RESEARCH ARTICLE

Determining Optimum Parameters for Adhesion Properties of Ta-DLC Films Using Taguchi Experimental Design

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Abstract

DLC films have high biocompatibility. But due to low adhesion, the element must be doped. Therefore, in this study, the high biocompatibility Ta element was doped into DLC film and optimum parameters for the adhesion of Ta-DLC films were determined. The Taguchi experimental system was used to determine the optimum parameters. Ta target current, substrate bias voltage and duty cycle were selected as variable parameters. The deposition was performed with HiPIMS-CFUBMS system. Ta-DLC films were grown on Ti6Al4V substrates using Taguchi $L_9 3^4$ orthogonal experimental system. The optimum deposition parameters for critical load of Ta-DLC films are 3A, -80V and %5 for Ta target current, substrate bias voltage and duty cycle, respectively.

Keywords: Ta-DLC, Magnetron Sputtering, HiPIMS, Taguchi, Adhesion

1. Introduction

DLC films are preferred in many fields from medical to manufacturing due to their high hardness, low friction coefficient and biocompatibility [1, 2]. However, DLC has low adhesion due to the internal stress of the films [3]. For this reason, metal (Ti, Ta, Cr) or nonmetal (N₂, B) is doped to increase adhesion [4-8]. Ta is a chemically stable, highly melting and biocompatible element. When literature review is made, the publication of Ta-DLC film is very limited [9-14]. In fact, there is no study about the adhesion of Ta doped DLC films. DLC films are commonly coated with CVD and PVD techniques [2]. High Power Impulse Magnetron Sputtering (HiPIMS) technique is the latest technology among PVD techniques. The films coated with HiPIMS have better adhesion, dense structure and higher hardness than the films coated by conventional magnetron sputtering methods [15, 16]. When the literature is searched, the parameters affecting the adhesion of DLC films are target current, substrate bias voltage, gas content. In addition, parameters affecting adhesion in HiPIMS system are duty cycle, target voltage, gas content and substrate bias voltage [5, 17-19].

In this study, optimum parameter conditions were determined for the adhesion value of Ta doped DLC films. Taguchi $L_9 3^4$ orthogonal experimental design was chosen to determine optimum parameters. Ta target current, substrate bias voltage and duty cycle were selected as variable parameters.

2. Experimental methods

Taguchi experimental method was used to determine the optimum deposition parameters for adhesion properties of Ta-DLC films. The test parameters selected for this study were: Ta target current (A), substrate bias voltage (-V) and duty cycle (%). For this parameters,

three different values were obtained as shown in Table 1. Also, the constant parameters are given in Table 1. Taguchi experimental design ($L_9 3^4$ orthogonal experimental design) is given in Table 2. Ta-DLC films were deposited onto Ti6Al4V materials used as implant materials using combination of HiPIMS and pulsed-dc. The coating system was arranged as Closed Field Unbalanced Magnetron Sputtering (CFUBMS). In the system two C, one Ta and one Ti targets, Ar and N₂, C₂H₂ gases were used. C targets were supplied by HiPIMS and pulsed-dc was applied on Ti and Ta targets.

Table 1. Variable and constant parameters

Variable Parameters	LEVELS		
	1	2	3
Ta target current (A)	3	4	5
Substrate Bias Voltage (-V)	80	90	100
Duty cycle (%)/Duty time (µs)	4/80	5/100	6/120
Constant Parameters			
Frequency (Hz)	500		
C target voltage (-V)	800		
Working Pressure (Pa / mTorr)	0.33 / 2.5		
C ₂ H ₂ content	%30 (3sccm)		
Ti interlayer	Ti: 5A (10 min)		
Ta doped c-BN deposition time	90 min		
Frequency and duty time for Pulsed-dc (kHz/µs)	150/2		

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Table 2. Taguchi experimental design

Test No	R1	R2	R3	R4	R5	R6	R7	R8	R9
Ta target current (A)	3	3	3	4	4	4	5	5	5
Substrate Bias Voltage (-V)	80	90	100	80	90	100	80	90	100
Duty cycle (%)	4	5	6	5	6	4	6	4	5

SEM (Zeiss Sigma 300) device was used to determine thickness of Ta-DLC films. Raman (WITech Alpha 300R) were conducted to obtain the sp^3/sp^2 ratio of Ta-DLC films. The hardness values were measured using microhardness tester (Buehler Micromet 2001) with the load of 10gf and indentation time of 15s. To determine critical load values of Ta-DLC films, scratch test (Anton Paar (Formerly CSM)) was carried out with indenter of Rockwell C (200 μ m radius), sliding speed of 10mm/min and loading rate of 100N/min.

3. Results and Discussions

A typical cross-section SEM image for Ta-DLC film was given in Fig 1. The highest film thickness obtained from Ta-DLC films was 1,318 μ m from R7 parameters and the lowest film thickness was 158 nm from R1 parameters. The highest film thickness was obtained at the highest Ta target current (5A) and highest duty cycle (6%) parameters. The lowest film thickness was obtained at the lowest Ta target current (3A) and the lowest duty cycle (4%) parameters. There was no effect of substrate bias voltage on film thickness. Efeoglu et al. (2018) reported Ta-DLC film thickness increased with increasing Ta target current [12]. Decho et al. (2016) stated that the film thickness increased with increasing duty cycle while keeping the other parameters constant [20].

Figure 1. A typical cross-section SEM image for Ta-DLC film

Raman graphs of Ta-DLC films deposited with Taguchi experiment design were given in Fig. 2. All films have D and G bands. The low D peak improves the quality and performance of DLC films [21, 22]. The sp^3/sp^2 ratio of Ta-DLC films was given in Table 3. The highest sp^3/sp^2 ratio was 0.527 in the R8 film and the lowest sp^3/sp^2 ratio was 0.471 in the R3 film (Table 3).

Figure 2. Raman graphics of films deposited with R1-R9 parameters

The hardness of the DLC films increases with increasing sp^3 ratio [23]. Table 3 shows the sp^3/sp^2 ratio and hardness values of the Ta-DLC coatings. As shown in Table 3, the R8 film with the highest sp^3/sp^2 ratio (0.527) had the highest hardness (41.2 GPa).

Table 3. sp^3/sp^2 ratio and hardness values of Ta-DLC films

	R1	R2	R3	R4	R5	R6	R7	R8	R9
sp^3/sp^2 ratio	0.4	0.4	0.4	0.5	0.5	0.5	0.5	0.5	0.5
Hardness value (GPa)	30	24	18	34	38	40	38	41	37

Normal load and friction graphs obtained from scratch tests for Ta-DLC films were given Fig. 3-Fig.11. The highest critical load and the lowest critical load values were obtained as 30N in R3 coating and 5N in R8 coating, respectively. The highest critical load value was obtained at the lowest hardness value. The hardness values of Ta-DLC films increase and critical load values decrease with increased substrate bias voltage and target current [24].

Figure 3. Normal load and friction graph for Ta-DLC film deposited R1 parameters

Figure 4. Normal load and friction graph for Ta-DLC film deposited R2 parameters

Figure 5. Normal load and friction graph for Ta-DLC film deposited R3 parameters

Figure 6. Normal load and friction graph for Ta-DLC film deposited R4 parameters

Figure 7. Normal load and friction graph for Ta-DLC film deposited R5 parameters

Figure 8. Normal load and friction graph for Ta-DLC film deposited R6 parameters

Figure 9. Normal load and friction graph for Ta-DLC film deposited R7 parameters

Figure 10. Normal load and friction graph for Ta-DLC film deposited R8 parameters

Figure 11. Normal load and friction graph for Ta-DLC film deposited R9 parameters

The calculated signal/noise (S/N) rates for the measured critical load values according to the Taguchi experimental design are given in Table 4. As very well known; the S/N ratio is a measure of the process variability. By optimizing the process parameters by using S/N rate according to Taguchi experimental design, it is verified that optimum process conditions are effective and stable, meaning that they have minimum process variability [25]. For critical load values of Ta-DLC films, the higher is the better. Therefore, S/N values (see in Table 4) is calculated by the following Equation 1;

$$S/N = -10 \log \frac{1}{n} \sum_{i=1}^n \frac{1}{y_i^2} \dots\dots\dots (1)$$

Where S/N is Signal/Noise ratio, y is experimental data and n is experimental number.

Table 4. The calculated S/N rates for the critical load of Ta-DLC films

	R1	R2	R3	R4	R5	R6	R7	R8	R9
The critical load (N)	23	26	26	22	8	6	13	5	15
S/N	27.2	28.3	28.3	26.8	18.1	15.6	22.3	13.9	23.5
Level	Ta target current (A)		Substrate Bias (-V)		B ₄ C target voltage (V)				
1	Sum	Mean	Sum	Mean	Sum	Mean			
2	S/N	S/N	S/N	S/N	S/N	S/N			
3	85.1	28.4	76.4	25.4	56.8	18.9			
2	60.5	20.2	60.3	20.1	78.7	26.2			
3	59.8	19.9	68.6	22.9	69.8	23.3			

Calculated mean S/N values for Ta target current, substrate bias voltage and duty cycle (see Table 4) are plotted according to the deposition parameters of the graphs are given in the Fig. 12. According to Fig.12, the optimum deposition parameters for critical load of Ta-DLC films are 3A, -80V and %5 for Ta target current, substrate bias voltage and duty cycle, respectively.

Figure 12. Parameter level versus Mean S/N graphs of variable parameters for critical load values of Ta-DLC films

4. Conclusions

The results of Ta-DLC films deposited with CFUBMS-HiPIMS technique are as follows:

- Taguchi L_9 3^4 orthogonal experimental design was chosen to determine optimum parameters for critical loads. Ta target current (3A, 4A and 5A), substrate bias voltage (-80V, -90V and -100V) and duty cycle (%4, %5 and %6) were selected as variable parameters.
- The highest and lowest film thickness values were 1,318 μm and 158 nm, respectively. Film thickness increased with increasing Ta target current.
- The highest and lowest sp^3/sp^2 ratio were 0.527 and 0.471, respectively. The ratio sp^3/sp^2 is a measure of hardness. For this reason, the highest hardness was obtained from the film having the highest sp^3/sp^2 ratio (41.2 GPa).
- The critical load values of Ta-DLC films decreased with increasing hardness. Also, critical load decreases with increasing substrate bias voltage and target current.

- The optimum deposition parameters for critical load of Ta-DLC films are 3A, -80V and %5 for Ta target current, substrate bias voltage and duty cycle, respectively.

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