by ultrasonic cleaning in deionized water for 10 min. Tab. 1 shows the details of input parameters like hydrogen (H2) flow rate, CH4-Argon (Ar) flow rate and temperature for the deposition (Td) of DLC coatings. The morphological analysis of the DLC coating is done with the help of Innova SPM atomic force microscope. The Young’s modulus (E) of the DLC coatings were calculated by using a nano-hardness tester (NHTX-55-0019) of CSM Instruments having Berkovich indenter. The radius of curvature of the indentor (B-I 93) is 20 μm. The indentation was considered at three different locations and the average of that value is considered and the maximum load is taken as 10 mN. The Oliver–Pharr method [14] is applied for the calculation of Young’s modulus (E). The nano scratch tests were performed over the coating by using CSM instrument with a spherico-conical diamond indenter (R = 2 μm, SB-A63), by applying a load of 20 mN with a scratch speed of 1 mm/min over a 0.5 mm scratch length.
2.2 Predictive Modeling with Polynomial Regression

In this work, the metamodels for Young’s modulus and the coefficient of friction are built by fitting a second-order polynomial regression equation of the following form.

\[
y_i = b_0 + b_1 x_1 + b_2 x_2 + b_3 x_3 + b_4 x_1 x_2 + b_5 x_1 x_3 + b_6 x_2 x_3 + b_7 x_1^2 + b_8 x_2^2 + b_9 x_3^2 \] (1)

Here \( b \)'s are the coefficients of regression. These coefficients of regression help in describing the response \( (y_i) \) as a function of predictor variables \( (x's) \). \( x_1, x_2 \) and \( x_3 \) represent hydrogen (H\(_2\)) flow rate, CH\(_4\)-Argon (Ar) flow rate and deposition temperature \( (T_d) \) respectively.

Using the Box–Behnken experimental design in Tab. 1, Eq. (1) is fitted based on multiple regression fitting scheme. The difference between the predicted value \( (\hat{y}_i) \) and the actual experimental value \( (y_i) \) of the response is called the residue [15].

\[
\epsilon_i = y_i - \hat{y}_i \] (2)

\( \beta \)'s in Eq. (1) are computed such that the residual sum of squared (RSS) is minimized.

\[
RSS = \sum_{i=1}^{n} \epsilon_i^2 \] (3)

where \( n \) is the number of experimental points in Tab. 1.

Table 1: Experimental readings of Young’s modulus and coefficient of friction measured at selected BBD sample points

| Exp. no. | CH\(_4\)-Argon flow rate | H\(_2\) flow rate | Deposition temperature | Young’s modulus (Gpa) | Coefficient of friction |
|---------|--------------------------|-------------------|------------------------|-----------------------|------------------------|
| 1       | 0.5                      | 40                | 100                    | 146.80                | 0.100                  |
| 2       | 2                        | 30                | 80                     | 137.50                | 0.126                  |
| 3       | 1                        | 30                | 100                    | 178.00                | 0.087                  |
| 4       | 2                        | 30                | 120                    | 154.00                | 0.082                  |
| 5       | 1                        | 20                | 120                    | 138.80                | 0.150                  |
| 6       | 0.5                      | 20                | 100                    | 152.30                | 0.142                  |
| 7       | 1                        | 20                | 80                     | 128.56                | 0.110                  |
| 8       | 2                        | 40                | 100                    | 179.20                | 0.102                  |
| 9       | 1                        | 40                | 80                     | 110.24                | 0.200                  |
| 10      | 0.5                      | 30                | 120                    | 186.54                | 0.090                  |
| 11      | 1                        | 30                | 100                    | 166.20                | 0.130                  |
| 12      | 0.5                      | 30                | 80                     | 106.10                | 0.160                  |
| 13      | 1                        | 40                | 120                    | 242.00                | 0.070                  |
| 14      | 1                        | 30                | 100                    | 171.30                | 0.131                  |
| 15      | 2                        | 20                | 100                    | 146.23                | 0.180                  |
2.3 Optimization with NSGA-III

In this work, non-dominated sorting genetic algorithm III (NSGA-III) \([16,17]\) is used for carrying out the Pareto optimization. The multi-objective optimization problem is stated as,

Find \(X = (x_1, x_2 \text{ and } x_3)\) which maximizes \(Y_1 = f_1(X)\) and minimizes \(Y_2 = f_2(X)\) \hspace{1cm} (4)

subject to \(x_1^l \leq x_1 \leq x_1^u; \ x_2^l \leq x_2 \leq x_2^u; \ x_3^l \leq x_3 \leq x_3^u\)

In Eq. (4), \(Y_1\) and \(Y_2\) are Young’s modulus (E) and coefficient of friction (COF) respectively.
NSGA-III is realized in this work by using the following pseudo-code.

---

START
Define objective functions \(Y_1\) and \(Y_2\)
Initiate generation counter \(t = 0\)
Initiate a random population of \(n_{\text{pop}}\) individuals
Calculate the fitness of each individual
Conduct non-dominated sorting of individuals
Assign ranks and select parents
Generate child population
Tournament selection
Crossover and mutation

Do
    Do for all individuals
    Calculate fitness
    Conduct non-dominated sorting
    Generate Pareto fonts
    Determine crowding distance
    Loop inside by adding solution to next generation from the first front until \(n_{\text{pop}}\)

END
Select points on lower front with high crowding distance
Create the next generation
Tournament selection
Crossover and mutation

Until \(t = t_{\text{max}}\).
Report the Pareto front having \(P_{\text{nd}}\) non-dominated solutions

END
3 Results and Discussion

3.1 Predictive Modeling

Using the training data listed in Tab. 1, second-order polynomial regression metamodels are developed for the prediction of Young’s modulus (E) and the coefficient of friction (COF). The coefficients of regression for the metamodels of E and COF are mentioned in Tab. 2. Fig. 1 shows the variation of the predicted values of E and COF for their respective experimental values. It should be noted that closer the values are to the diagonal (identity) line in Fig. 1, better are the estimations of the metamodel. In general, the metamodel for E is seen to have better performance than COF. To further analyze the utility of the two metamodels, the residuals in each case are evaluated against their respective predicted values, as shown in Fig. 2. A random scatter is seen in both cases, which indicates that the residues do not show any trend with the predicted values. Thereby it can be concluded that the metamodels are appropriate as they can quantify the variance in the training data. Further analysis of the residuals is done by plotting their normal probability plots in Fig. 3. No outliers are seen in Fig. 3, which further confirms the efficacy of the metamodels.

| Table 2: Regression coefficients for the second-order metamodels |
|---------------------------------------------------------------|
| Regression coefficient | Young’s modulus | Coefficient of friction |
| β₀                    | −72.8926        | −2.31E-01               |
| β₁                    | 156.2961        | 2.66E-02                |
| β₂                    | −12.7616        | 1.20E-02                |
| β₃                    | 4.9633          | 4.71E-03                |
| β₄                    | 0.9127          | −1.82E-03               |
| β₅                    | −1.1282         | 3.47E-04                |
| β₆                    | 0.1519          | −2.13E-04               |
| β₇                    | −26.6650        | −2.83E-03               |
| β₈                    | −0.0342         | 1.65E-04                |
| β₉                    | −0.0338         | 2.14E-19                |

3.2 Influence of Process Parameters on the Young’s Modulus

Fig. 4 shows the effect of H2 and CH4—Ar flow rate on Young’s modulus (E) of the DLCs. It is seen that deposition temperature (Td) has a significant effect on the role that of H2 and CH4—Argon flow rate plays on E. For example—at a lesser Td, the E of the DLCs increase with the increase in CH4—Argon flow rate but the E decreases with an increase in H2 flow rate. The trend is the opposite when higher levels of Td is considered. Similarly, Fig. 5 shows the interactive effect of Td and CH4—Ar flow rate on E at various levels of H2 flow. It is observed that the E increases as the Td and CH4—Argon flow rate increases. Fig. 6 shows that the increase in H2 flow, in general, decreases the E of the DLCs.

3.3 Influence of Process Parameters on the Coefficient of Friction

Fig. 7 shows the effect of H2 and CH4—Ar flow rate on the coefficient of friction (COF) of the DLCs. It is seen that the trend of the COF of the DLCs is also significantly affected by the Td. At low Td, the increase in H2 and CH4—Argon flow rate increases the COF of the DLCs, whereas at higher levels of Td, the COF decreases with an increase in H2 flow rate but CH4—Argon flow rate has a negligible effect on it. In Fig. 8, the trend of COF with Td and CH4—Argon flow rate is similar for mid and high level H2 flow rate but is
significantly different for low H₂ flow rate. At mid and high H₂ flow rate, the COF increases with a decrease in Tₐ, but is not much affected by variation in CH₄—Argon flow rate. However, as seen in Fig. 9, the behaviour of the COF of the DLCs is similar at all levels of CH₄—Argon flow rate.

Figure 1: Predicted versus the experimental output responses. (a) Young’s modulus, (b) Coefficient of friction

Figure 2: Variation of the residuals with predicted output responses. (a) Young’s modulus, (b) Coefficient of friction
Figure 3: Normal probability plots for the residuals of the metamodels. (a) Young’s modulus, (b) Coefficient of friction

Figure 4: Variation of Young’s modulus with $H_2$ flow rate and $CH_4$—Argon flow rate at different deposition temperatures. (a) $T_d = 80°C$, (b) $T_d = 100°C$, and (c) $T_d = 120°C$
Figure 5: Variation of Young’s modulus with deposition temperature and CH$_4$—Argon flow rate at different H$_2$ flow rates. (a) 20 sccm, (b) 30 sccm, and (c) 40 sccm

Figure 6: Variation of Young’s modulus with deposition temperature and H$_2$ flow rate at different CH$_4$—Argon flow rates. (a) 0.5 sccm, (b) 1 sccm, and (c) 2 sccm
Figure 7: Variation of the coefficient of friction with H\textsubscript{2} flow rate and CH\textsubscript{4}—Argon flow rate at different deposition temperatures. (a) T\textsubscript{d} = 80\degree C (b) T\textsubscript{d} = 100\degree C (c) T\textsubscript{d} = 120\degree C

Figure 8: Variation of the coefficient of friction with deposition temperature and CH\textsubscript{4}—Argon flow rate at different H\textsubscript{2} flow rates. (a) 20 sccm, (b) 30 sccm, and (c) 40 sccm
3.4 Pareto Optimization

Based on the discussion on the effect of the DLC deposition process parameters on Young’s modulus and COF in the previous two sections, it is seen that the optimal setting of the two response parameters has conflicting requirements in terms of process parameter settings. Thus, it is not possible to arbitrary decide the optimal parameter combination that would simultaneously maximize the E and minimize the COF. Thus, a Pareto optimization using non-dominated sorting genetic algorithm is carried out and depicted in Fig. 10. The continuous Pareto front in Fig. 10b shows that as the E of the DLCs improve, there is an increase in COF as well. Thus, each solution within the Pareto front in Fig. 10b represents a possible compromise solution to the multi-objective problem. Since it is not possible to arbitrarily draw a particular solution out of the Pareto front to represent a feasible solution, a multi-criteria decision-making approach called EDAS is used to select the most plausible solutions pertaining to certain practical scenarios and are presented in Tab. 3.

3.5 Experimental Confirmation of Optimal Results

Confirmation experiments as per the optimal process parameters are conducted and the experimental values are reported in Tab. 3. The 3-D and 2-D atomic force microscopy (AFM) images of DLC coatings for the validation of optimal point of scenario (A/B/C), (D) and (E) are shown in Figs. 11–13, respectively. The surface roughness ($R_a$) of the coatings for scenario (A/B/C), (D) and (E) are 15.5 nm, 28 nm, and 32 nm respectively. From the figures, it is observed that small, agglomerated particles are formed for all the cases. The maximum and average particle size for the experimental results for scenario (A/B/C) is 7 nm and 1.5 nm, for scenario (D) the sizes are 15.4 nm and 3.4 nm, for scenario (E) the sizes are 44.2 nm and 7 nm respectively. In all the different scenarios the $H_2$ flow rate and deposition
temperature are the same, however, the CH\textsubscript{4}–Argon flow rate is different. From the experimental results shown in Tab. 3, it is observed that the COF of the DLC coating decreases with an increase in the CH\textsubscript{4}–Argon flow rate. From the AFM images, it is observed that the DLC coating having less COF has a smooth surface and result is confirmed from the \(R_a\) value. Overall, the confirmation experiment values are seen to be close to the predicted optimal solutions.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure10.png}
\caption{Multi-objective optimization of Young’s modulus and COF (a) Dominated and non-dominated solutions, (b) Enlarged view of Pareto frontier}
\end{figure}

\begin{table}[h]
\centering
\begin{tabular}{llllllllll}
Scenario & \(W_1(\%)\) & EDAS performance measure & CH\textsubscript{4}–Ar flow rate & H\textsubscript{2} flow rate & \(T_d\) & E (Gpa) & COF \\
\hline
A & 10 & 0.97832 & & & & & 0.03538 \\
B & 25 & 0.93497 & 2.00 & 40 & 120 & 208.08595 & 214 & 0.066 \\
C & 50 & 0.80491 & & & & & 0.04379 \\
D & 75 & 0.62844 & 1.41\textsuperscript{2} & 40 & 120 & 227.92423 & 212 & 0.04379 \\
E & 90 & 0.86518 & 1.14\textsuperscript{3} & 40 & 120 & 230.71794 & 219 & 0.04698 \\
\end{tabular}
\caption{EDAS selected optimal solution from the Pareto front}
\end{table}

\textsuperscript{1} \(W_1\) is the weight or importance attributed to the criteria Youngs modulus. \(100-W_1\) is the weight attributed to the criteria COF.

\textsuperscript{2} For experiments CH\textsubscript{4}–Argon flow rate was considered as 1.5.

\textsuperscript{3} For experiments CH\textsubscript{4}–Argon flow rate was considered as 11.
Figure 11: AFM images of deposited DLCs as per scenario A/B/C

Figure 12: AFM images of deposited DLCs as per scenario D
4 Conclusion

Finding an optimal combination of process parameters that enhances the performance of a process is a realistic goal with tremendous practical implications. In this work, such an effort for optimizing the DLC deposition process parameters is undertaken to suitably enhance Young’s modulus and coefficient of friction of DLC thin film coatings. Based on the study the following conclusion are made:

- Second-order polynomial regressions can serve as reliable metamodels for DLC process modelling that can be subsequently used for process parameter effect study or in case of optimization scenarios.
- Non-dominated sorting genetic algorithm (NSGA-III) is a viable tool for Pareto optimization of such critical processes. Improvement in Young’s modulus of the DLCs was in general accompanied by worsening of the coefficient of friction. Thus, given such conflicting process performance, the Pareto set, as opposed to single-objective solutions, can provide the designer with a lot of flexibility regarding setting the preferred process parameters.
- In general, a higher level of hydrogen flow rate and deposition temperature was found to be suitable in augmenting the young’s modulus of the DLCs.
- Confirmation experiments conducted as per the optimal process parameters showed that the polynomial regression—NSGA-III—EDAS approach is reliable and accurate.

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