Comparative Study on Aluminum Coating Methods on Ti-6Al-4V - Review

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Abstract— Titanium alloy is widely used especially in the aerospace industry. Though it has excellent favorable properties, it also possesses poor tribological properties. It is thus important to enhance its properties by various surface modification techniques. This review comprises of a comparison between the various methods of coating titanium alloy with aluminum. Seven different processes namely laser surface alloying process; plasma electrolytic oxidation process, thermal diffusion process, ion-plating process, vacuum evaporation process, magnetron sputtering process and dip coating process were evaluated and discussed. It was concluded here that dip coating is the most effective means of coating.

Index Terms— Coating methods, Ti-6Al-4V alloy, Aluminum coating, surface modification, research comparison

1 INTRODUCTION

Titanium and Ti-alloys have wide applications in aerospace, offshore engineering, automobiles and even biomedical applications. This is due to their high strength to weight ratio, low density and excellent corrosion resistance [1][2]. However their applications are limited due to its high affinity towards oxygen at high temperatures and its poor tribological properties such as high and unstable coefficient of friction, severe adhesive wear and a low resistance to plastic shearing [3]. Therefore it is necessary to enhance the surface performance of titanium or Ti-alloys by surface engineering methods so as to increase the oxidation resistance and also wear resistance.

Coating is found to one of the best surface engineering method to enhance the tribological properties of Ti-alloy. Coating is the process by which thin liquid layers are formed and applied to a solid surface [4]. This is done to enhance the surface properties like oxidation, wear, corrosion or to achieve optical, magnetic and electrical properties. Some factors that affect the choice of a coating are service environment, compatibility of the substrate material, life expectancy, shape and size of the component. Before coating, properties of the substrate should be analyzed and also the properties that are desired out of coating process must be determined.

There are several coating methods which include laser surface alloying process [1], plasma electrolytic oxidation process [5], thermal diffusion process [6], ion-plating process [7], vacuum evaporation process [8], magnetron sputtering process [8] and dip coating process [9]. It is important that through this method of coating we not only achieve the best adhesion strength but also achieve this through an economic and simple method. The present review is on a comparative study of the mentioned processes to conclude on the most effective means of coating. The various methods were studied with their advantages and disadvantages and conclusions were drawn as to which could be the best suggested coating method by all means.

2 COATING METHODS

2.1 Laser Surface Alloying

Laser surface alloying is the process of melting a pre-deposited layer or concomitantly added alloying elements/ compounds with a part of the underlying substrate by the directed energy laser beam to form an alloyed interface in a very short interaction time period. Laser surface alloying has been a research hot spot in the surface modification of titanium alloys and titanium aluminides for the enhancement of wear, corrosion and oxidation resistance. Some of the advantages of the coatings fabricated by laser surface alloying are dense microstructure, good metallurgical bonding with the substrate, controllable thickness. Moreover, different substrates can be alloyed with most materials. A wide range of solid solution, microstructure, and coating thickness can be achieved by altering the laser processing parameters.
Baogang Guo did research on coating of titanium substrate with Ti-Al coatings by the process of laser surface alloying [10]. He studied the inter-surface morphology, hardness change and the wear resistance after the coating process. Fig. 1 shows the XRD patterns of the titanium aluminides coatings. As can be seen from Fig. 1, the laser surface alloying coating with a thickness of 400 μm preplaced Al powder layer (denoted as Ti3Al coating) was composed of single Ti3Al inter-metallic phase. The laser surface alloying coating with a thickness of 800 μm preplaced Al powder layer (denoted as TiAl coating) was mainly composed of TiAl phase, although a little portion of Ti3Al phase was formed. The existence of

Ti3Al phase was beneficial to improve the ductility of single TiAl phase [11]. The laser surface alloying coating with a thickness of 1000 μm preplaced Al powder layer (denoted as TiAl3 coating) was mainly composed of TiAl3 phase, which was accompanied by a little TiAl2 phase and Al phase. It was difficult to obtain a single TiAl3 phase coating by laser surface alloying because of the very narrow composition range of TiAl3 [12][13].

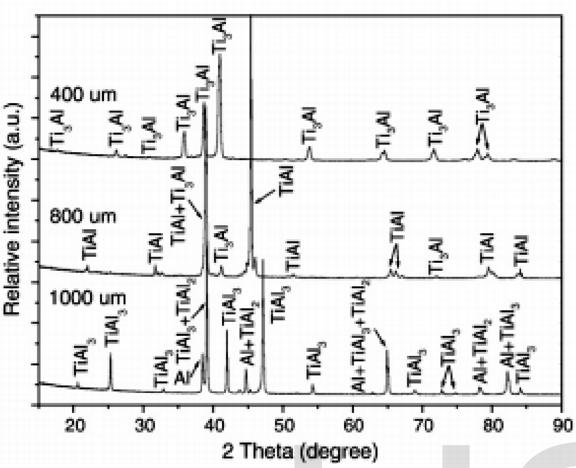

Fig 1. XRD patterns of the titanium aluminide coatings

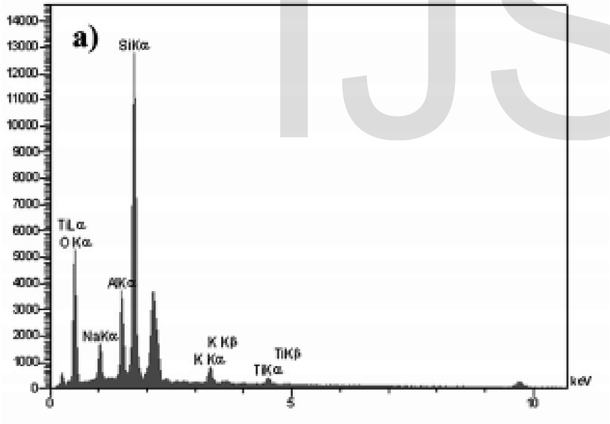

Fig 2a. EDS-SEM analysis of coated sample t_{10}

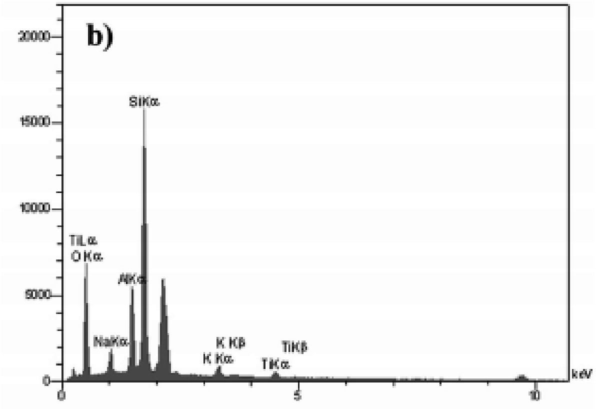

Fig 2b. EDS-SEM analysis of coated sample t_{20}

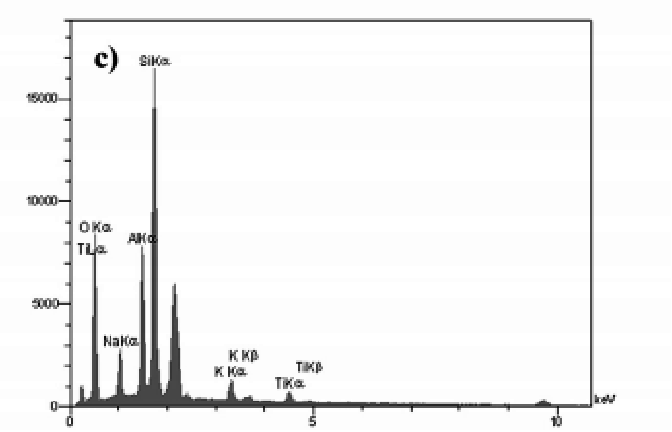

Fig 2c. EDS-SEM analysis of coated sample t_{30}

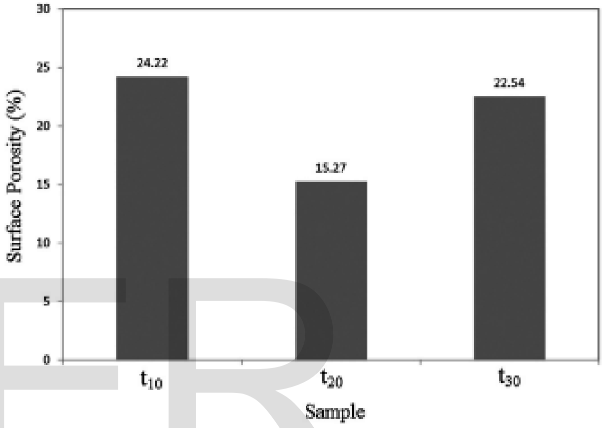

Fig 3. Surface porosity of samples t_{10}, t_{20}, t_{30}

2.2 Plasma Electrolytic Oxidation

Plasma electrolytic oxidation (PEO), is a new and attractive surface engineering method to create thick ceramic coating on the so-called valve metals such as aluminum, titanium, magnesium, zirconium, and other light metals as well as their alloys [14][15]. The PEO process is based on anodizing process with a high applied voltage and plasma discharge channels [16]. Some of the advantages of this method over the other surface treatments are single-step processing, excellent adhesion of coating to the substrate, environmentally friendly processing, and ease of controlling [17]. The composition, structure, and properties of coating produced by PEO process, depend on various parameters such as chemical composition and concentration of the electrolyte as the important ones.

In S. Sarbishei's work [5], alumina nano-particles were added to silicate based electrolyte to form alumina-silicate composite coating on titanium by the PEO process in order to modify the coating and to fill the porosities, resulting in enhanced corrosion resistance. The main objective of the present work is to discuss growth mechanism of the coating and probable reactions occurring during the process in a suspension containing alumina nano-particles. In the meantime, investigation of phase and chemical compositions, thickness, structure, and corrosion resistance of the coatings are the other attempts of this study. The EDS elemental analyses of the coatings with different processing time are shown in Figs. 2. As process time increases, the coating gets thicker, the micro arcs need higher energy and consequently porosities grow in size and decrease in number. Also slight micro cracks can be observed due to cooling effect of electrolyte and stress relief in the coating. As it can be seen in Fig. 3, there was an optimum processing time (20 min) to achieve minimum porosity.

2.3 Thermal Diffusion

Thermal diffusion process is one of the most widely used surface micro-alloying methods due to its economy and convenience. It could fabricate various surface alloyed coatings on the substrate via penetrating the alloying elements into the surface under the action of heat. The outstanding advantage of thermal diffusion is good metallurgical bonding between the surface alloyed coating and the substrate. The bonding strength between the alloyed coating and the substrate is high, and the alloyed coating is not easy to fall off from the substrate. The common thermal diffusion alloying elements used to improve high temperature oxidation resistance of titanium alloys and titanium aluminides include Al, Si, Cr, Nb, C, S and Mo.

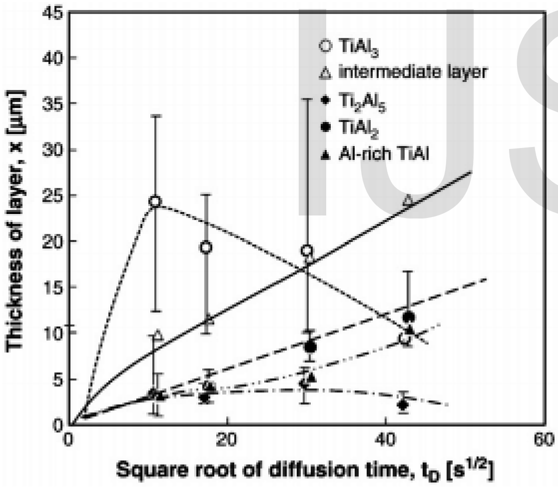

Fig 4. Changes in thickness of intermetallic layer, x, during diffusion process

In the study done by T. Sasaki [6], coating of aluminide on a TiAl-based alloy (49.1 at.% Al) was carried out by thermal spraying pure aluminum and subsequent diffusion treatment at 1100 °C. The growth of a Ti–Al inter-metallic layer in the coating layers as well as the oxidation resistance of the aluminized TiAl-based alloy was investigated. Fig 4 shows the changes in the thickness of the inter-metallic layers in the aluminized coating for the same diffusion times as for the cross-sectional observations. The total thickness of Ti2Al5, TiAl2, and Al-rich TiAl in the intermediate layer is also shown in the figure. The error bar indicates the variance of the thickness in the layer. Although TiAl3 shows a large variance in thickness caused by the rough surface, it grows rapidly in the initial stages of the diffusion treatment, up to 120 s. The thermal sprayed aluminum layer melts because of the diffusion treatment at a temperature higher than the melting point of aluminum. The rapid increase in thickness of TiAl3 in the initial stage indicates that the growth can be attributed to high diffusivity in liquid aluminum.

2.4 Ion-plating

Ion implantation is a surface engineering process by which ions are accelerated in an electrical field and impacted into the surface of materials. In comparison to other surface treatments, ion implantation has some outstanding advantages. Nearly all elements can be implanted into the surface of almost any materials, including metallic ions, such as Al, Nb, Mo, and Si, or non-metallic ions, such as C, Cl, F and I. Ion implantation is a low temperature process, which can avoid thermal defects caused by high temperature diffusion and distort of the work-piece. There is no adhesion problem since the implanted coatings exhibits metallurgical bonding with the substrate. The implantation depth and concentration of implantation ions can be well controlled and reproducible. Due to these unique advantages, the application field of ion implantation has expanded rapidly in the last decade. Numerous systematic studies have proved that ion implantation can improve high temperature oxidation resistance of titanium alloys and titanium aluminides for 100 h or more.

In the work reported by D. G. Teer and F. B. Saleem [7], aluminum was ion plated onto titanium and titanium alloys, over a wide range of plating parameters, and the coatings were tested in corrosive environments. During the course of this work it was found that, at the higher ion current densities, the aluminum diffused into the titanium to depths of up to 25 μm. Furthermore, the structure of the interfaces was studied by X-ray diffraction using a counter diffractometer. The X-ray patterns revealed that the interface consisted of two phases: a solid solution of aluminum in titanium and the inter-metallic phase TiA13. Pure titanium has a hexagonal structure with a c/a ratio of 1.587. The addition of aluminum in solid solution in the titanium caused a reduction in both a and c and also an increase in the c/a ratio, the value of c/a depending on the amount of aluminum in solution. The c/a ratio was related to the composition by etching the interface to a preselected depth, by measuring the composition of the new surface by microprobe analysis and by measuring the lattice parameters by X-ray diffractometer. The results are presented in Fig. 5. It can be seen that the c/a ratio is increased to a maximum value of 1.61 at 40 wt. % A1.

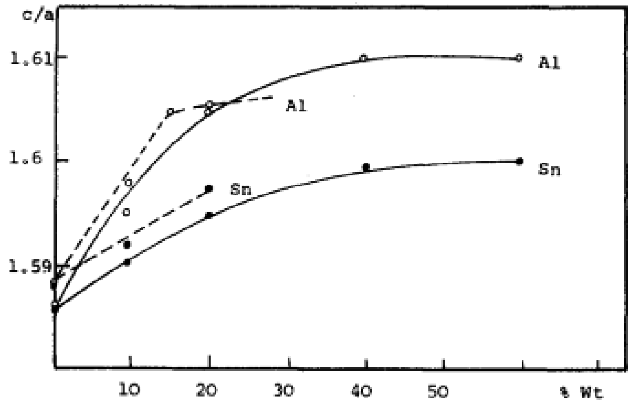

Fig 5. The effect of the percentage of film metals on the c/a ratio of titanium

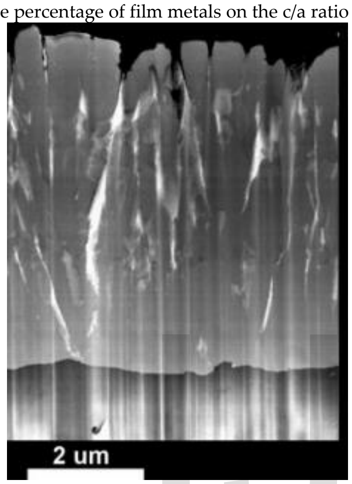

Fig 6. Microstructure of aluminum layer deposited by vacuum evaporation

2.5 Vacuum Evaporation

Vacuum evaporation is one of the most widely used methods for deposition of films on to different substrates. Vapor particles are directly deposited onto the substrate using the vacuum. The vapor particles then condense back to a solid state, forming a functional coating. The vacuum evaporation process takes place in two stages: the evaporation of the coating material and its condensation on the substrate. Vacuum evaporation technology (VET) is advantageous as it provides resistance to wear, corrosion, high temperatures, oxidation and radiation. It also enhances the conductivity, permeability and insulation properties of the substrates. Evaporation techniques have also been used for the metallization of polymer packaging films. The main purpose of the metallization of the packaging films is to isolate the product from the passage of light, oxygen or water vapor.

H. Garbacz performed research on vacuum evaporation technique by coating aluminum onto titanium alloy substrates [8]. The study of surface topography of the aluminum coating deposited by vacuum evaporation show that it is more developed; its microstructure is comparatively less homogeneous and more porous. It does not induce residual stresses in the coatings and the texture is very weak. This can be seen in Fig 6. which shows the microstructure observations of the coating. These pores, localized between the elongated grains, are formed due to the shrinkage of the deposited layer during the solidification process. The temperature of the titanium substrate in this process did not significantly increase, so there are no changes in the volume of substrate. As a result, the tensile stresses are generated in the layer during the cooling, which are sufficiently high to break weak diffusion/adhesion bonding between the columnar grains.

2.6 Magnetron Sputtering

Magnetron Sputtering is a Plasma Vapor Deposition (PVD) process where the positively charged ions from the plasma created are accelerated by an electrical field, which is superimposed on the negatively charged electrode also known as "target". The positive ions are accelerated by potentials ranging between hundred to thousand electron volts. It strikes the negative electrode with enough force so as to dislodge and eject atoms from the target. These atoms are ejected from the face of the target in a typical line-of-sight cosine distribution. It will then condense on surfaces that are placed near to the magnetron sputtering cathode.

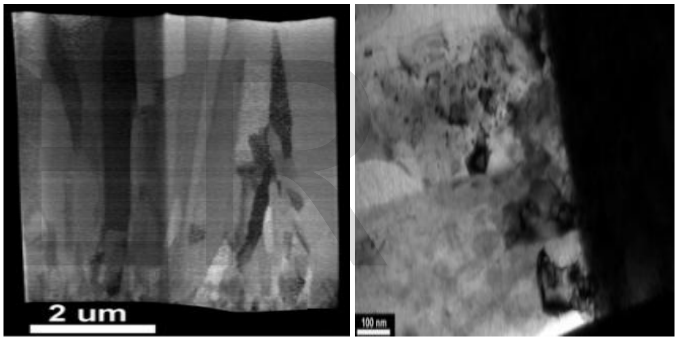

Fig 7. Microstructure of aluminum layer deposited by magnetron sputtering with a magnification of (a) 2µm(b) 100µm

H. Garbacz also performed research on magnetron sputtering process [8]. Microstructure observations showed that Al layers produced have a very refined microstructure with elongated nano-grains (Fig. 7a). It has been also found that, the layer obtained has a better quality, being more homogeneous, free of pores and cracks. However the Al layers obtained exhibit lower roughness. The microstructure of this layer consists of the elongated grains, 3–4 µm long and the 200–400 nm in diameter (Fig. 7a). Near the interface with the titanium substrate a small equiaxed grains of Al are observed (Fig. 7b). The size of these grains is in order of tens of nanometers. The grains become elongated at a longer distance from the interface. The presence of the very small equiaxed grains near the substrate can be explained by a rapid solidification of the sputtered Al during the deposition on the 'cold' titanium substrate.

2.7 Dip-Coating

Dip coating is process of depositing a wet liquid film by withdrawing the substrate from the liquid coating medium.

The complete process of film formation takes place in several stages, as shown in Fig. 9. The process starts by immersing the substrate in the solution of the coating material. When the substrate is withdrawn from the coating fluid, a coherent liquid film is formed on the surface of the substrate. A thin layer of coating is formed upon evaporations of solvents and any chemical reactions that are accompanied in the liquid film. Normally a post treatment such as curing or sintering is done to obtain the final coating. Dip coating technique is similar to sol–gel coating technique, although the process is faster and a complete transition can be achieved within a few seconds if volatile solvents are used [18]. Dip coating is fairly popular in various applications like the industry and in laboratory due to its low cost, simple processing steps and high coating quality.

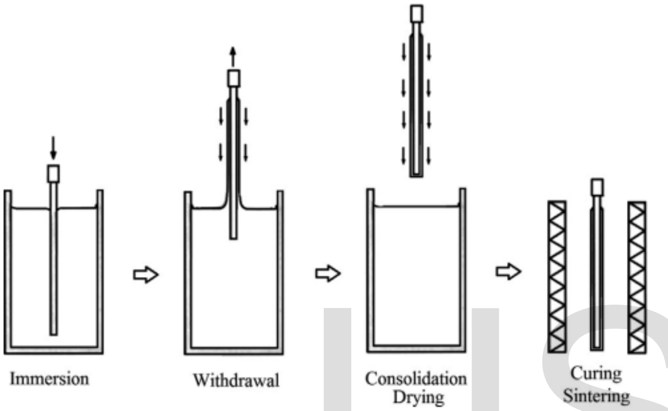

Fig 8. Fundamental stages of dip-coating (the finer arrows indicate the flow of air)

Various study have been reported on dip-coating of titanium alloy with aluminum. As by Wei Jiang, who in his study, prepared an aluminized coating on Ti-6Al-4V alloy by hot-dip aluminizing and subsequently diffusion treatment. We can see inter-metallic layer being formed at the interface between the two surfaces. This coating surface forms a protective layer against oxidation and wear of the titanium alloy surface. Figure 10 illustrates the micro-structural characteristics of the aluminized Ti-6Al-4V alloy. As shown in the XRD pattern, TiAl3 was identified to be the main phase of the aluminized coating. The cross-sectional morphology of the aluminized coating is shown in Fig. 10(b). The inter-metallic coating presented a fine microstructure and a flat, smooth interface with the substrate. The regional semi-quantitative analysis EDS1 (Fig. 10c) indicates that the atomic ratio of this aluminized coating was approximate to the stoichiometric ratio of TiAl3. The formation of TiAl3 coating could be attributed to the fact that Ti and Al atoms moved outward and inward through the elevated-temperature diffusion at 650 C. TiAl3 coating presented the micro-hardness of 500-585 HV.

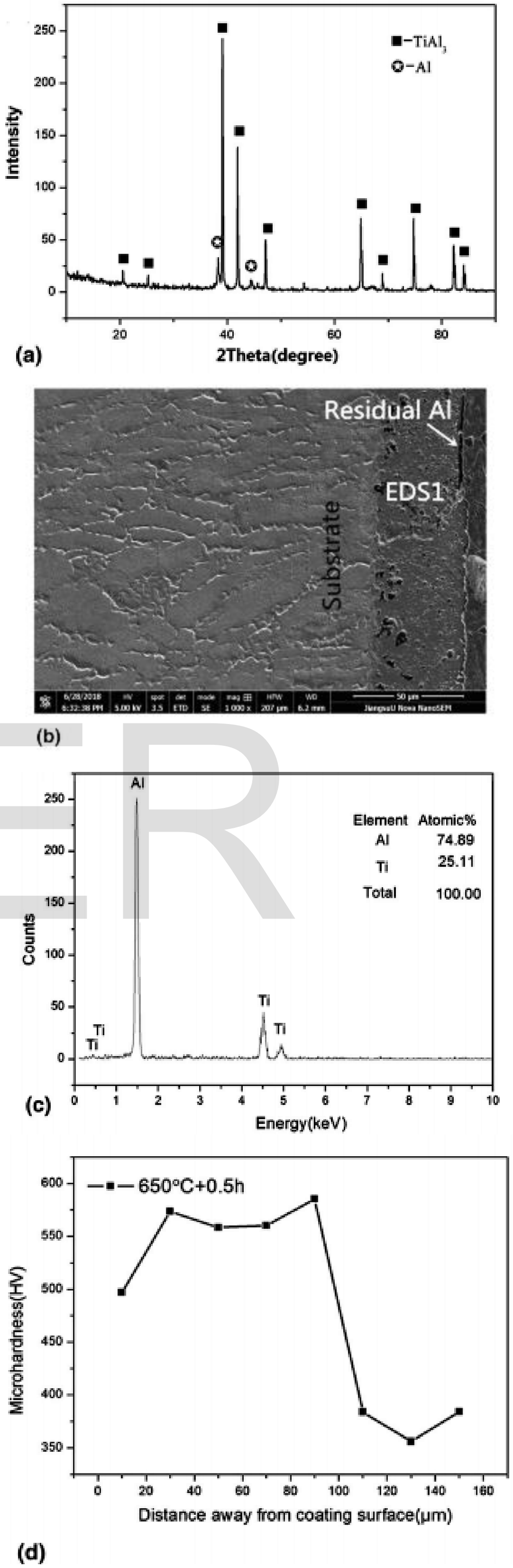

Fig 10. Micro-structural characteristics of the aluminized coating on Ti-6Al-4V alloy: (a) XRD pattern, (b) cross-sectional morphology, (c) EDS analysis, (d) micro-hardness distribution

3 DISCUSSION

COATING METHOD	SURFACE MORPHOLOGY	THICKNESS	ADVANTAGES	DISADVANTAGE
Laser Surface Alloying	TiAl,Ti3Al,TiAl3 layers	<1000 μm	High quality metallurgical bonding, controllable thickness, better hardness, wear resistance and oxidation resistance.	Pores and cracks were observed on the etched surfaces of TiAl coating and TiAl3 coating, hardness of coating in certain locations was even lower than that of pure Ti.
Plasma Electrolytic Oxidation	TiAl3 layers are formed. The surface is rough and hence the TiAl3 thickness varies.	<20 μm	Thick coatings can be achieved, single-step processing, excellent adhesion of coating to the substrate, environmentally friendly processing, and ease of controlling	Pores and micro crack were observed and stress relieving is required.
Thermal Diffusion	Ti2Al5, TiAl2, and TiAl3	50 to 150 μm	Good metallurgical bonding, high diffusivity, high deposition rate and economical.	The thermal sprayed layer contains some pores and oxides, lack of uniformity, crack formations, high temperature process leads to thermal defects.
Ion-plating	The X-ray patterns revealed that the interface consisted of two phases: a solid solution of aluminum in titanium and the intermetallic phase TiA13.	<25μm	Low temperature process, which can avoid thermal defects, metallurgical bonding, excellent corrosion resistance, high temperature oxidation resistance.	Increased variables to take into account when compared to other techniques, uniformity of plating not always consistent, excessive heating to the substrate, compressive stress.
Vacuum Evaporation	Ti-Al intermetallic Layers with higher porosity.	0.5-3 μm	resistance to wear, corrosion, high temperatures, oxidation and radiation, enhances the conductivity, permeability and insulation properties of the substrates.	Less homogeneous microstructure and more porous, weak texture and tensile stresses are generated in the layer during cooling, more porous.
Magnetron Sputtering	Ti-Al intermetallic Layers with uniform composition and lower porosity.	0.5-3 μm	layer obtained has a better quality, being more homogeneous, free of pores and cracks.	Stress measurements by X-ray show that the MS layer is under tension, low roughness
Dip coating	The inter-metallic coating TiAl3, TiAl and TiAl3 presented a fine micro structure and a flat, smooth interface.	<10-200 μm	low cost, simple processing steps and high coating quality, oxidation and wear resistance.	Light parts tend to float and film thickness can vary from top to bottom, fatty edges develop on the bottom of parts as excess coating drains, and the tank removes some of the coating.

The coatings fabricated by laser surface alloying are dense microstructure, good metallurgical bonding with the substrate, controllable thickness. The coatings obtained from laser surfacing method were free of pores and cracks and with high quality metallurgical bonding with the Titanium substrate. But the microstructure of the titanium aluminides coatings after etched in Kroll's solution resulted in pores. The coating resulted in better corrosion resistance.[19] PEO which gives single-step processing, excellent adhesion of coating to the substrate, environmentally friendly processing, and ease of controlling depends on the following analysis. The EDS elemental analysis of the coatings with different processing time is done. As process time increases, the coating gets thicker, the micro arcs need higher energy and consequently porosities grow in size and decrease in number. Also slight micro cracks can be observed due to cooling effect of electrolyte and stress relief in the coating. Metallurgical bonding between the surface alloyed coating and the substrate is developed. The bonding strength between the alloyed coating and the substrate is high [20].

In ion-plating the aluminium diffused into the titanium to depths of up to 25 μm at the higher ion current densities. The deep graded interfaces were formed only when the ion plating was performed at high power. The formation of the interfaces depended on the power. Aluminum was deposited onto titanium and then was heat treated at 600 °C little diffusion occurred. The TiAl3 in the interface simply hardens the interface region. The *c/a* ratio more nearly approaches the value for a perfect hcp structure and as it is known that h.c.p metals have better friction and wear characteristics. The wear rate is extremely low [21], [22].

Similarly microstructure observations of the coatings showed that Al layers produced by vacuum evaporation and magnetron sputtering have a very refined microstructure with elongated nano-grains. It has been also found that the layer obtained by magnetron sputtering has a better quality, being more homogeneous, free of pores and cracks. In the case of layer deposited by vacuum evaporation, a large amount of pores was observed. The high surface roughness of the Al layer is therefore disadvantage in this case of inter-metallic coatings deposited by duplex method. The Al layers obtained by magnetron sputtering method exhibits lower roughness. Stress measurements by X-ray show that the magnetron sputtering layer is under tension, whereas the vacuum evaporation does not reveal residual stresses [23], [24], [25].

Dip coating process forms inter-metallic layers in the order of TiAl3/TiAl2/TiAl/Ti3Al. the layers are found to be uniform and increases the hardness of the coating material. It forms a protective layer against oxidation, wear and corrosion. Due to the formation of the inter-metallic aluminide layers the adhesion is found to be good. The protection of the coating layer can be extended at high temperatures upto 800˚C.[26], [27], [28]

4 CONCLUSION

The various coating methods to obtain aluminum coating on titanium alloy substrate have been reviewed. Seven commonly used processes such as laser surface alloying process, plasma electrolytic oxidation process, thermal diffusion process, ion-plating process, vacuum evaporation process, magnetron sputtering process and dip coating process have been studied and discussed. The microstructure of the coatings achieved by the above mentioned methods were evaluated. We can conclude that

- Comparing all parameters in the coating process including cost, set-up construction, bonding strength of the coating, microstructure analysis of the coating, extent of property enhancement, we can say that dip-coating is found to be the most effective means of coating.
- Dip-coating is a low cost process. The set-up for the procedure is simple and can be easily customized according to the substrate shape and size. Also there is minimal loss of material during this process.
- The bonding strength is found to be comparatively good considering the diffusion treatment and formation of aluminide layers. The coating forms a protective layer against oxidation at high temperatures up to even 800˚C. It also protects the substrate against wear during sliding.
- The coating thickness can be controlled by certain process parameters such as dipping time, withdrawal rate, etc.

Thus, we can conclude that dip-coating process is the most effective way of obtaining aluminized coating on titanium alloy. Further study is being carried out on the same to achieve a more controlled and functional coating.

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