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RESEARCH ARTICLE

Investigation of nanoporosities fabricated on metallic glass surface by hydroxyapatite mixed EDM for orthopedic application

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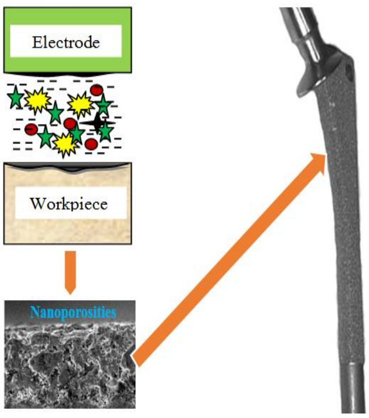
Graphical abstract



Abstract

Bulk metallic glasses (BMGs) have exceptional biomechanical characteristics like low elastic modulus, outstanding fracture strength, superior wear and corrosion resistance compared to routinely used biomaterials. The major downside of BMG is its inability to osteointegrate to the surrounding living tissues. To solve this problem, a biocompatible and bone-like nanoporous layer is normally imparted on the implant surface. In this study, a very hard, biocompatible and nano-porous layer was deposited on the Zr-based metallic glass surface, by hydroxyapatite mixed electrical discharge machining (HA-EDM). A 2-level factorial design and 3 factors was designed using design expert 10.0 software. The experiment was carried out using a die-sinker EDM machine. FESEM was employed to observe the pore distribution, geometry, and sizes. The result reveals the formation of rough, narrow craters and interconnected nanoporosities in the range of 558.2 nm to 893 nm in diameter and surface area of 244764 nm^2 to 626596 nm^2. However, the XRD and EDX characterization revealed the deposition of ZrC, TiC and $CaTiO_3$ on the HA-EDMed surface. The surface produced by HA-EDM are expected to facilitate higher tissue ingrowth and bone-implant adhesion.

Keywords: HA-EDM, metallic glass, nanoporosities, orthopedic implant

INTRODUCTION

The long-term stability of orthopedic implant depends on several factors such as the implants material, manufacturing, design and the surface modification techniques. Biomaterials such as titanium and its alloys, cobalt chromium and stainless steel are the most frequently used orthopedic implant materials(Odekerken, Welting, Arts, Walenkamp, & Emans, 2013). Despite the higher strength of the metal based implant compared to the natural bone, most of the implants failed at early stage of implantation (less than 15years). This period is comparatively shorter than the life span of human (even the elderly person with a life expectancy of 17.9 years) (Chen & Thouas, 2015). Thus, the patient most undergoes a revision surgery, which is not normally recommended due the complexity of the process (Ashkanfar, Langton, & Joyce, 2017; DeFrances, Lucas, Buie, & Golosinskiy, 2008). The potential causes of this implant pre-mature failure may be due to several reasons such as incompatible mechanical properties of the implant or fixation device, the development of undesired osteointegration between living bone and the synthetic surface, poor implant surface finishing as well as poor corrosion and wear resistance (Ashkanfar et al., 2017; Bahraminasab et al., 2012; Martin & Trousdale, 2013). Fig. 1 shows the hip anatomy, components of the total hip replacement and the merged hip implant fits into the body.

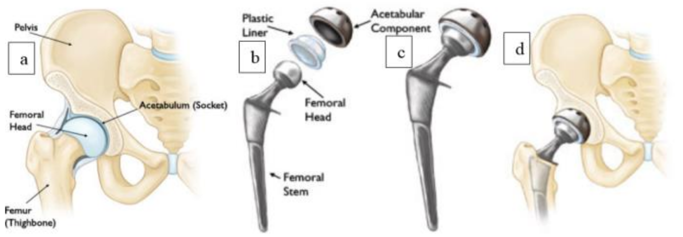


Fig. 1 (a) Anatomy of normal hip (b) components of total hip replacement (c) Merged component (implant) (d) Implant fits into the body

The number of hip or knee revision surgery is rising every year as depicted in Fig. 2. Kurtz, Ong, Lau, Mowat, and Halpern (2007) projected that the cases of total hip and knee revision surgery will rise with over 137% by 2030 in United State. Fig. 3 depicts a normal revised hip and the loosen screw after revision surgery.

Bulk metallic glass (BMG) is non-crystalline alloys also called amorphous alloys (liquid metal), which have exceptional properties when compared with crystalline solids. BMGs are superior than the routinely used biomaterials (titanium alloys, cobalt-chromium and stainless steel) in terms of strength, modulus of elasticity, wear and corrosion resistance as well as fatigue endurance (W.-H. Wang, Dong, & Shek, 2004). Thus, BMGs are continuously gaining attention by several researchers, especially in the biomedical area. Fig. 4 compares the mechanical properties of cortical bone with biomedical materials. Based on the above BMG potentialities. It is highly expected that, if the challenges of the Zr-based BMG could be addressed, by enhancing its surface bioactivity and improved its current size, this material could replace the currently used biomaterials in terms of applications as summarized in Table 1.

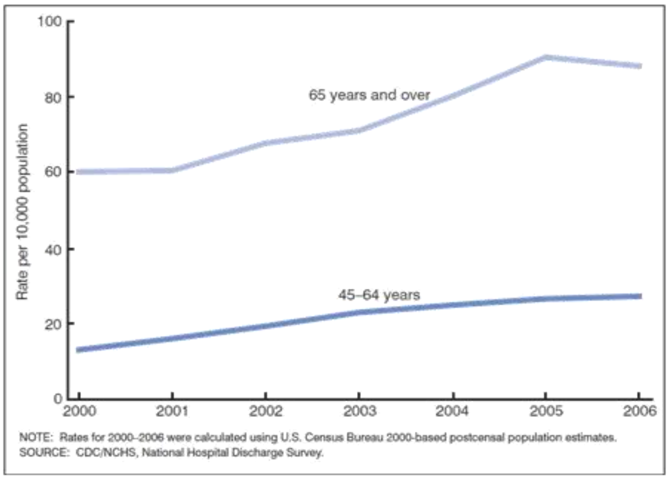


Fig. 2 rate of knee replacements in United States for hospital inpatients of 45 and above years old (2000 - 2006) (DeFrances et al., 2008)

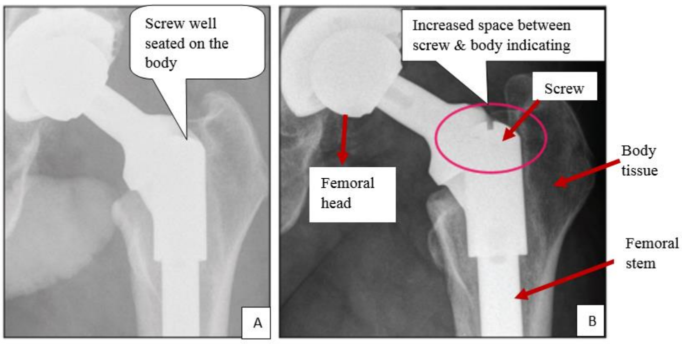


Fig. 3 Screw seated in the proximal body after revision surgery (A) A clear space between the screw and the proximal body, indicating loosening, 11 months after the index revision surgery (B) (Martin & Trousdale, 2013)

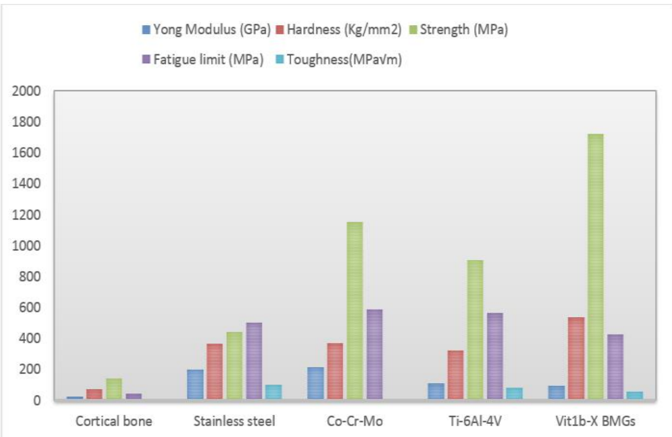


Fig. 4 Comparison of mechanical properties of the cortical bone and the most commonly used metallic biomaterials (Abdul'Azeez Abdu Aliyu et al., 2017)

Table 1 Routinely used metallic biomaterials and their primary applications. (Abdul'Azeez Abdu Aliyu et al., 2017)

Routinely used biomaterials	Applications
Stainless steel	Orthopedic: For total hip replacement and production of temporary devices such as plates, screws, pins, nails etc.
Co-based	Orthopedic: For total joint replacement Dentistry castings
Ti-based	Orthopedic: For stem and cup total hip replacement, production of permanent devices such as nails, face makers Dentistry: Dental screws (permanent implant)

Despite the continuous emergence of new biomedical materials, there are still unsolved issues. For instance, BMG have a bio-inert oxide layer, which is characterized by low surface hardness and poor wear resistance. Thus, this layer does not promote strong physical bonding with the living tissues. Several authors considered coating the implant surface with a biocompatible, bioactive and nanoporous layers through several kinds of coating methods as a primary solution to this problems (Odekerken et al., 2013; Stojanovic et al., 2007). Calcium phosphate (CaP) containing compounds, such as hydroxyapatite (HA) which contain the main inorganic component of the bone were adopted as coating material by many researchers (Asri, Harun, Hassan, Ghani, & Buyong, 2016; L.-N. Wang & Luo, 2011; Xu et al., 2016; Y. Zhang et al., 2014). CaP/HA surface coating increases the biocompatibility and provides a very tight bonding between the implant and the surrounding tissues (Asri et al., 2016; Dorozhkin, 2015). In addition, porous layer is believed to have a reasonable and adjustable modulus of elasticity(Prakash, Kansal, Pabla, & Puri, 2015; L. Zhang, He, Zhang, Jiang, & Zhou, 2016). Among the hydroxyapatite deposition techniques, plasma spray is the most common and commercially available. However, this method is expensive, produces surface cracks and require very high temperature, which might change the initial performance of the substrate material. Powder mixed electric discharge machining (PMEDM) is an emerging technology which expected to acts as a machining and surface modification technique.

The use of EDM in mold, tools, automobiles and aerospace industries have been largely documented (Amorim & Weingaertner, 2004; Cabanes, Portillo, Marcos, & Sánchez, 2008). Lately, the concept of material transfer from the tool and the dielectric additives to the workpiece surface during powder mixed EDM (PMEDM) process was reported by several authors (A. A. Aliyu, Hamidon, & Rohani, 2014; A. A. A. Aliyu, Rohani, Rani, & Musa, 2017; Batish, Bhattacharya, Singla, & Singh, 2012; Liew, Yan, & Kuriyagawa, 2013; Saxena, Agarwal, & Khare, 2016). Fig. 5 demonstrate the mechanism of material migration at machining zone during PMEDM of Zr-based BMG.

Higher elestic modulus of most biomaterials compared to that of bone is considered the major reason for implant loosening which leds to revison surgery. Porous metallic materials have large specific area, good ductility, low density and controlled modulus of elesticity. Although, the use of porous materials to replace hard tissues have been largely documented, processing of these material to achieve bone match pore size (20 – 59 % volume fraction) is quite difficult (Zou, Yu, Li, Guo, & Li, 2016). This study aimed at fabricating a bone-like nanoporosities and depositing a hard corrosive and wear resistive film machined Zr-based BMG surface using hydroxyapatite mixed electrical discharge machining (HA-EDM).BMG surface. SEM and XRD characterization techniques were used to exermine the HA-EDMed BMG surface. An extremely hard surface, with dense interconnected nano pores were fabricated. The EDX patterns shows the deposition of minute amount of calcium and potassium on the machined surface. The presence of some carbide layers were also noticed on the XRD pattern.

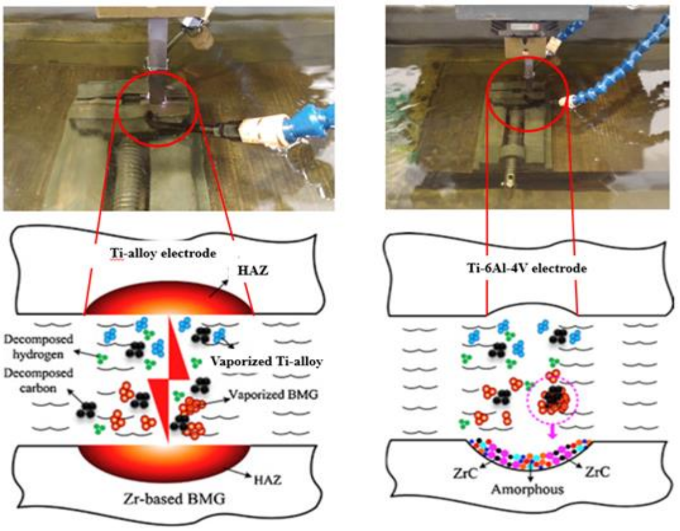


Fig. 5 PMEDM gap (a) discharge time: melting and vaporization of workpiece and tool materials (b) off-time: deposition of the carbide layer on the workpiece surface.

EXPERIMENTAL

Materials and method

A commercial Zirconium based BMG was choosen to be the workpiece material. The received as-cast BMG (100 x 110 x 5 mm) was cut into pieces (9 x 11 x 5 mm) using wire-cut EDM machine. 30 micron hydroxyapatite (HA) powder was adopted to be the additive of the EDM dielectric fluid. However, pure titanium is considered as the electrode material. The weight of both the tool and the workpiece eletrodes were measured before and after the experiment. 5g/L of hydroxyapatite powder concentration was mixed with the hydrocarbon EDM oil in the fabricated 38.5L capacity PMEDM main tank. HA mixed EDM oil was circulated and convayed to fabricated metallic machining tank (Fig. 6). The BMG was machined using the die sinker EDM machine and titanium electrode.

A 2 level experimentel design and 3 factors (discharge current (Dc), discharge time (Dt), and Electrode polarity (Ep)) gives a total of 8 experimental runs was carried out. Machning parameters and levels were summarized in table 2. The constant parametrs used were given in table 3. The HA-EDMed BMG surface was examined using Scanning Electron Microscopy (SEM). Fig. 7 and Fig. 8 shows the SEM micrograph and EDX of the BMG (as received) respectively. Fig. 9 depicts the XRD pattern of the Zr-based BMG (as-cast). The absence of sharp peaks and the presence high bump between 30 to 50 position, justify the amophours nature of the received BMG.

Table 2 Machining conditions and respective levels

Machining conditions	Levels	
	1	2
Discharge current (A)	8	12
Discharge time (μs)	4	16
Electrode polarity	+	-

Table 3 Constant machining conditions

Conditions	Unit	Descriptions
Gap voltage (V)	V	80
Powder concentration	g/L	5
Workpiece	mm	Plate: 5 x 10 x 11
Electrode (tool) type	-	Rectangular: Pure Ti
Dielectric fluid type	-	Hydrocarbon oil
Flushing type		Fluid emission
Powder type	-	Hydroxyapatite
Powder size	μm	30
Machining depth	mm	0.3

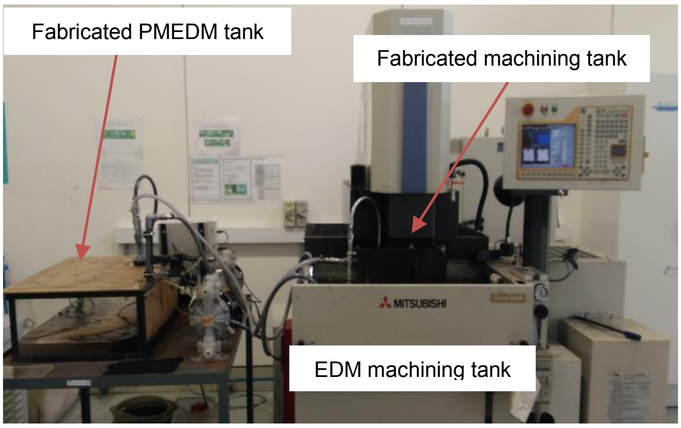


Fig. 6 Fabricated PMEDM sysem

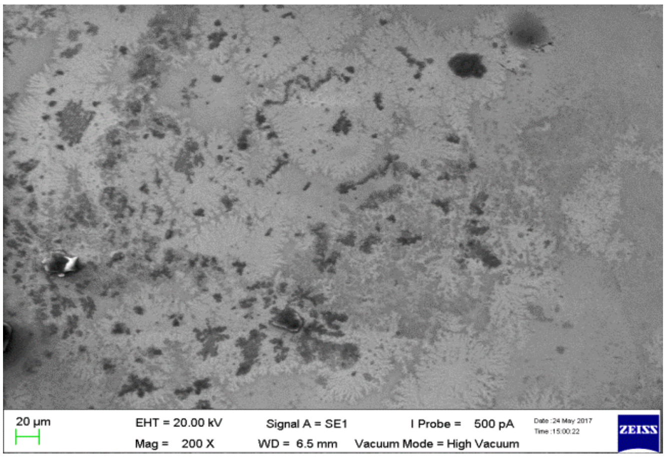


Fig. 7 SEM micrograph of the Zr-based BMG (as-received)

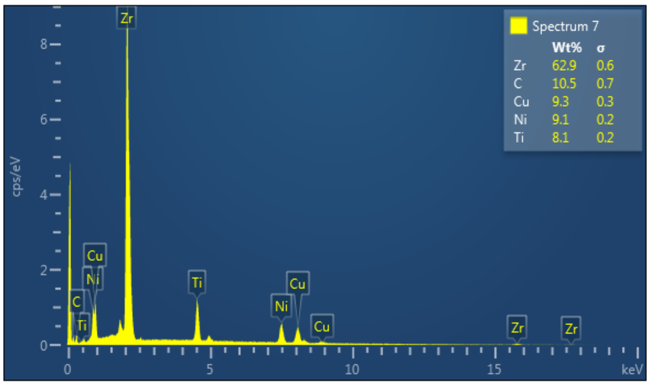


Fig. 8 EDX pattern of the Zr-based BMG (as-received)

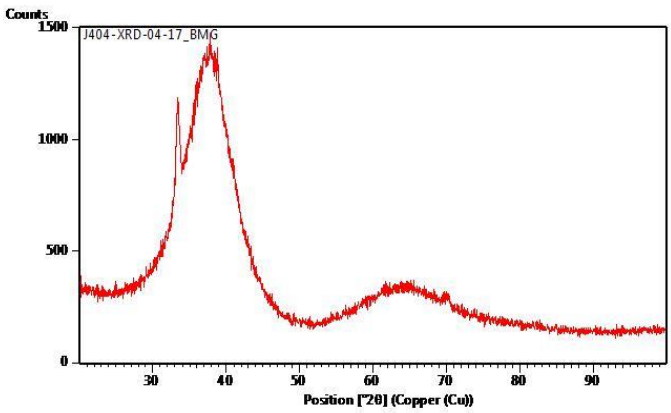


Fig. 9 XRD pattern of the Zr-based BMG (as-received)

RESULTS AND DISCUSSION

Nanoporosities

SEM migrographs of EDMed BMG and HA-EDMed BMG are depicted in Fig. 10. Numerous interconnected micro, submicro and nano pores can be observed in both EDMed and HA-EDMed BMGs. However, the pores in the EDMed BMG (Fig. 10a) are scanty compared with HA-EDMed (Fig. 10b). This may be due hydroxyapatite powder added to the dielectric fluid and the low energy (Dc and Dt) used. The added powder stabilizes the EDM process by increasing the discharge gap and insulating the dielectric strength. In addition to porosities, a deep and wide craters could be noticed on the EDMed surface, while the craters are narrow, interconnected and shallow in the HA-EDMed surface. A pore size of 558.2 nm to 893 nm in diameter could be observed in the highly magnified macrograph of both EDMed and HA-EDMed BMG surface (Fig. 10c and 10d). This indicates that, the addition of HA powder does not have much influence on the pore size. However, the pore surface area ranged between 244764 nm^2 to 626596 nm^2 were observed.

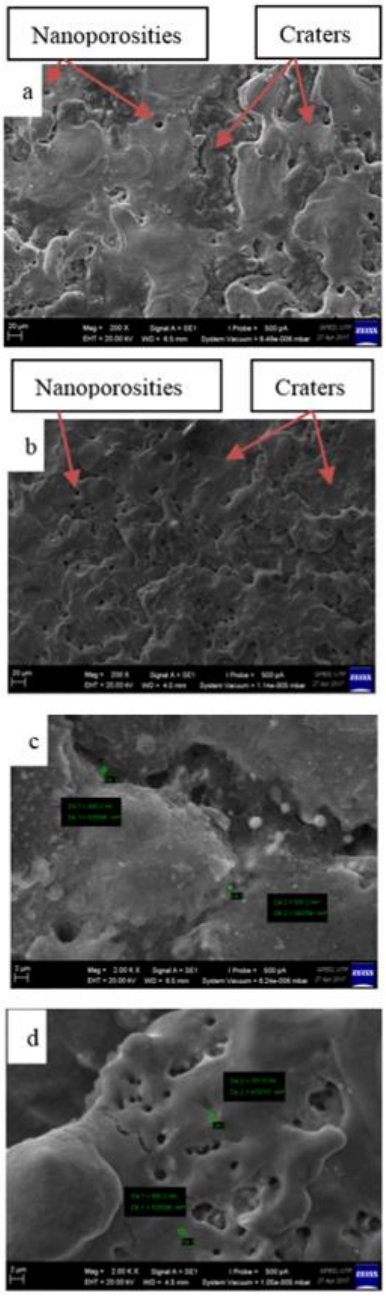


Fig. 10 FESEM micrograph of (a) EDMed BMG (Dc = 12A, Dt = 16μs, Pc = 0) (b) HA-EDMed BMG (Dc = 8.0A, Dt = 4.0μs, Pc = 5) (c) pore size of EDMed BMG (d) pore size of HA-EDMed BMG (e) sectional view of HA-EDMed BMG.

Surface Analysis

The formation of different layers can be seen in Fig. 11. The topmost layer is the extremely hard carbide layer, which is formed due to the reaction of carbon in the dielectric hydrocarbon fluid and the workpiece alloying elements. The recast and oxide film constituted the middle layers. The recast layer is produced because of re-solidification of unflushed metallic debris in the craters (Kruth, Stevens, Froyen, & Lauwers, 1995). The last layer is the heat affected zone, (heated but non-melted layer). This layer is formed due to heating and subsequent quenching during the EDM process. A thin recast layer is achieved due to the HA powder and hydrocarbon dielectric fluid used. Despite that hard and adhesive recast layer enhanced the corrosion and wear resistance of the metallic implant, a thick recast layer should be avoided or polished, to reduce the risk of the implant mechanical failure (Huang et al., 2015). The thinner and porous recast layer as well as finer and harder surface acheiched is similar to result reported by Kumar, Kumar, and Kumar (2016).

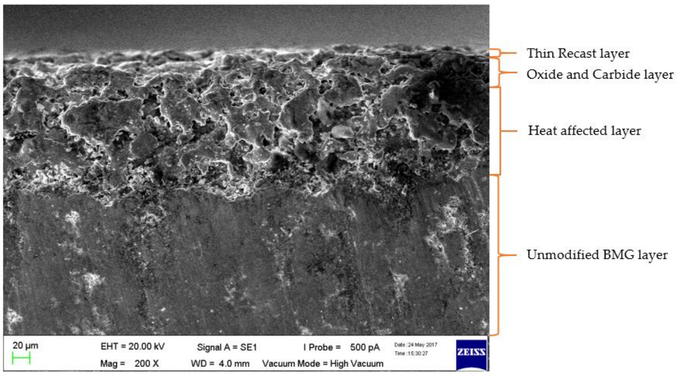


Fig. 11 Cross-sectional view of the H-EDMed BMG showing different layers.

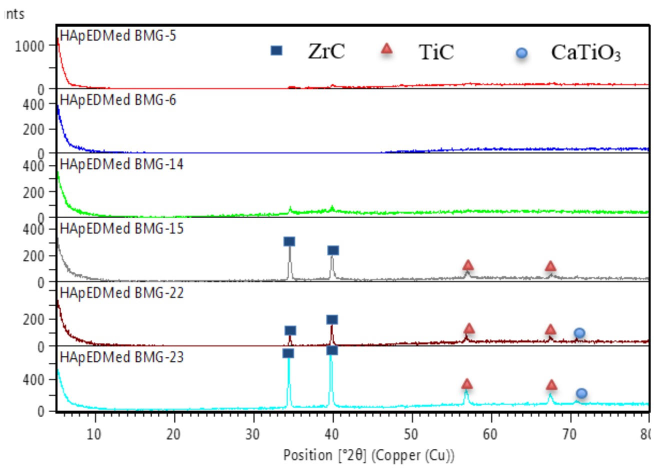


Fig. 12 XRD patterns of HA-EDMed BMG using different EDM parameters setting.

The XRD pattern presented in Fig. 12 depicts the presence of zirconium carbide (ZrC), titanium carbide (TiC) and calcium phosphate (CaTiO3). The carbide produce is due to the reaction of carbon to the zirconium which is the main alloying element of the workpiece material and the titanium (electrode material). It is reported by many authors that carbide layer generated by EDM provide not only strength but also strong wear and corrosion resistance to the EDMed surface (Prakash, Kansal, Pabla, & Puri, 2016; Prakash, Kansal, Pabla, Puri, & Aggarwal, 2015; Zou et al., 2016). As dipicted in Fig. 12, HA-EDMed samples 5, 6 and 14 were machined using negetive tool polarity while 15, 22 and 23 using positive tool polarity. Sharp and high peaks of ZiC (at 35.55^0 and 40.66^0) and TiC (at 57.50^0 and 67.75^0) could be observed on the XRD patterns of the samples machnined using negetive tool polarity. The long and sharp peaks indicate a high crystallization of BMG and the deposition of carbon on the machined surface. In addition, prolonged heating due to negative polarity and high energy enhanced by the discharge time might contribute to the formation of crystalline ZrC and TiC. However, a small bump for $CaTiO_3$ could be noticed at 71.00^0. This is due to reaction of calcium in the HA powder to the workpiece and the electrode elemnetal composition. On the other hand, in the XRD of HA-EDMed BMG 5, 6 and 14, shows that the amorphours nature of the BMG is highly maintained. This confirmed the suitablity of positive tool polarity to machine BMG without much effect on its initial crystal sturucture.

Fig. 13 depicts the Energy-dispersive X-ray spectroscopy (EDX) of the HA-EDMed sample. About 1.5% by weight of calcium and 0.7% of potassium could be observed. This indicate the ability of HA-EDM process to deposit some powder mixed in the deielectric fludid, under suitable parameters setting.

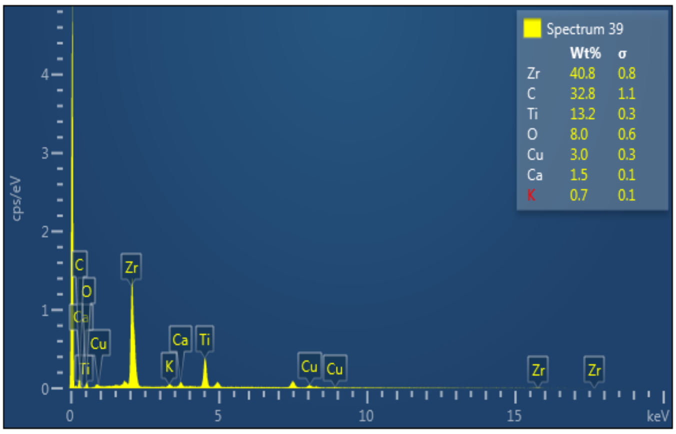


Fig. 13 EDX pattern of HA-EDMed pattern

CONCLUSION

It can be concluded that EDM machine is suitable for not only fabricating implant but also imparting a nanoporous hard carbide and catium titanium oxide on the implant surface. However, the addition of hydroxyapatite powder facilitates the formation of the nanopores and shallow craters. A low discharge current of 8.0A and discharge time of 4.0µs were found to have a great contribution to the formation of interconnected nanoporosities. A nanopores of about 558.2 nm to 893 nm diameter and 244764 nm^2 to 626596 nm^2 surface area were fabricated on the Zr-based BMG surface. The craters produced were larger and wider on the EDMed BMG surface compared to HA-EDMed surface. It is believed that nonoporosities and narrow craters implant surface fabricated by HA-EDM possses greater bone-implant adhesion, higher strength bonding and full tissue interlocking when compared with conventionally fabricated EDM surface.

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