

## Review of diamond-like carbon films in medical applications

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### Abstract

A review on the diamond like carbon (DLC) film as coating material for biomedical application, nowadays DLC as coating material is coming larger in biomedical application with variation of time because of its properties and biocompatibility, when we go back in history we noticed that the first report on the hard carbon films were published in the 1950s and nearly 20 years later then began worldwide doing a lot of researches activity on diamond like carbon and nowadays it is more and more for the therapeutic and diagnostic application in biomedical field. DLC films has a lot of strengths which is hemocompatibility, low friction, and low roughness, good adhesion, stability, corrosion resistance and more. When we talk about production of DLC, there are two major methods to produce the DLC which is Physical vapor deposition (PVD method) and the other method is Chemical vapor deposition (CVD method) which is the common method (widely used) to produce the diamond like carbon and its depends on using of hydrocarbon gas, and it is simple in structure compare by other procedure, it has advantages among other procedure which is low cost and low roughness and low friction. Nowadays according to Increase the world's population and the increase in the average of weights of people that caused in increasing a number of load-bearing joints that need to be replaced by artificial implants such as hip joints implant, knee joints implant all of them is coated by DLC to provide high adhesion and more, also for the DLC-coated stent it is widely used in this time, and for advanced biomedical applications (new trends) of the DLC Coatings we can use it as Nanotechnology such as biomolecular monitoring, cancer therapy and neural cell.

**Keywords:** Diamond-like; Carbon films; Medical applications; Coatings

### 1. Introduction

#### 1.1. Biomaterials

Bio-materials are synthetic materials used to replace a part of a living system and to work in direct contact with body tissues [1].

The design and selection of biomaterials depends on the specific applications that will be used in to be useful and their properties can be guaranteed for the required period of time, without being rejected by the body [2]. In addition, the most important things in biomaterial that it does not cause inflammation and toxic reactions and allergic symptoms in the body, and it must be biofunctional, biocompatible, bioinert, bioactive and sterilizable (Fig1) [3, 4].

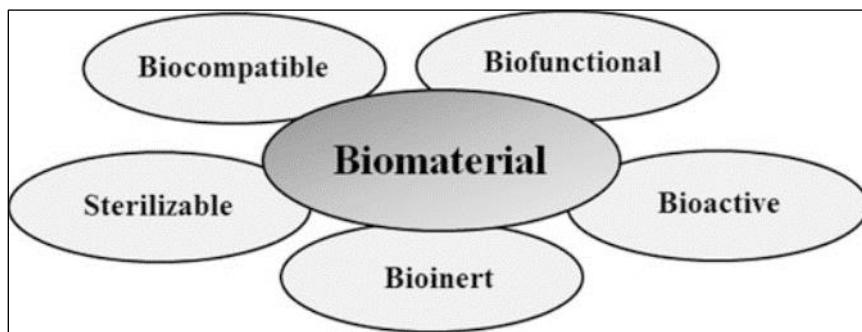
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Sterilization is also important and must be taken into consideration before using biomaterials in order to avoid being rejected by the body [4].

The surface of the biomaterial directly relates to living tissue and plays a crucial role in the compatibility of the biotic material with the tissue and the properties of the biomaterial can be used to improve tissue interaction [3].

The concept of compatibility has changed completely in the last few years. In the past it was thought that the completely inert material that has no response from the biological medium to its presence in the human body is the biocompatible material [5].

Biomaterials can be used for various applications in countless areas of the human body. However, the use of medical implants of biomaterial increased within the last decades as a result of the population ageing, boost in expectancy and style of life and enhancements in implant technology [6].



**Figure 1** Characteristics that a material must present to be a biomaterial

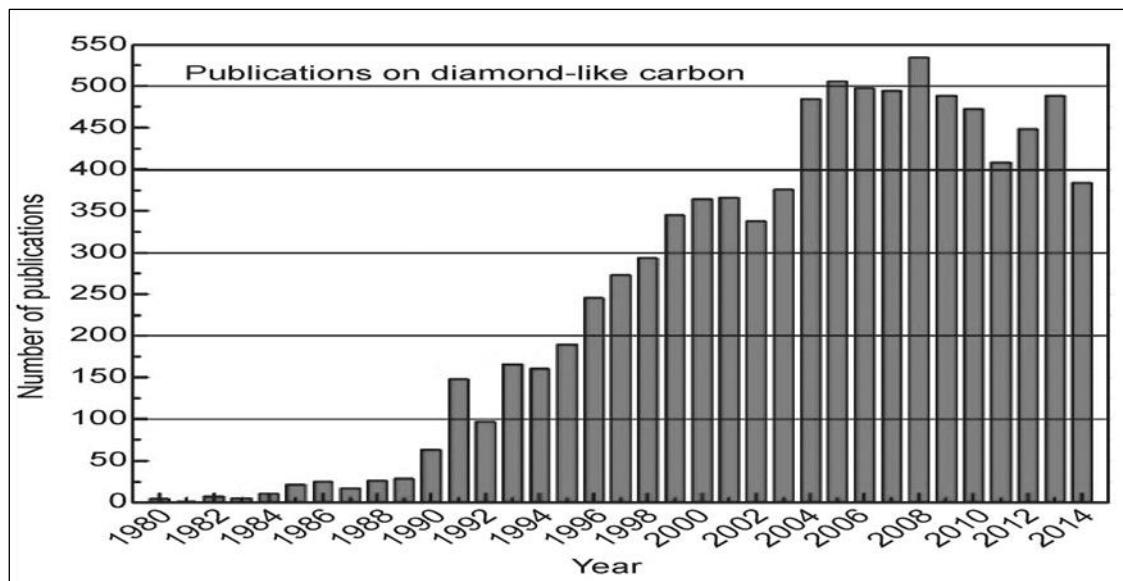
### 1.2. Diamond like carbon (DLC)

Carbon has been the key element during the last three decades in the synthesis of thin coating of diamond, **diamond-like carbon**, carbon nitride, boron carbide, and a myriad of transition metal carbide and carbo-nitride coating. Because of their excellent mechanical and tribological properties, these coatings are now used in an extensive range of engineering applications to control friction and wear [7].

DLC Film is amorphous in structure also has the properties nearly comparable to that of diamond. The properties of these films predominantly depend on the deposition methods and process conditions employed. Even today, the nature of these materials is not understood to the full extent. These films are broadly classified as hydrogenated and non-hydrogenated. The hydrogen content predominantly affects the DLC film structure. Hydrogen content is less than 1% in non-hydrogenated DLC films whereas, it can be about 60% in hydrogenated DLC films. Usually, these films have a combination of  $sp^3$ ,  $sp^2$  and sometimes  $sp^1$  bonds. The presence of hydrogen not only stabilizes the dangling bonds but also contributes in achieving wide optical gap and high electrical resistivity. DLC films have a smoother surface compared to diamond and hence have many applications that cannot be fulfilled by diamond films [8].

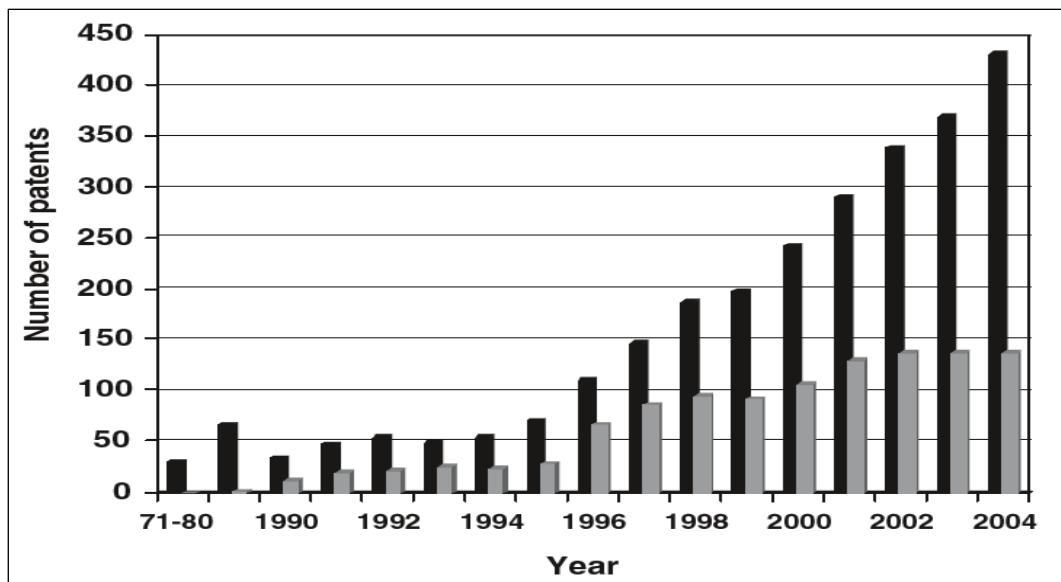
### 1.3. Historical overview

The family of DLC coatings is the largest and represents one of most studied between all other coatings. These coatings were first discovered in the 1950s by Schmellenmeier [9] but didn't attract much attention until almost two decades later by Eisenberg and Chabot work [10]. During the 1980s, a few researchers developed interest in these films, while during the 1990s, the research on DLC films increased momentum [11,12]. As can be deduced from Figs 2–3, almost each year since the early 1990s, numerous papers and patents have been dedicated to DLC films. Since 2000, these films have attracted even more interest, and they are still the subject of numerous scientific studies [12].



**Figure 2** DLC coating publication from 1980 to 2014.

Since the early 1990s, patents and numerous papers have been devoted to DLC films. Since 2000, these films have attracted even more interest, and they are still the subject of numerous scientific studies until today [7].



**Figure 3** US patents issued Number per year on DLC coatings (black bar) and on their tribology (grey bar)

### 1.3.1. Inception and Early studies

Historically, in 1953 when Heinz Schmellenmeier reported a black carbon film derived from  $C_2H_2$  gas in glow-discharge plasma, in addition, was the earliest attempts to produce DLC films. His film exhibited great hardness and, hence, it was very resistant to scratching by other hard objects. Later work by Eisenberg and Chabot produced such films on negatively biased metallic substrates in the early 1970s by using an ion beam deposition system [10]. Their films were very hard and hence resistant to scratching and possessed a high index of refraction, high dielectric constant, excellent optical transparency, and high resistance to corrosion in strongly acidic solutions. Because DLC films are typically dense and amorphous, they were less prone to pin-hole defects. During the mid-1970s, Holland et al. and a few other researchers were as well able to synthesize DLC films from other hydrocarbon sources by basically applying a radio frequency (RF) bias to the substrate materials and thus producing a glow-discharge plasma [11-13]. Because of the remarkable mechanical properties of these films, some researchers had speculated they were perhaps composed of

crystalline diamond, but systematic microscopic studies during the late 1970s by Weissmantel et al. Dispelled these speculations by confirming that the DLC films had an amorphous structure [13].

There were a few other reported papers on DLC films during the 1980s. Review paper by Arnoldussen and Rossi from IBM predicted and discussed in detail the potential usefulness of thin DLC films for magnetic recording media in a 1985 [14]. In 1981 by King such a possibility had already been explored, who was able to show far superior performance and efficiency for the DLC-coated disk media than the other types of overcoat materials used in hard disks at that time [15]. Based on King experimental findings, his concluded that the use of these films in magnetic recording media might lead to some major advances in the hard disk industry, and obviously King predictions were right. During the 1980s, only a few tribological studies were performed on DLC [16-17]. Robertson published a comprehensive review paper on amorphous carbons, plus DLC films, and summarized the details of the most important characteristics of such materials in 1986s [18].

in the 1990s, we have A large rise in DLC-related research activities, with the number of publications and granted patents (see Figs 2-3) and continued in subsequent years. Many novel types of DLC films were formulated during these years; diamond like carbon (DLC) films were produced on an industrial scale and systematic tribological studies were carried out. Because of such increased manufacturing and scientific activities, our information base on DLC film increased tremendously [7].

## 2. Properties

### 2.1. Adhesion

The strong adhesion is very important of the films for enabling the use of DLC as an applications of implant coating in high mechanical stress. The high-quality DLC coatings produced with method (PVD) have high internal stresses originated during the deposition process. During the deposition molecular species arriving with high energies to the surface can become implanted into the film where they may occupy interstitial sites and therefore produce compressive of stress. That is why DLC films have considerable intrinsic, compressive, up to 10-12 GPa stresses [19, 20]. This property is directly correlated to the fraction of sp<sup>3</sup> bonds in the films [21-22].

### 2.2. Biocompatibility

Biomaterials within the human body are usually good tolerances. If the implant can function effectively for the time period required, it is called biocompatibility. Generally, the biocompatibility is the ability of a material to perform with an appropriate host response in a specific application [1]. However, all implant material, regardless of how "biocompatible", stimulates a tissue response. This response could be very small and the patient is not aware of any problem as long as an implant performs its function. In other side, the patient's life could be in danger. When the subcutaneous implant is placed inside the body, a biomaterial will stimulate the inflammatory response. This is the primary reaction of the host defense mechanism and also the beginning of the wound healing response. Human body responses to the wound are known, but those surgical implants of a medical device are less understood. The common method for evaluating implant biocompatibility is to characterize hemocompatibility and cytotoxicity by measuring protein adsorption or cell adhesion on the surface of the implant material. [23]

#### 2.2.1. *In vitro*

Effects of DLC coatings on the activation and growth of macrophages have been studied.

Macrophages are tissue immune cells resulting from mononuclear monocyte precursors circulating in the blood. Macrophages are known for their ability to phagocytize ("eat") extracellular particles. Experiments using murine [24, 25, 26], or macrophages and human monocytes [26, 27] showed consistently a biocompatibility of hydrogenated and non-hydrogenated DLC coatings in association with an appropriate high albumin-to-fibrinogen adsorption ratio measured on the DLC surface [24].

#### 2.2.2. *In vivo*

Many studies have been done in vivo tests of DLC coating since mid-90s [28, 29, 30, 31, 32, 33]. The following major findings and experiments were published:

- DLC coated stainless-steel cylinders were implanted into sheep's muscular tissue and cortical bone for four and twelve weeks. Results showed the biocompatibility of DLC coatings [28].

- DLC coated implants within the tibiae of Wistar rats were better Osseo integrated during thirty-days than the reference titanium implants [30].
- DLC-coated cobalt chromium cylinders were implanted in intramuscular location in rats and transcortical sites in sheep for ninety days. histologic analysis of samples retrieved after surgery showed that the DLC coated samples were well tolerated in each sites [29].
- DLC-coated Ti samples were implanted in skeletal muscle of rabbits for one, three, six and twelve months. The tissue cell interaction information indicated that DLC was biocompatible with skeletal muscle [31].
- DLC coated implants in SV129 mice were monitored up to 6 months and showed a benign tissue response in subcutaneous tissue [32].
- DLC-coated cylinders were inserted into femoral condyles and muscular tissue of rats for four and twelve weeks. after histological surgery analyses showed that the DLC coating was well tolerated in these sites, demonstrating in vivo biocompatibility of DLC [33].

In summary, the results collected above strongly support that the DLC coating is biocompatible in both in vitro and in vivo experiments.

### **2.3. Hemocompatibility**

The body reactions to a blood contacting implant rely on the surface conditions of the implant like the surface texture, chemical composition and local flow conditions.

Two main characteristics of the characterize hemocompatibility are evaluated: blood clotting and coagulation cascade. This is assessed by investigating the adhesion of human platelets, albumin and fibrinogen to the surface of the material. The high absorption ratio of albumin / fibrinogen protein measured on the implant surface represented a low tendency of thrombus formation. Studies show that DLC, has a higher proportion of absorbed fibrinogen proteins / albumin compared to surfaces. [23]

study of platelet adhesion has showed that DLC surfaces have lower numbers of adhering platelets when compared with other implant materials [34]. The hemocompatibility of the material can also be assessed by measuring the ratio of hemolysis (the lower the hemolysis ratio, the better of hemocompatibility) [35]. Recent studies by Sharma et al. show that the ratio of hemolysis on DLC coated PMMA is comparable to that in normal saline. They reported that DLC is a biocompatible material at the nano level [36].

Combining all the above results, it can be inferred that DLC is a highly hemocompatible material.

### **2.4. Cell attachment and cytotoxicity testing**

For successful tissue growth and contact on the implant, the cells should spread, proliferate and attach well on the implant surface. It has been observed that fibronectin plays a major role in mediating and promoting the adhesion, growth, spreading and differentiation of cells on biomaterials. so, fibronectin behavior is typically applied in testing of materials biocompatibility. [37]

Material is considered as cytotoxic if cells don't proliferate well or die upon contact with the surface of the material.

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## **3. Production**

### **3.1. Introduction**

There are many ways to produce diamond-like-carbon and all of these methods depends on the lower density of the  $sp^2$  than  $sp^3$  carbon. By using pressure and catalysis or combination of these or impact at the nanometer scale (atomic scale) to make the  $sp^2$  bond carbon atom closer to the  $sp^3$  bond carbon atom [38].

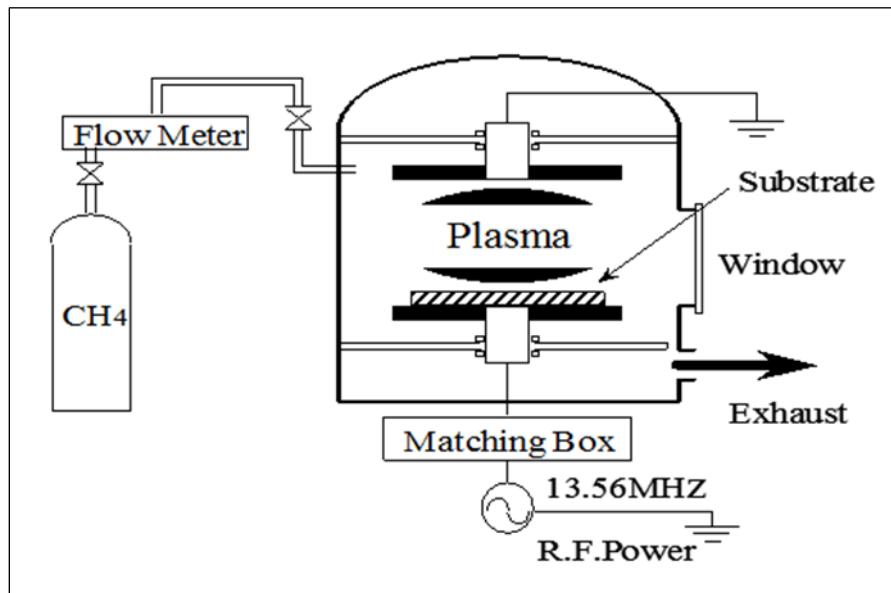
the common methods for production are two methods: Physical Vapor Deposition (PVD) and Chemical Vapor Deposition (CVD). The physical vapor deposition depends on using of solid graphite (being as carbon source) and the common result for this method is laser vapor deposition. [39]

And the chemical vapor deposition depends on using of hydrocarbon gas, and its widely used for production of diamond like carbon films in large area because it has a good feature which is simple in structure compare by other procedure, also the chemical vapor deposition has too many advantages which is very important if you compare it with another procedure or technique. The important advantages of CVD method are low cost (very important for the manufacturing), low roughness and low friction. [40]

For medical application we need materials for insulting coating to avoid problems such as corrosion.

### 3.2. Experimental procedures for producing DLC (r.f method)

The schematic diagram on the Fig.4 shows the experiment for diamond like carbon deposition, and the chamber and electrodes are made as stainless steel.



**Figure 4** The experiment for diamond like carbon deposition, and the chamber and electrodes are made as stainless steel

This system on the Fig 4 including the following:

- Radio frequency generator
- Anode side electrode
- Cathode side electrode
- CH4 gas
- Plasma
- Matching box
- Vacuum pump
- Electrical power

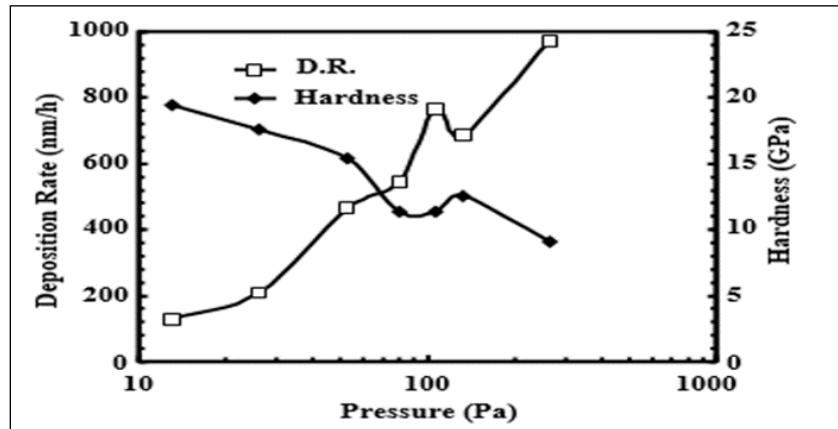
The radio frequency generator is (model SS-310 AAE, from Fuji company) and the cathode side electrode contains aluminum cylindrical electrode, the matching box is (model1397 Sargent-welch company), the vacuum pump is (model-GR143030 Gore & associates company), the electrical power it was held at 100W (constant value), CH4 gas pressure is at 10Pa and the deposition time is 30min. [42]

#### 3.2.1. Procedure:

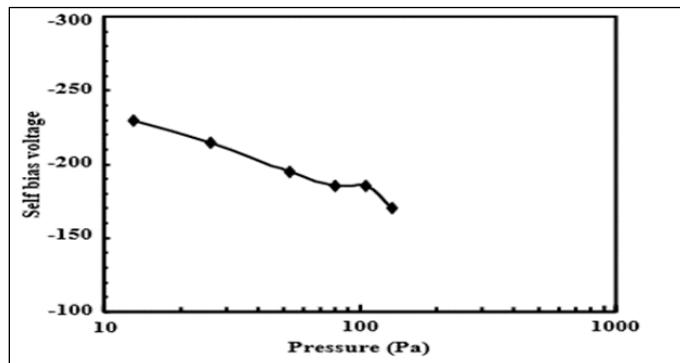
By applying radio frequency (13.56MHz) to both electrodes and to keep the clearance between both electrodes (the upper and lower electrodes) at 100 mm, the look is seem as disk for electrodes 50mm (half diameter) and vacuum chamber is set to unhand at 3-10 Pa by the pumps. After that the methane gas (CH4) entered to the device in vary status of pressure and the power is 100W over the deposition of DLC film. the condition which applied to the deposition modified for each single sample. [41]

#### 4. Result and discussion

On the Fig 5 it shows that the relations between hardness, pressure, and deposition rate. The hardness of the DLC films is decreases as the pressure is increase, and this relationship is in interaction with self-bias as shown in Figure 6 while the negative-bias voltage decrease within the same time, the plasma density and the deposition rate is suddenly increase with the pressure. [43]

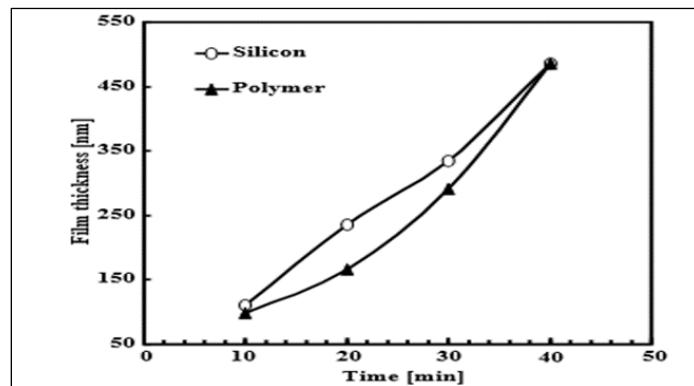


**Figure 5** The deposition rate and hardness of DLC films as a function of pressure



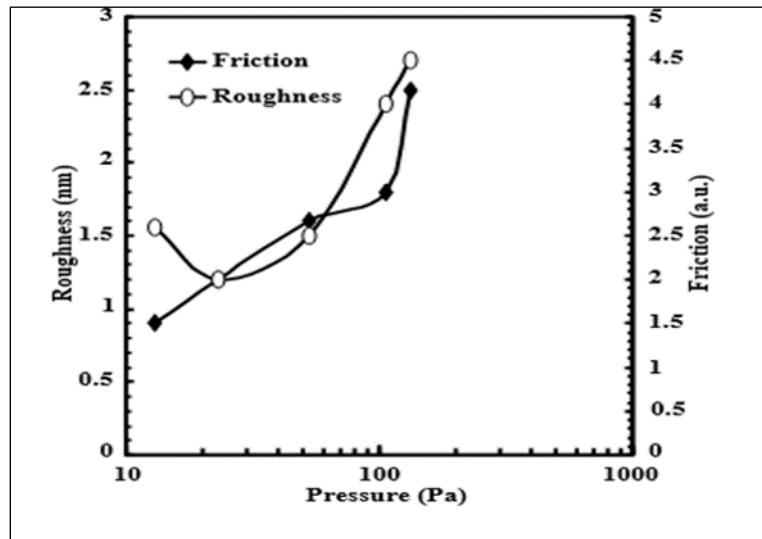
**Figure 6** Relationship between self-bias voltage and pressure of methane gas the deposition of DLC film

In Fig 7 polymeric material used for the deposition rates, the deposition rate on the polymeric substrates is similar to the Si substrate. [44]



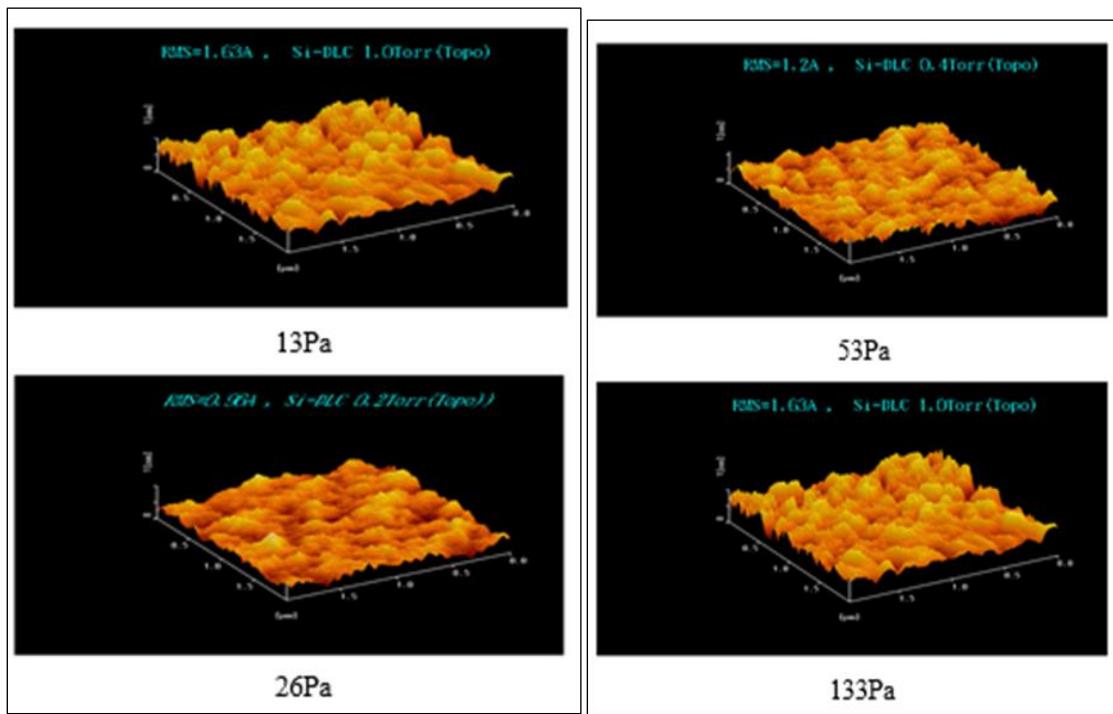
**Figure 7** Film thickness of DLC films on polymeric substrate and Si at pressure of 53 pa

On Fig. 8 shows that RMS (root mean square) roughness and friction as a function of the pressure, the friction and roughness are increase with pressure, these value is used to evaluate variation of DLC film friction and roughness. [45,46]



**Figure 8** Friction and roughness of DLC films deposited at Si substrate as function of pressure

On the Fig. 9 AFM (Atomic Force Microscope) images show us the surface roughness of the diamond-like-carbon films deposited at Si substrates.



**Figure 9** Surface roughness of the diamond-like-carbon films deposited at Si substrates.

## 5. Biomedical application

### 5.1. Introduction

After we talk about the proprieties of the DLC, we conclude that the DLC film has a proprieties and biocompatibility that we need to use in different biomedical application.

There are many applications of DLC film in different biological system and it is become larger and more common with time and more advance in use in many areas such as orthopedic (especially in tribology), cardiovascular system and optical areas

Nowadays according to Increase the world's population and the increase in the average of weights of people that caused by to many reason (fast food, Neglecting sports , etc ..) that caused in increasing a number of load-bearing joints that need to be replaced by artificial implants , for example ( hip joints implant , knee joints implant and also spinal disk but it is in lesser extent ) all of them is coated by DLC [47] , also in cardiovascular system ( in the stent ) it is widely used in this time and it will come larger because of the same reason we talked about.

For advanced biomedical applications of the DLC Coatings we can use it as Nanotechnology, such as: biomolecular monitoring, cancer therapy and neural cell culture. [48]

## 5.2. Orthopedic Applications of DLC

DLC become more widely used especially in orthopedic (tribology), nowadays because of increasing in the average of weights of people and other reason such as people does not do sport that caused to increase the number of load-bearing joints and high friction in joints that must be replaced by artificial implants to provide ability to movement.

The major problem with the prosthetic joints is in its wear and corrosion after long term of use after years. if two material surfaces slide with each other and they are in relative motion, then that caused to the biomaterial which have lower hardness it will worn out so cause to loosening of the implants (for both implants) and this biomaterial will be also in contact with the body blood, so we need these implants coated by material need to be hard and also inert to prevent the corrosion and to be fixed in long time The inertness, corrosion and wear resistance, high hardness, low frictional coefficient, as we talked before on the properties and compatibility of DLC which is corrosion resistance, wear resistance, very low frictional coefficient and its high hardness we concluded that the DLC is suited [49].

All of these proprieties make DLC a promising for biomedical application on the surfaces of orthopedic implants such as DLC-coated knee joint and DLC-coated femoral heads. as it shown on the Fig. 10 one of application of DLC-coated femoral heads [50].



**Figure 10** one of application of DLC-coated femoral heads

## 5.3. DLC-Coated Stents

First, we need to define the stent: "A stent is a metal tube that is inserted permanently into an artery". It will help to open an artery to make the blood flow in it. " nowadays the stents has become more common and more widely used in the world for cardiac implantation for many reasons related to the diseases of the heart, for example people nowadays are over-eating in fast food that is full of fat that cause thrombus so we need to the stent to keep the artery open so the blood can flow [51].

In past years some researchers did in vitro and in vivo studies on the DLC-coated stent and its effect on the platelet activation and metal release. it was observed that all of the metals have corrosion after implantation [52]. So, we need

to the biomaterial-coated stent to avoid some problem like this, so as we talk before about the hemocompatibility of the DLC and the important properties of it such as corrosion resistant, we conclude that it is a good choice and suitable.

When we go back to the history we see that the first successful implantation of stents that coated by DLC was in the year 2004 for endovascular treatment of femoral artery occlusive disease. [53]

#### **5.4. DLC-Coated Guidewires:**

First, we need to defined the medical guidewires: Tools used to insert some medical devices such as catheter and stent inside the human body. the stainless steel is used as the material of the guide wire, but for better frictionless, they coated the stainless steel by polytetrafluoroethylene (PTFE) or silicon overlays. Also, these coatings stick with the side effects of nonuniformity, non-stable and its poor adhesion with the stainless steel which result in releasing the coated materials. It must be noted that the guidewire should not cause any implant impact or damage the vessel walls during its insertion. The medical guidewires required important properties that is must have to be inserting in the human body ( for the biocompatibility and other procedures ) which is high flexibility , inertness, Friction less, hemocompatibility, and also it has a properties that is not including to the polytetrafluoroethylene-coated stainless steel or silicon overlays and all of these properties we have it in the diamond like carbon properties ( such as good adhesion , stable) some studies were made about it so it's a good choice to decide to be coated with the DLC. [54,55,56]

#### **5.5. DLC-Coatings for Advanced Biomedical Applications:**

DLC nowadays is a many-sided coating material that is used in a many of mechanical and biomedical applications, such as endo prostheses and dental implants [57]. As we say before in biocompatibility and properties, DLC provides mechanical hardness and cell-compatibility in the same time, high hardness, frictionless, high wear resistance to make it more sustainable [58]. Also, DLC coating is noncytotoxic and antithrombogenic. so, they are being explored for several in-vivo and in-vitro biomedical applications extend from orthopedic applications to cardiovascular, DLC coatings has been certified as biocompatible in both in vitro and in vivo studies because of their strong C=C bonding environment [59,60]. Nowadays Graphene and carbon nanotubes are widely extend carbon allotropes for advanced Nanotechnological applications, but they are not popular for biomedical applications because of their cytotoxic effects. In this respect DLC become mechanically stable and noncytotoxic, recently, DLC through proper functionalization, is become as strong material for advanced biomedical applications, either in therapy or diagnosis (monitoring) for example bimolecular monitoring, cancer therapy and neural cell culture. [61]

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## **6. Conclusion**

After reviewing the diamond like carbon coating material studies have shown the ideal properties of DLC, which are high biocompatibility, which showed the results of studies that it is biocompatible in vivo and in vitro experiments. Studies have also shown the homeocompatibility with DLC and also lack of toxicity in this biomaterial. All these results reinforce confidence in the use of DLC for medical purposes.

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### **Compliance with ethical standards**

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#### *Disclosure of conflict of interest*

No conflict of interest to be disclosed.

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## Appendix

- AFM Atomic Force Microscope
- CVD: chemical vapor deposition, general term for deposition method in which the gaseous deposit contains reactants that decompose and recombine to form the desired thin film DLC: Diamond-like Carbon
- Si: silicon
- sp1: oligomer hybridization of carbon valence electrons
- sp2: planar hybridization of carbon valence electrons
- sp3: tetrahedral hybridization of carbon valence electrons
- Ti: titanium
- PMMA: poly (methyl methacrylate)
- PVD: physical vapor deposition, general name for deposition methods in which the atoms of the deposit are ejected in the vapor either by thermal evaporation or by atomic impact.
- RF: radio frequency
- Adhesion: adhesion reflects the behavior of cells shortly after contact to a surface.
- Adsorption: Increase in the concentration of a substance at the interface of a condensed and a liquid or gaseous layer owing to the operation of surface forces.
- Artificial: Qualifier for something that is made by human activity, rather than occurring naturally
- Bioactive: Qualifier for a substance that provokes any response from a living system.
- Biofunctional: Describing a material whose function is dependent on a biological content.
- Bioinert: are ones which do not initiate a response or interact when introduced to biological tissue. In other words, introducing the material to the body will not cause a reaction with the host.
- Biocompatibility: Ability to be in contact with a living system without producing an adverse effect.
- Biomaterial: *Material* exploited in contact with living tissues, organisms, or microorganisms.
- Hemocompatibility: Compatibility of blood.
- Nanoindentation: is a variety of indentation hardness tests applied to small volumes.
- Polymer: Substance composed of macromolecules.
- Toxicity: Consequence of adverse effects caused by a substance on a living system.