

## ORIGINAL ARTICLE

## Performance Study of Biocompatible Recast Layer Formation on Ti6Al4V by using Electrical Discharge Coatings

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**ABSTRACT** – Long-term implantation of titanium-based alloy, Ti6Al4V can be harmful in human bodies due to the release of aluminium and vanadium elements. Thus, a biocompatible barrier coating can be applied towards corrosion and wear resistance of the implant. In this research, the surface of a biomedical grade of Ti6Al4V was coated with a thin film of biomaterial ceramic by the electrical discharge coatings (EDC) using a pure graphite electrode. Polarity, discharge duration and pulse interval were varied to investigate the formation of recast layer thickness (RLT) on the surface of titanium alloys. RLT was measured from cross-sectioned samples using a high magnification optical microscopy. From the statistical analyses of variance, the response was significantly influenced by the pulse interval, followed with electrode polarity. Additionally, the interaction of polarity to discharge duration and pulse interval also significantly affect the RLT. In order to obtain a more uniform recast layer formation, the process condition should be in reverse polarity with a low setting of pulse interval.

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### INTRODUCTION

Titanium alloys are widely used in orthopaedics and dental applications because of their excellent biocompatibility. One of the most common titanium-based alloys is the Ti6Al4V. Ti6Al4V alloy is widely used for endoprosthesis application [1]. However, the alloy has a deficiency in term of poor wear resistance [2]. In body fluids, implants are significantly susceptible to wear and corrosion issues. Low wear and corrosion can result in the release of metal ions from the implants into the human body. The release ions cause inflammatory or allergic reactions which reduce the longevity of the implant and consequently, require revision surgery on the patient. In the case of Ti6Al4V, the release of vanadium and aluminium ions creates cytotoxicity that results in an allergic reaction. The ions also may cause Alzheimer disease and neuropathy [3]. To prevent these issues, biocompatibility of thin-film coating on the base material's surface is essential in reducing the number of released harmful ions. For this purpose, a biocompatible barrier coating of carbide alloy is to be developed on the surface of Ti6Al4V by using electrical discharge coatings (EDC). The carbide composition layer can effectively increase the wear resistance of the titanium alloy surface [4] and improve the osseointegration and proliferation processes in the human body that increases the biocompatibility of the implant [5].

Electrical discharge coatings (EDC) or also known as electrical discharge alloying (EDA) or electro-spark alloying (ESA) is a revolutionary technique from the conventional EDM process that creates a coating layer on a workpiece from a tool in dielectric fluids. From the working principle of the EDM process, the workpiece is melted, vaporized, and then cooled rapidly by the dielectric fluids. The tool is also eroded during the process. The material transfer from the tool and re-solidification of the molten workpiece material leads to the formation of recast layer on the substrate surface [6]. During the formation of the recast layer, the melted particles from the electrode tend to combine with the localized melt pools from the workpiece. This would develop craters on the workpiece prior to rapid cooling/quenching which forms alloying deposits. Multiple sparks also generate multiple overlapping of craters to form a continuum coating across the workpiece [7].

A survey of the literature indicated that a number of significant parameters on the EDC process on the surface modification. Amorim et al. [8] investigated the performance of the EDC process on the Ti6Al4V using a special grade of graphite electrodes based on the particles size for different regimes. The regimes were roughing, semi-finishing and finishing mode, which was based on the variation of discharge durations. They found that the highest recast layer thickness and hardness can be achieved from the roughing regime, which arises from the highest setting of discharge duration. Meanwhile, Chang-Bin et al. [9] studied on the formation of TiC layer on Ti6Al4V by graphite electrodes in air, nitrogen and silicon oil. They claimed that the EDC modified layer remarkably improved the hardness, wear and corrosion resistance of the surface layer of the substrate. From in vitro cytotoxicity tests, the substrates with the EDC modified layer showed no toxicity. Also, the change of cells shapes indicated it at growth states and influence for cell proliferation. Sindhu and Banwait [10] studied the effect of EDC parameters on EN-31 die steel by semi sintered with powder

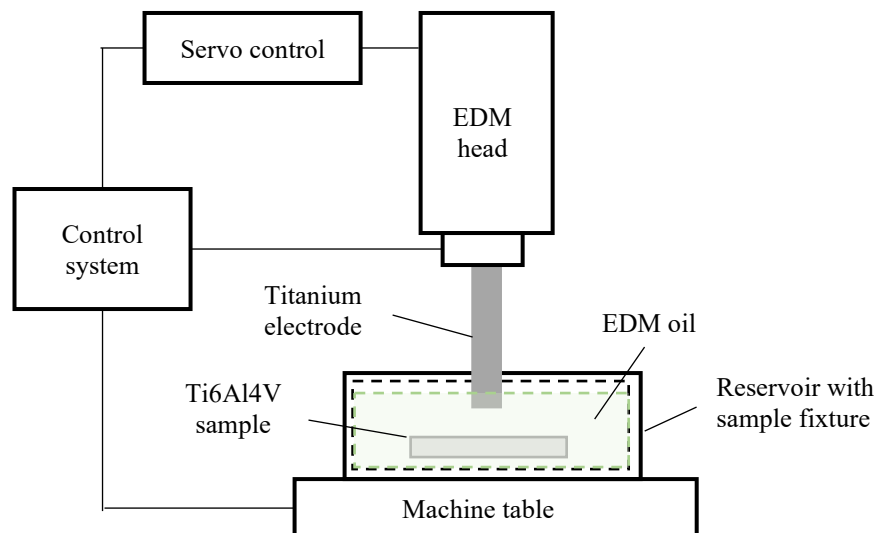
metallurgy electrodes. They concluded that the pulse interval influence significantly on the surface deposition rate (SDR) and roughness. The increase of pulse interval can lead to a decrease of SDR due to the less heat energy generated within the spark zone and consequently reduces the melting and evaporation of both electrodes. Apart from that, Gill and Kumar [11] studied the micro-hardness of the EDC layer by varying several parameters. From their results, the analyses indicated that micro-hardness was affected by the polarity of the electrode. They asserted that the positive of an electrode (reverse polarity) tends to develop a highly uniform alloyed layer on the surface of the substrate as to that of the negative polarity.

In this study, the Ti6Al4V samples were coated using EDC process by varying the parameters of polarity, discharge duration and pulse interval to evaluate the recast layer thickness and the uniformity of the adhesion from the topography images. The novelty of the research was the inclusion of polarity during EDC process on the Ti6Al4V alloys by the graphite electrode. Polarity has a high potential to influence the coating process. In addition, the use of hydrocarbon fluid (EDM oil) has the likelihood to increase the adhesion of the carbide alloy on the Ti6Al4V surface. Statistical analyses on the recast layer thickness (RLT) was carried out on the experimental data that was arranged in a full factorial experimental design.

## EXPERIMENTAL SETUP AND PROCEDURE

### Experimental Setup

EA8 Mitsubishi die sinker machine was utilised to machine the titanium alloy by a series of experimental trials based on different machine settings. The machine is capable of travelling in the area of  $300 \times 250 \times 250$  mm for the X, Y, Z axes respectively. It is equipped with internal circulation system with a liquid capacity of 250 L. During the experiment, a fabricated dielectric fluid reservoir with an internal dimension of  $150 \times 150 \times 40$  mm was used as to avoid flushing pressure from the circulation system. This is also to increase the material adhesion from the electrode. The reservoir was designed with a fixture to firmly hold the sample, as shown in Figure 1.



**Figure 1.** Schematic of the experimental setup.

### Material Preparation

Titanium alloys (Ti6Al4V) with a dimension of 50 mm in diameter were cut into 10 mm of thickness and ground using sandpaper of 500, 1000 and 2000 grit sizes. Then, the samples were polished with a misting of 6M monocrystalline diamond suspension until a mirror surface was produced. Then, the samples were etched using Kroll's reagent (6 ml of HNO<sub>3</sub>, 2 ml of HF, and 92 ml of distilled water). This method of material preparation was also applied in the metallurgical preparation of cross-sectioned samples to measure the RLT.

### Experimental Condition and Procedure

Graphite electrode of 99.99% purity and 10 mm in diameter was used to machine the Ti6Al4V with a depth of 0.2 mm. In this experiment, a jump function was activated to reduce the high accumulation of debris that could cause a short circuit. Initially, the sample was submerged approximately 10 mm in the dielectric fluids. The machining parameters; polarity, discharge duration and pulse interval were varied in the machine control panel, based on the experimental conditions tabulated in Table 1.

In this work, two levels of full factorial experiments with three parameters were specified to design the layout of the experiment. The centre point runs were replicated to obtain the experimental errors. Table 1 shows the machining conditions of the experimental work. Polarity, discharge duration and pulse interval were selected as the variable

parameters while other machining parameters such as the peak current, open gap voltage and jump up time were kept constant. The discharge voltage of the process was approximately 25 V.

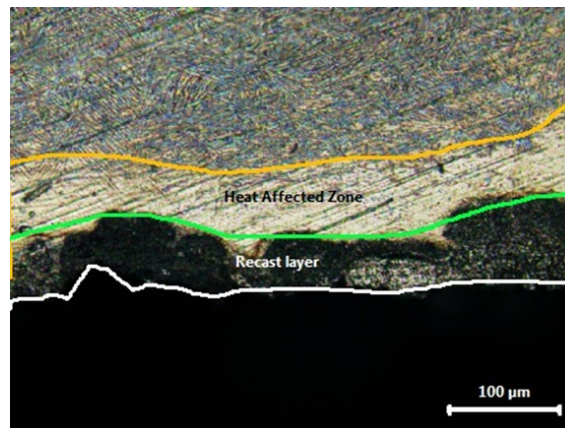
The samples were prepared for cross section observations by grinding, polishing and etching processes, similar to that of the previous material preparation. Xopton X80 series of high power optical microscopy (at maximum 1000× optical magnification) was used to observe and measure the thickness of the RLT. The chemical composition of the modified layer was examined using D2 PHASER X-Ray Diffraction (XRD) testing machine from Bruker Corporation. XRD test was implemented at a scan rate of 0.1 s/step for a 2θ range varying from 20° to 80°.

**Table 1.** The experimental conditions

Parameters	Unit	Conditions
Material		Ti6Al4V
Electrode		Pure Graphite 99.99%
Dielectric fluid		EDM oil
Polarity	-	Straight (-), reverse (+)
Discharge duration	μs	10, 480
Pulse interval	ms	3, 7
Peak current	amp	10
Open gap voltage	volt	30
Jump up	μs	3

## RESULTS AND DISCUSSION

Figure 2 shows a cross-sectioned sample under 1000× magnification that depicted a dark and white layer which can be seen clearly on the edge of the machined sample surface. This dark layer is the adhesion of alloying material from the electrode, workpiece and the dielectric fluids. In this study, the thickness of the layer that obtained is about 4 μm to 38 μm based on the parameters settings. Heat affected zone (HAZ) appeared on the upper border of the recast layer. The HAZ layer was formed due to the effect of heat that changes the grain structure from the original structure [12].



**Figure 2.** Microscopy observation of RLT from a cross-sectioned sample under 1000× magnification.

Figure 3 shows the XRD pattern of the phases on the surface of the workpiece before and after the EDC process. It can be seen that the coating layer was mainly composed by the titanium-vanadium carbide (CTi<sub>0.42</sub>V<sub>1.58</sub>) phase. The presence of titanium carbide (TiC) phase was indicated by the small peaks. In term of the biocompatibility, the high amount of vanadium element on the coating layer could be harmful to human bodies. Yet, the composition of CTi<sub>0.42</sub>V<sub>1.58</sub> consists of the vanadium carbide (VC) phase, which contributed towards the extremely hard and high wear-resistant layer.

The measurement results of recast layer thickness (RLT) based on the factorial design is shown in Table 2. From the ANOVA analysis, the significance of the model term is shown in Table 3 indicated an acceptable and significant of the experimental data. The most significant parameter which influences the RLT was the pulse interval with a contribution of 23% to the response, followed by the polarity with a contribution of 15%. Discharge duration was insignificant to the response and the trend was almost comparable for both levels of the settings. Thus, it can be assumed that the discharge duration mainly does not affect the response. The jump function acted to reduce the sparking energy that contributed by the parameter. Thus, less material removal rate on the workpiece and electrode resulted in a lower amount of material adhesion on the workpiece. However, the parameter has a remarkable effect when interacted with the polarity, which contributed up to 23% to the response. Another interaction was the pulse interval with the polarity of 19% contribution. These results indicated that the interaction between the parameters has a large impact on the formation of RLT.

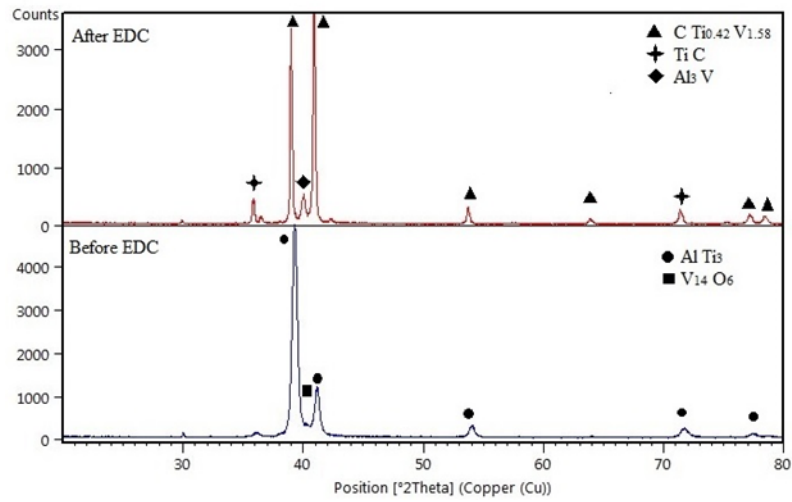


Figure 3. XRD profiles on a sample surface.

Table 2. Measurement results of recast layer thickness (RLT).

Std. no.	Polarity	Discharge duration (µs)	Pulse interval (ms)	RLT (µm)
1	straight	10	3	30.57
2	reverse	10	3	7.66
3	straight	480	3	21.21
4	reverse	480	3	14.27
5	straight	10	7	12.8
6	reverse	10	7	4.4
7	straight	480	7	5.86
8	reverse	480	7	16.06
9	straight	245	5	8.55
10	reverse	245	5	7.84
11	straight	245	5	9.10
12	reverse	245	5	5.38

The main effect of the pulse interval harms the RLT formation as shown in Figure 4(a). In theory, the increase of the level of settings can lead to the reduction of the duty cycle and discharge energy for the process. During high discharge energy, a large number of materials eroded either from the electrode or the workpiece. In the EDC process, a high erosion on the electrode rather than the workpiece is preferred. A high erosion of material from the workpiece can prevent the molten electrode from evenly adhered on the machined surface. Figure 5 shows this evidence based on the different topography images at a different level of pulse interval for the straight polarity. The light colour area exhibits the image of recast material from the workpiece. A more uniform coating surface can be achieved from the higher setting of pulse interval, see Figure 5(b). Although the RLT is lower than at a low level of pulse interval settings, for medical application, the uniformity of the coating is essential to the implant to be a barrier from the migration of harmful metal ions into the living tissues.

Table 3. ANOVA result of recast layer thickness.

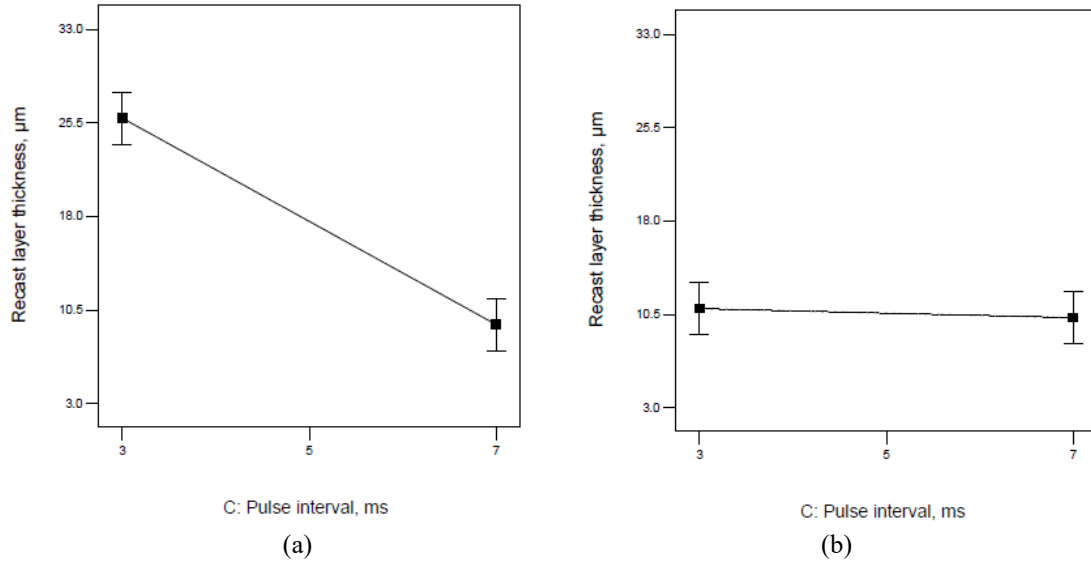
Source	Sum of squares	df	Mean square	F value	p-value Prob > F	
Model	512.6	5	102.52	37.22	0.0019	significant
A-Polarity	98.35	1	98.35	35.7	0.0039	
B-Discharge duration	0.49	1	0.49	0.18	0.6958	
C-Pulse interval	149.52	1	149.52	54.28	0.0018	
AB	149.35	1	149.35	54.22	0.0018	
AC	125.35	1	125.35	45.51	0.0025	
Curvature	124.21	2	62.1	22.55	0.0066	significant
Residual	11.02	4	2.75			
Lack of Fit	7.84	2	3.92	2.47	0.2883	not significant
Pure Error	3.18	2	1.59			
Cor. Total	647.83	11				

This result showed that the RLT was only affected by pulse interval under the straight polarity. RLT was almost not affected by pulse interval under the reverse polarity setting as shown in Figure 4(b). However, the coating was well uniform for the reverse polarity as to that of the straight polarity at pulse interval of 3 ms as shown in Figure 6, with

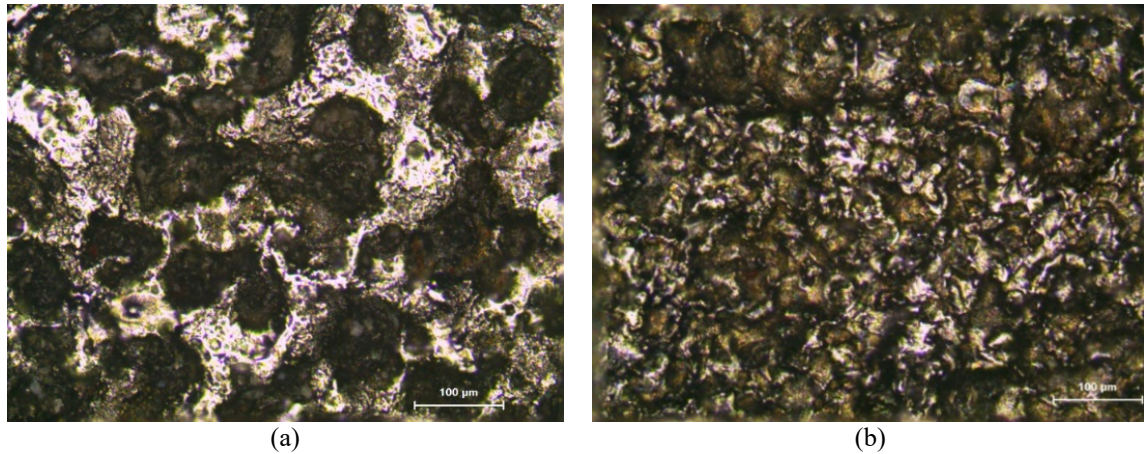


smaller RLT. At this condition, the migration of molten material from the graphite electrode to the workpiece surface was highly significant. Due to the high melting temperature ( $T_m$ ) of the graphite (approximately 5000 K) [13], high energy is required to melt this material. Since the reverse polarity was selected, the electrons tend to accelerate towards the electrode, which then collides to the electrode surface to increase the temperature before evaporation. Simultaneously, dissociation of carbon ions in EDM oil tends to adhere on the electrode surface and develop a hard layer of carbon.

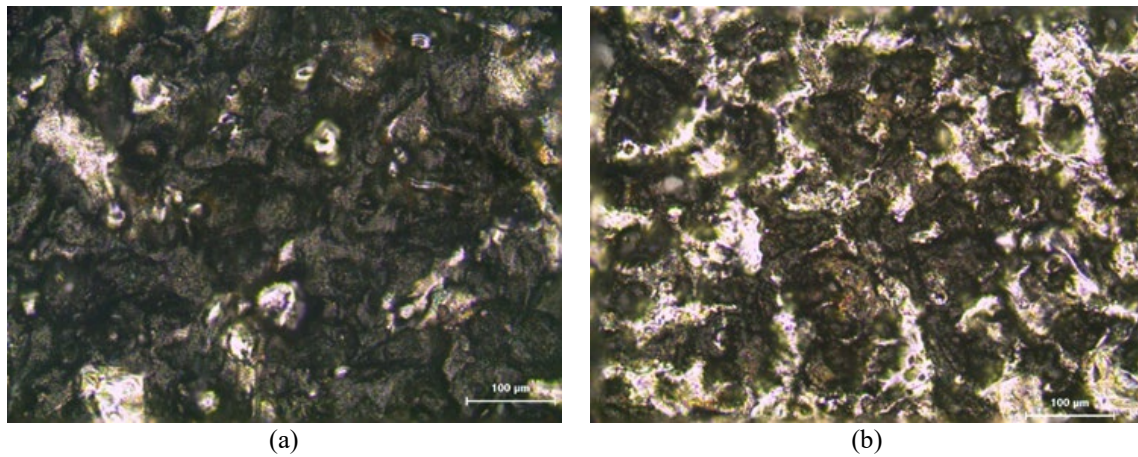
In comparison with the straight polarity, the negative polarity of the electrode was attacked with the positive ions which leads to a lower erosion. Then, the workpiece surface received a high sparking energy from electron acceleration, thereby more material was melted and evaporated before re-solidified on the workpiece surface. This phenomenon leads to an increase in the volume of workpiece's material and then re-solidified on the substrate. As a result, more recast material from the workpiece adhered on the substrate surface rather than the material from the electrode.



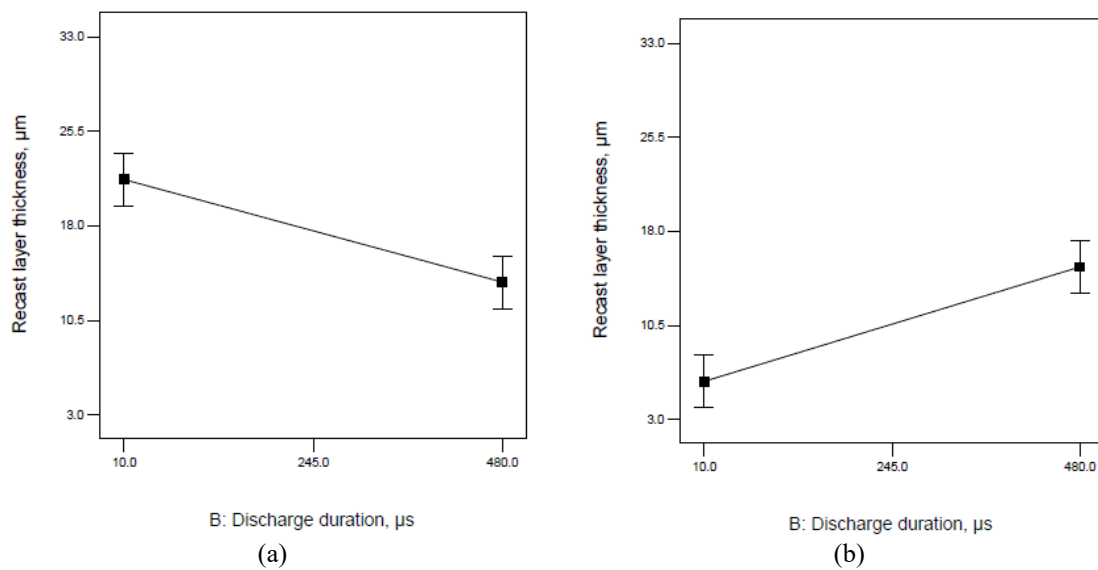
**Figure 4.** The effect of pulse interval on the thickness of recast layer with discharge duration of 245.5  $\mu\text{s}$  under (a) straight polarity and (b) reverse polarity.



**Figure 5.** The topography images of modified surface under 1000 $\times$  magnification with straight polarity and discharge duration of 245.5  $\mu\text{s}$ . At (a) pulse interval of 3 ms, RLT = 21.2131  $\mu\text{m}$  and (b) pulse interval of 7 ms, RLT = 5.858  $\mu\text{m}$ .



**Figure 6.** The topography images of modified surface under 1000× magnification with reverse polarity and discharge duration of 245.5 μs. At (a) pulse interval of 3 ms, RLT = 14.27 μm and (b) pulse interval of 7 ms, RLT = 16.06 μm.



**Figure 7.** The effect of pulse duration on the thickness of recast layer with pulse interval of 5 ms under (a) straight polarity and (b) reverse polarity.

In general, the discharge duration contributed only 0.08% towards the RLT formation. However, the parameter has significantly influenced the response when interacted with the polarity. Figure 7 shows a different trend of the RLT against the pulse duration under different electrode polarity. RLT tends to decrease until 8.1 μm with the increasing of discharge duration under the straight polarity. Conversely, an increasing trend of the RLT was evident when the reverse polarity was used. Theoretically, pulse duration controls the sparking period of the EDC process. Raising this parameter can increase the sparking energy in the spark gap that leads to significant material evaporation. Similar to that of pulse interval, erosion of the workpiece material was higher than that of the electrode under the straight polarity setting. As a result, the machined surface was covered up with most of the workpiece's material. This makes it difficult to observe a clear formation of the coating layer on the titanium alloys.

## CONCLUSION

This paper investigated the effect of machining parameters (polarity, discharge duration and pulse interval) of the EDC process towards the formation of recast layer coating on Ti6Al4V alloys. Pure graphite electrode was used to adhere to the carbide alloy on the surface of the substrate. From the EDC process, titanium-vanadium carbide (CTi0.42V1.58) was found as the main composition of the coating layer on the workpiece surface rather than the titanium carbide (TiC). Due to the presence of the vanadium element, further investigation should be implemented to test the biocompatibility of the coatings. For the statistical performance, the results indicated a significant effect of the pulse interval and the polarity towards the formation of RLT. Discharge duration only influenced the RLT when interacted with the polarity. The results of topography images indicated that a well uniformed coating can be achieved by reversing the polarity settings due to the significant erosion on the graphite electrode prior to the alloying of the Ti6Al4V surface. The straight polarity has a negative effect on the coating uniformity due to the high-volume erosion from the workpiece material.

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