Deposition of diamond-like carbon coatings: conventional to non-conventional approaches for emerging markets

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Abstract

Diamond-like carbon (DLC) coatings are recognized for a broad range of industrial applications due to their superior mechanical properties such as high hardness, low friction, and promising wear resistance. DLC coatings are commonly produced with physical vapour deposition (PVD) and plasma-enhanced chemical vapour deposition (PECVD) methods. New DLC markets are emerging in electronics, biomedical, additive manufacturing and textiles sectors with industrial transformations. The conventional PVD and PECVD methods may have limited usage for depositing emerging DLC products due to their elevated thermal and high vacuum environment, lack of localized deposition function, and production throughput restrictions.

This review begins by briefly describing DLC coatings background, the volume of research outcomes and the global revenue in the past decade and projections for the future. DLC structural designs made with conventional deposition methods and corresponding operational parameters are then discussed in detail and enhancement in conventional methods to improve DLC coating quality and to resolve unaddressed problems are summarized. The emerging DLC applications and potential of non-conventional methods to produce DLC coatings are critically analysed with specific attention to scientific, technological and economical aspects. Representative investigations suggest that DLC coatings can be produced with hardness values up to ~ 20 GPa using dielectric-barrier-discharge deposition, hydrophobicity up to ~ 167° with electrospray assisted plasma jet coating, high deposition rates up to ~ 6 µm/min with microwave resonator deposition, and critical load of ~ 30 N with a friction coefficient of ~ 0.1 when deposited with the plasma gun technique. The review concludes by recommending systematic investigations to optimize geometric and operational

parameters of non-conventional DLC deposition methods which can produce high-quality DLC coatings at low temperatures and atmospheric pressures with scalability to meet emerging market demands.

Keywords: Carbon; Diamond-like carbon; Coatings; Deposition; PVD, Micro-plasma; Emerging Markets

1. Significance of Diamond-like carbon coatings

Diamond-like carbon (DLC) coatings are widely used in aerospace [1], automotive [2], defence [3], food and beverage [4], electronics [5], mechanical [6], biomedical [7], surface cosmetics [8] and digital [9] industries. Their recognition in this broad range of industries is due to their superior and multifunctional features. They have excellent mechanical properties i.e., high hardness and Young's modulus, good scratch resistance, lower friction, and high wear resistance. Similarly, these coatings have proven biocompatibility, and are chemically inert and thermally stable up to ~ 300 ^{o}C [10]. Engine components, dies and moulds, mechanical components and cutting tools; like stamping, drill bits, end mills, and similarly gears and bearings are popular DLC products. Likewise, DLC coated stents, and surgical instruments for the biomedical industry, hard discs for the computer industry, watches in the jewellery industry, transistors in the electronic industry, solar panels for the renewable energy sector are also representative examples of DLC products.

The DLC global revenue is continuously growing due to these increasing industrial demands. The projected trends for growth in the DLC global revenue for the period 2009 to 2025 are summarized in Fig. 1A. The projected figures [11] were \$782 M, \$905 M, and \$1700 M for 2009, 2010, and 2015 respectively. Similarly, \$1859 M by 2022 [12], \$1930 M by 2024 [13] and \$2120 M by the end of 2025 [14] have been forecasted by marketing research companies. This hefty increase in the DLC market of more than 4 % compound annual growth rate (CAGR) is a key incentive and motivation to boost DLC research and development. Moreover, as shown in Fig 1B., the number of DLC publications indexed in ScienceDirect database in the last 10 years increased by over 150 % i.e. from ~ 2800 in 2010 to ~ 7500 in 2020. This significant growth in global revenue and associated increase in research outputs reflect the growing importance and high demand of DLC coatings.

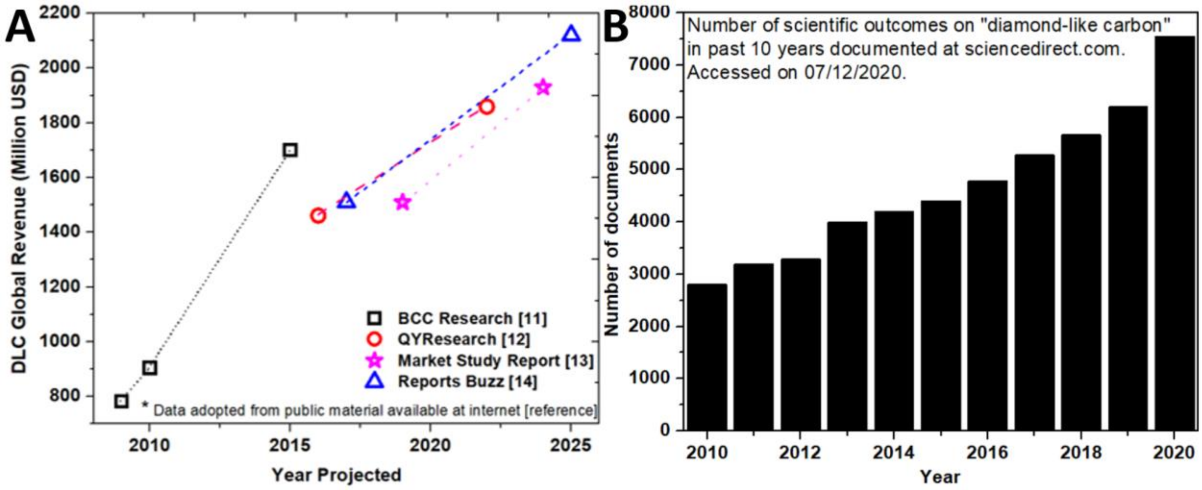

Figure 1. (A) Global revenue of DLC market projected from 2010 to 2025, (B) The number of DLC research outputs indexed in ScienceDirect.com from 2010 to 2020.

2. Conventional methods and designs to deposit diamond-like carbon coatings

Carbon coatings were first explored in 1954 [15] and retained graphite-like nature until the diamond-like feature emerged in the 1970s [16,17]. The DLC coatings were adopted in industry relatively earlier than research labs if compared with graphene, and MXenes materials. The scientific investigations to further improve the quality of the DLC coatings, to minimize their limitations, re-coating matters, and growth mechanisms commenced in the 1980s.

The graphite-like and diamond-like features are dominated by sp^2 and sp^3 fractions respectively. The transformation from graphite-like to diamond-like features desires higher sp^3 fractions in the coatings, which guarantee their superior performance. Therefore, these coatings are further classified on the basis of sp^3 fractions as amorphous carbon (a-C) and tetrahedral amorphous carbon (ta-C) coatings. Usually, the sp^3 starts from 20 % in a-C coatings and reaches to 85 % in ta-C coatings [18]. Hydrogenated DLC (a-C:H) coatings are another broad area and can be sub-classified as polymer-like a-C:H (PLC:H), diamond-like a-C:H (DLC:H), graphite-like a-C:H (GLC:H), and hydrogenated tetrahedral amorphous carbon films (ta-C:H) based on their hydrogen content [19]. The DLC structure is capable of containing 17 to 61 at.% of hydrogen atoms [20]. Hence, typically a-C:H (PLC:H), a-C:H (DLC:H), ta-C:H, and a-C:H (GLCH) coatings contain hydrogen contents up to 40-50 at.%, 20-40 at.%, ~25 at.% and less than 20 at.% respectively [19]. The structural configuration [21] of DLC coatings changes with the amount of hydrogen content, which governs the number of new atomic bonds formed between carbon and hydrogen atoms and consequently changes their mechanical [22], electrical, and physical properties, and tribological performance [23]. It is deduced that the in-situ [24, 25] or ex-situ [26] addition of hydrogen promotes polymer-like characteristics and boosts the superlubricity phenomenon. To increase the sp^3 fractions, DLC coatings are being deposited with numerous methods such as sputtering, arc methods, ion beams, radio-frequency glow, microwave plasma, and their hybrids. Table 1 summarises these popular DLC deposition methods, along with the corresponding deposition parameters, such as ion energy, substrate and target bias voltage, target current, vacuum pressures etc., which significantly influence the DLC growth, quality, and corresponding properties.

Table 1. Common technologies and experimental parameters for DLC deposition with conventional methods.

Sr.	Deposition Techniques	Further reading	Sr.	Process Influencers	Further reading
1	Sputtering		1	Ion Energy	[45]
	○ Balanced magnetron	[27]	2	Ion flux	[46]
	○ Unbalanced magnetron	[28]	3	Pulse Power	[47]
	○ Radio frequency	[29]	4	Pulse frequency	[48]
	○ Reactive	[30]	5	Discharge power	[49]
	○ High Power Impulse (HiPIMS)	[31]	6	Target current	[50]
2	Arc discharge / Filtered Cathodic Vacuum Arc	[32]	7	Substrate bias voltage	[51]
			8	Target bias voltage	[52]
3	Microwave plasma (MV)	[33]	9	Duty Cycle	[53]
4	Radio frequency glow (RF)	[34]	10	Deposition rate	[54]
5	Pulsed laser depositing (PLD)	[35]	11	Chamber pressure	[55]
6	Electron beam	[36]	12	Magnetic field intensity	[56]
7	Ion beam (IB)	[37]	13	Substrate roughness	[57]
8	Ion source	[38]	14	Source-to-Substrate Distance	[58]
9	Plasma Immersion Ion Implantation and deposition (PIIID)	[39]	15	Substrate Rotation	[59]
			16	Substrate Orientation	[60]
10	Hollow Cathode Plasma Immersion Ion Processing (HCPIIP)	[40]	17	Coating thickness	[61]
			18	Buffer layer thickness	[62]
11	RF + PECVD	[41]	19	Deposition temperatures	[63]
12	MW + PECVD	[42]	20	Pre/Post heat treatment	[64]
13	IB + Sputtering	[43]	21	Reactive gas/precursor orientations	[65]
14	PLD + Sputtering	[44]			

It is important to note that if a higher sp^3 fraction is an inspiration to produce good quality DLC coatings with high hardness, low friction and higher wear resistance; then such coatings also have some demerits, such as higher residual stresses, low toughness [66], and poor interface design. These issues have evolved the area of DLC structural design including, monolayers, multilayers, doped DLC, and hybrid microstructures as shown in Fig. 2. The monolayers of pure carbon may have structural variants of amorphous (Fig. 2A) [67], nano-crystalline (Fig. 2B) [68], or granular (Fig. 2C) [69] texture made with operational alterations such as temperature-dependent or glancing angle depositions. Similarly, the multilayer design may be composed of pure carbon layers (Fig. 2D) [27] deposited with different layer thickness and layer-specific bias voltage; or amorphous and non-amorphous hybrid structure i.e., bilayer amorphous carbon and carbon nanotube layers (Fig. 2E) [70]; or the combinations of carbon with non-carbons (Fig. 2F) [71]. In the same way, the DLC has been doped (Fig. 2G to Fig. 2I) [72-74] with a broad range of metallic, non-metals, ceramics, or gaseous elements to improve their toughness and wear resistance. The elements to be doped in DLC are specifically selected for certain purpose and application. Usually, the DLC toughness improves by metal doping but the coating loses its hardness at the same time [75]. Therefore, a significant compromise on DLC features is required while boosting its specific properties, such as biocompatibility, corrosion resistance, oxidation resistance, wear resistance, thermal stability etc. More details on DLC characteristics, scientific and technological aspects, and applications can be studied from comprehensive review articles [76-83].

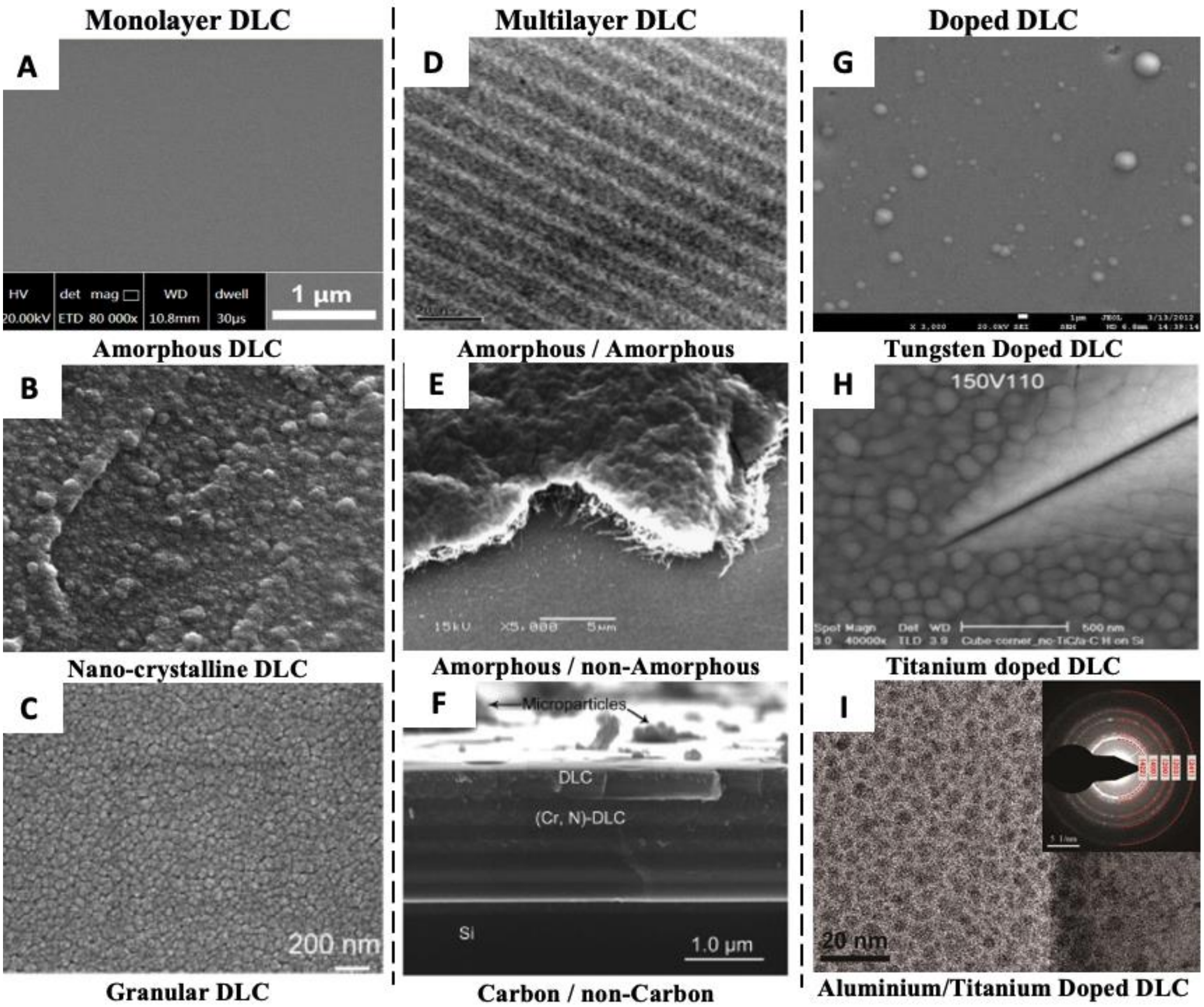

Figure 2 Architecture based classification of DLC coating i.e., monolayers, multilayers, doped-DLC produced with conventional PVD and PECVD methods. Reproduced with permissions: A [67], B [68], C [69], D [27], E [70], F [71], G [72], H [73], I [74].

3. Diamond-like coatings deposition with enhancement in conventional methods

Researchers are working to continuously improve long unaddressed problems, such as thermal stability at elevated temperature and toughness. Usually, DLC coatings are thermally stable up to 300 ^{o}C and further increase in temperature transforms them into nano-crystalline graphite [84] and the coatings lose DLC features of high hardness and superior wear resistance. By achieving thermal stability at elevated temperatures, the DLC coatings are expected to capture new products in aerospace, automotive, and turbomachinery.

Similarly, the graphene nanocrystallite embedded carbon coatings [85, 86] emerged early in this decade and have demonstrated improved friction behaviour. Fig. 3A and Fig. 3B present the plane and cross-sectional views of graphene nanocrystallite embedded carbon coatings produced with electron cyclotron resonance plasma sputtering. The coating growth and graphene nanocrystallite size were governed by electron irradiation densities. It is important to note that the friction coefficient increases with graphene nanocrystallite size, therefore, an optimized graphene crystallite size is desirous to receive superior tribological performance, for example, a 1.09 nm crystallite size corresponds to a friction coefficient of 0.03; 1.34 nm corresponds to 0.11, and 1.67 nm corresponds to 0.16, respectively [86]. Similarly, Fig. 3C and Fig 3D present plane and cross-sectional views of isolated carbon nanoparticle embedded carbon coatings [87, 88]. The isolated carbon nanoparticles were created in-situ by unbalanced magnetron sputtering with controlled size and distribution ranges and simultaneously embedded in an amorphous carbon coating matrix at controlled depths, in a single-step process [89, 90]. The new DLC coatings were reported with ~ 20 % higher hardness, simultaneously to ~ 10 % increase in toughness and 60 % decline in wear rate at ~ 3 GPa contract pressure. The continuation in advanced DLC research with enhanced-PVD methods and hybrid microstructural design is expected to report superior developments in the coming years.

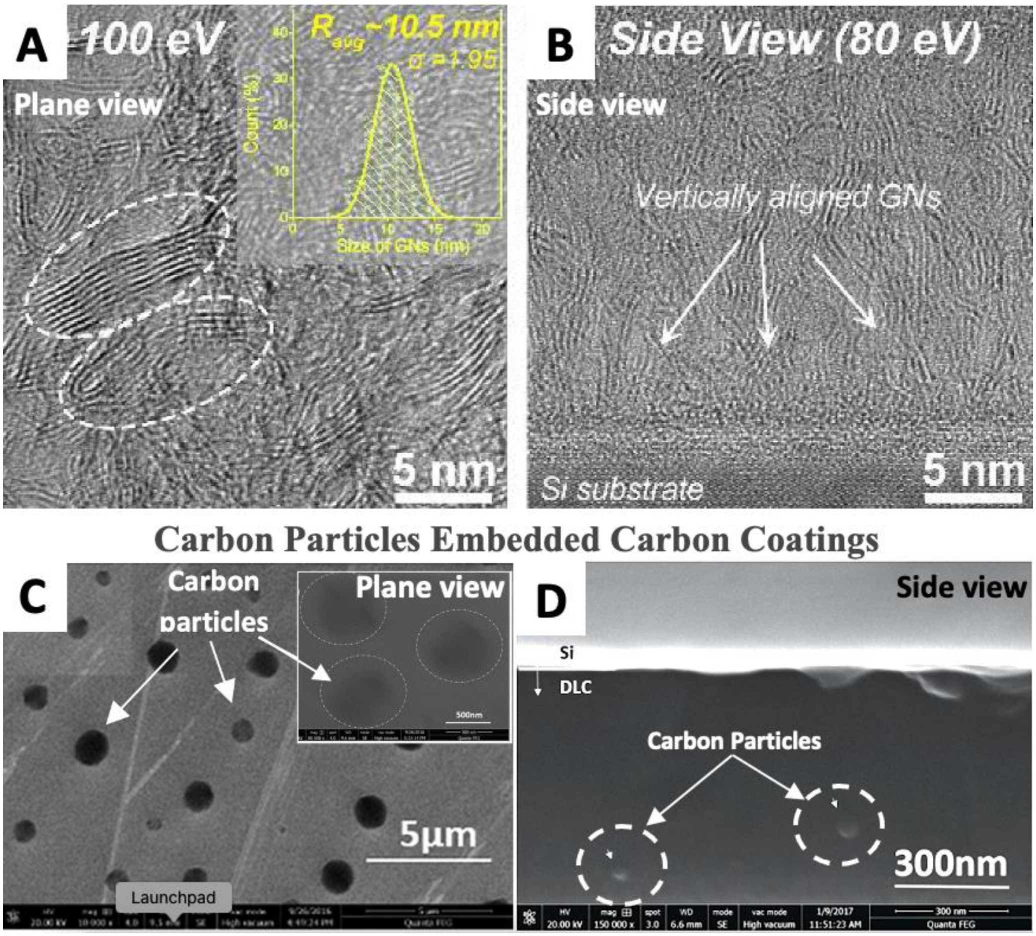

Figure 3 (A) Plane view and (B) cross-sectional view of graphene nanocrystallite embedded carbon coatings, reproduced with permissions [85]. (C) Plane view and (D) cross-sectional view of isolated carbon particle embedded carbon coatings, reproduced with permissions [87].

4. Diamond-like carbon coatings and emerging markets

DLC coatings have been actively used by numerous manufacturing industries for the past 40 years. However, the last 10 years have remained important due to the industrial transformation which has inflated process digitalization, role of advanced materials, and state-of-art manufacturing technologies for new and emerging applications. Some of the DLC markets are shrinking while new DLC markets are emerging in different industrial sectors. The impact of industrial transformation on DLC can be elaborated with an example from the automotive industry, which claims a 58.87 % [13] share in the DLC market. Fuel engines which represent the largest DLC proportion in the automotive market, save mechanical losses by coating the engine components with hard, low friction and wear resistance DLC, which also reduces energy losses by minimizing unburned fuel and CO_2 emissions. However, electric vehicles are now rapidly replacing fuel cars and consequently, DLC is losing one of its biggest markets. The electronics dominated future is also expected to contribute to further decline in the DLC contribution from mechanical sectors such as bearings, magnetic discs, mechanical components, etc.

At the same time, biomedical, electronics, acoustics, textiles and cutting tools are emerging sectors for DLC coatings. DLC coated textiles have presented excellent filtration [91] and antibacterial properties [92]. Surgical and electro-surgical [93] instruments, heated filaments, and implants are common examples from the medical industry. In the coming years, DLC coatings have a potential to actively serve in the healthcare sector for anti-bacterial touch surfaces in elevators, escalator rails, trains and buses etc. Similarly, DLC coatings are gaining popularity in acoustic devices such as audio speakers and headphones. DLC coated diaphragms [94] in loudspeakers and earphones help to improve the frequency response [95]. In 2010 [11], a 33.5 % CAGR for biomedical devices and 35 % CAGR for audio speaker applications was projected for 2015 and is likely to grow further in coming years.

In the electronic industry, the DLC coatings have proven applications for thin-film transistors, solar cells and micro-electromechanical system (MEMS) [96]. In particular, DLC is expected to play the same role for MEMS as the mechanical industry but it will be at micro and nanoscales rather than macro scale. If DLC coatings were reducing friction in engine pistons, then they are expected to perform similar functions for micro-sized electronic motors, where they can offer low friction, self-lubrication, smooth surface finish, mechanical durability and additional features of controlled insulation or conductivity barriers. Carbon coatings are not only used as typical functional coatings but are now becoming the essential design criteria for battery materials. For example, carbon coatings are integrated with lithium titanium oxide to store energy for flexible wearables [97]. Similarly, these carbon coatings are reducing the limitation of potential battery materials, such as, olivine-structured LiFePO4, which have promising capabilities to act as a cathode in lithium-ion batteries for electric vehicles but are currently restricted due to their poor conductivity [98]. DLC coatings also have increased revenue in the digital industry where they are emerging for 3C products i.e., computer, consumer electronics, and communication devices such as mobile phones. DLC has a proven record of reducing contact friction in data storage drives [99,100] in the computing industry and is now improving the performance and durability [101] of numerous flat [102], flexible [103] and liquid crystal displays [104] and digital screens.

DLC coatings were also proposed for process industry pipelines about twenty years ago [105] but are now actively emerging in the oil and gas industry due to technological advancements in deposition methods, such as radial anode ion sources [106] and hollow cathode discharge to coat ~ 10 μm [106] thick DLC at inner surfaces of oil and gas pipelines [108]. The uniform axial distribution [109] of DLC coating inside such long tubes and pipelines is still an active research area to enhance product life and performance.

DLC is still progressing for specialized mechanical applications. For example, diamond-coated cutting tools are currently used for aluminium machining. The manufacturing of light-weight electronic products encapsulated in aluminium housing, such as mobile phones, tablets, laptops, cameras etc. have drastically increased in the last decade. Therefore, the demands for diamond-coated cutting tools are also increased. However, pre-requisites like acid treatments, slow growth rate, expensive raw materials like diamond slurry for seeding, and longer operational time make diamond coatings very expensive. Nonetheless, DLC coatings are still being actively researched and tested for this purpose and expected to grab a big market share soon. Thicker DLC coatings are now emerging which are desirous for extended product life in numerous applications such as tribological performance. Previously, the thickness of DLC coating was needed to be compromised due to higher compressive stresses which reduce their adhesion with the substrate. The adhesion of DLC coatings [110] is still an active [111] research domain and now being addressed with foreign element doping [112] and interlayer designs [113]. A ~50μm thick DLC coating has reported [114] in current years made with precise buffer layer design and exhibits good adhesion as high as ~ 73 N.

Similarly, additive manufacturing is another revolutionary area of the current decade. Researchers are actively investigating the fundamental mechanisms of additive manufacturing, while in parallel, research is also expanding into surface coated additively manufactured products. DLC coatings have an established history for PET bottles for food [115, 116] and the medical industry [117, 118]. Therefore, DLC coatings have good potential to improve surface performance of additively manufactured products and to widen their applications. For example, the load-carrying capacity, contact stress distribution, durability, and service life of additively manufactured plastic parts like gears and washers etc. can be further increased with application of a DLC coating on their surface. Along with surface hardness and surface finish, it is anticipated that DLC coatings which are thermally stable up to 300 ^{o}C, can also improve the thermal stability of plastics, which usually deform around 150 ^{o}C.

DLC coatings may raise technical concerns when adopted for new products and markets. For example, DLC coatings have demonstrated good potential in nano-mechanical testing, where DLC coated atomic force microscope (AFM) [119] tips are a representative example. Even though DLC has proven high hardness, smooth surface finish, and low friction, the materials response and properties change from macro-to-nanoscale applications. The atomic-scale wear of DLC coatings in AFM tips have been recently [120] observed, which urges significant further investigation to assure accurate and repeatable measurements with such DLC coated high precision devices.

5. Potential of non-conventional methods to deposit diamond-like carbon coatings for emerging markets

5.1 Significance of non-conventional methods for DLC deposition

It is important to note that conventional DLC deposition systems such as PVD and PECVD, like sputtering and cathodic vacuum arc sources may not fulfil the essential requirements for future demands and applications. The conventional PVD methods have limitations of finite chamber space, maintaining higher vacuum, and higher deposition temperatures. The deposition temperature is a system property that could be below ~ 150 ^{o}C [121] or between 180 ^{o}C to 250 ^{o}C [122] if DLC is deposited by ion beam or sputtering, respectively. For certain systems like filtered cathodic arc or evaporators, the substrate-to-target distance also governs thermal effects on DLC deposition. The deposition temperature greatly affects the coating growth and corresponding properties; therefore, the temperature aspects are essentially related with appropriate selection of DLC deposition method and application. Higher deposition temperature improves thermal migration [123] and structural ordering of carbon atoms, and is thus desirous for tribological performance [124]. Therefore, the process temperature is additionally increased sometimes by substrate [125] or radiative heating. In contrast, low temperature (usually less than 100 ^{o}C) [126] and room temperature DLC deposition methods have also been previously attempted for flexible [127] and plastic [128] materials.

DLC coatings have mainly served the mechanical sector in the past 30 years, thus their deposition through conventional PVD methods i.e., sputtering, filtered cathode vacuum arc etc. remained popular than room temperature or atmospheric pressure depositions due to thermal stability of substrates, high-quality DLC coatings, superior mechanical properties and tribological performance. With the industrial transformation, DLC coatings have been adopted in various new markets like textiles, healthcare, and electronics with increasing demands. The textile industry is generally practising non-plasma methods such as dip coating, spinning, sol-gel, and photo-catalyses to deposit carbon, titanium, and zinc-based nanomaterials for energy, antibacterial, and environmental applications. The COVID-19 pandemic has further increased the usability of safe metal ions to boost the antiviral properties of textiles either for face masks or hazmat suits. The conventional plasma-based deposition methods may have minimized the shortcomings of non-plasma methods, such as adhesion, roll-to-roll capacity, and large area depositions, but not adopted broadly due to depositions at a higher temperature and under high vacuums. Therefore, there is a need to evolve non-conventional DLC deposition methods such as, micro-plasma, dielectric-barrier-discharge (DBD), electro-deposition, microwave resonator, electrolysis, chemical vapour deposition and tribo-chemical transformation methods to meet the requirements of new markets, which generally require DLC deposition at low temperatures and atmospheric pressures.

5.2 Microstructure of DLC coatings produced with non-conventional deposition methods

Fig. 4 illustrates the representative micrographs of DLC coatings made with non-conventional approaches. It can be observed that the non-conventional methods are also capable of depositing amorphous, granular, columnar, rough, and fibrous DLC coatings. The non-conventional methods, especially micro-plasma, DBD, and microwave resonators have a proven background for nanomaterial synthesis, but they have also shown good potential for surface treatment and DLC coatings deposition. Referring to the textiles industry, the usability of DLC coated textiles have been validated for antibacterial [129], vascular implants [130], and smart bandages [131]. Similarly, the formation of DLC coatings have been demonstrated with DBD discharge (Fig. 4A) [132-138], micro-plasma (Fig. 4B) [139-141] electro-deposition (Fig. 4C) [142-146], electrolysis (Fig. 4D) [147-149], plasma guns (Fig. 4E) [150, 151], plasma jet (Fig. 4F) [152, 153], microwave resonator (Fig. 4G) [154], and plasma torches [155]. Recently, fibrous DLC films were prepared with the chemical vapour deposition (CVD) method by growing densely packed DLC nanofibers (Fig. 4H) [156]. The DLC nanofibers are similar to carbon nanotubes but with amorphous features and could have potential applications in sensing, electronics, and biomedical domains. Another non-conventional area for DLC coating synthesis is operation-based rather than manufacturing. The DLC coatings are conceived (Fig. 4I) from carbon enriched dry, wet, gaseous [157] sources during tribology or under shear processes, when two surfaces slide against each other either at ambient conditions or at a certain pressure and temperature. Advanced simulations [158] have shown increasing C-C bond formation during sliding. This area has received increasing attention in the past few years and is being actively explored with different hydrocarbons, catalysts [159], environmental conditions and applications like self-lubrication in automotive [129], mechanical components [161], microelectromechanical systems [162], and medical products [163]. The idea has further advanced to treat pre-deposited DLC coatings with carbon enriched lubricants to gain superior lubricity by yielding graphene [164, 165] or other by-products [166] through tribo-chemical interaction.

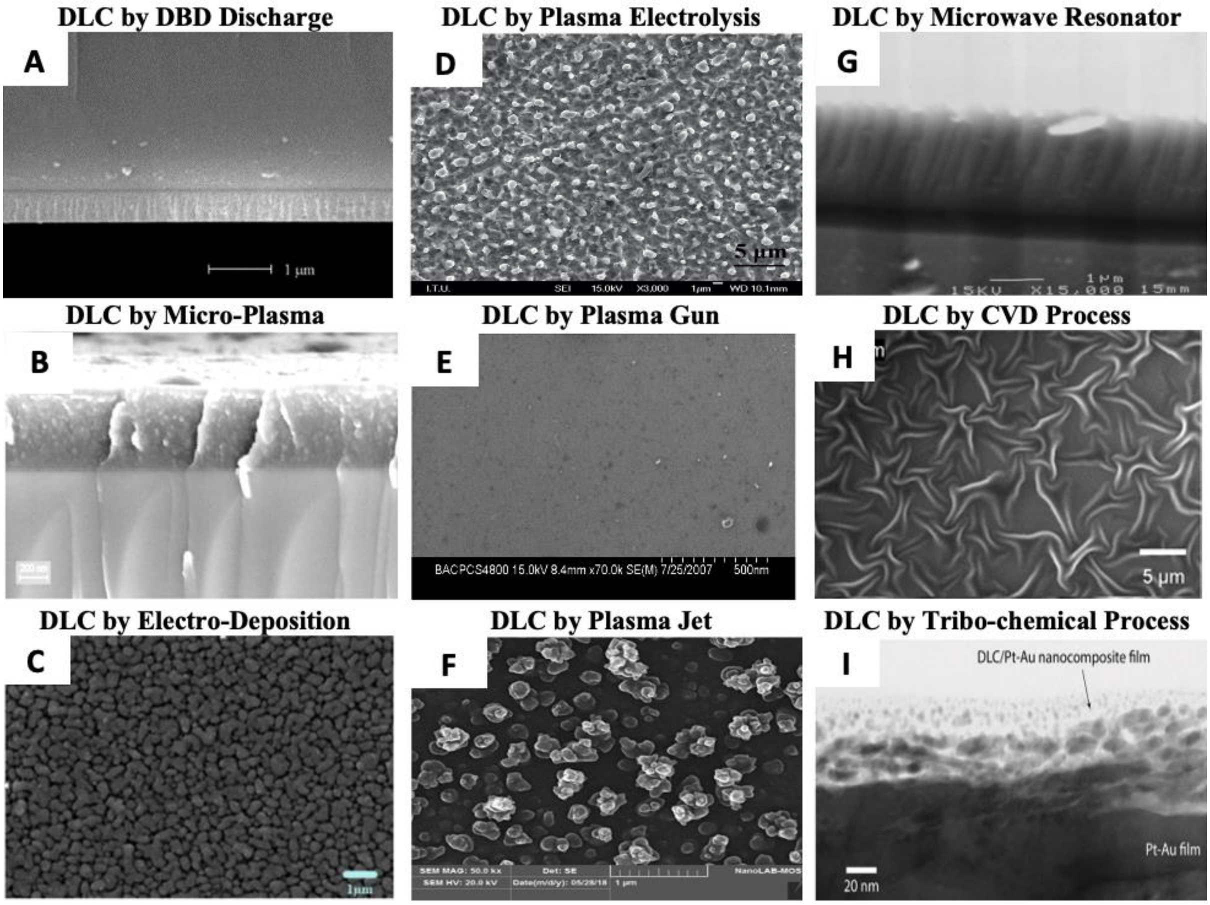

Figure 4 DLC coatings produced with non-conventional approaches. It can be observed that DLC coatings with numerous morphologies i.e., amorphous, granular, columnar, and fibrous can be produced with low-temperature non-conventional methods for emerging applications in textiles, electronics, and biomedical sectors. Reproduced with permissions: A [101], B [139], C [142], D [147], E [150], F [152], G [154], H [156], I [162].

5.3 Properties of DLC coatings deposited with non-conventional methods

Conventional methods mainly produce load-bearing DLC coatings. Generally, they have deposition rates of a few nanometres per minutes and their hardness ranges between ~ 15 GPa to ~ 35 GPa, Young's Modulus from ~ 170 GPa to ~ 300 GPa, while surface roughness and friction coefficients remain lower than 10 nm and ~ 0.1, respectively. In contrast, DLC coatings produced with non-conventional methods usually address non-load bearing applications. Most of the studies performed on non-conventionally produced DLC coatings cover plasma diagnostics, current-voltage relationships, self-cleaning, absorbance, reflectance, conductivity, biocompatibility features and rarely investigate mechanical and tribological applications. Non-conventional methods are not yet comparable for DLC performance, however, their features for DLC synthesis are being described for illustration. DLC coatings may have deposition rates as high as 6 µm/min with a hardness of ~ 6 GPa and Young's modulus of ~ 60 GPa when deposited with microwave resonator [154]. Similarly, contact angles as high as 167^{o} were observed when DLC coatings were deposited with electrospray assisted plasma jet [153]. DLC coatings made with DBD have demonstrated high hardness of ~ 20 GPa [132,133] and super-hydrophobicity up to ~ 160^{o} [134]. Whereas, DLC coatings made with DBD-based plasma gun have yielded a friction coefficient of 0.1 and good adhesion with a critical load of 31 N [151]. In general, electro-deposition of DLC coatings has resulted in a lower hardness of ~ 5 GPa, critical load of 40 mN, friction coefficients higher than 0.15 [142-146]. DLC fibres grown with CVD have hardness ~ 0.05 GPa, Young's modulus of ~ 1.5 GPa and contact angle as high as 159^{o} [156]. Tribo-chemical methods have shown super-lubricity with friction coefficients lower than 0.05 and hardness up to ~ 7 GPa [157-161]. Generally, the surface roughness of DLC coatings made with non-conventional methods remains between 5 nm to 20 nm. The mechanical, physical, and material properties of carbon coatings produced with non-conventional methods depend on the system and operational parameters. Table 2 summarize non-conventional methods used for DLC deposition along with common carbon precursors and core parameters. Most of the non-conventional methods, except tribo-chemical transformation, are sensitive to reactor design such as electrodes geometry, materials, and spacing between them. The outcomes depend on operational parameters which vary for different methods but commonly include electric power, frequencies, nature of precursor and its injection method, electric and magnetic potentials and similarly, working temperature and pressures. Whereas interfacial factors such as contact- pressure, area, and time essentially contributes to carbon film formation with tribo-chemical transformation.

Table 2. Non-conventional methods for DLC synthesis and their features - Analysis based on representative scientific outcomes

Sr. No	Non-conventional Methods for DLC synthesis	Common Precursors	Properties	Core Parameters	Representative Applications	Further reading
1	DBD discharge	Methane Acetylene	H ~ 20 GPa, CA > 150º, R ~ 0.1 nm, D ~ 26 nm/min	*System Properties* Electrodes configuration: geometry, orientation, distance, Material; Reactor design *Operational Parameters* Electrical power: AC/DC, magnitude, frequency, pulses; Precursor flow rate, Temperature, Pressure, Additional potential energies. *Material Properties* Precursor, Process gas, Catalyst.	Textiles, Smart Bandages, Sticky surfaces for robotics liquids analysis	[132-137]
2	Micro-plasma	Chloroacetic Acid, Acetic Acid, HydroxyEthylAmine Lactate, ACetoNitrile, Methanol, Ethanol, Acetonitrile	D ~ 20 µm/min		Inner surface of tubes	[138-141]
3	Electro-deposition	Methanol, Ethanol, Propanol, Acetone, Tetrahydrofuran, Acetonitrile	H ~ 5 GPa, Lc ~ 40 mN, COF > 0.15		Solar cell, Tribological Performance	[142-146]
4	Electrolysis	Methane	H ~ 22 GPa, R ~ 1.0nm Lc ~ 25N		Optoelectronic devices Fatigue life	[147-149] [167]
5	Plasma guns	Cyclohexane liquid	COF ~ 0.15, Lc ~ 31 N, R ~ 6 nm		Friction modifier	[150- 151]
6	Plasma jet	Ethane, Acetylene	CA ~ 167º		Cotton fabric	[152-153]
7	Microwave resonator	Acetylene	H ~ 1.5 GPa, E ~ 60 GPa, D ~ 7 µm/min		Roll-to-roll	[154]
8	Plasma torches	Acetylene	Lc ~ 19N		-	[155][168]
9	CVD	Isopropyl, Hexanes	H ~ 0.05 GPa, E ~ 1.5 GPa CA ~ 159º		Electric insulation	[156]
10	Tribo-chemical transformation	Methane, Acetylene	H ~ 7 GPa COF < 0.05	Contact pressure, Number of cycles, Catalyst	Lubrication	[151-161]

H: Hardness, E: Young's Modulus, CA: Contact Angle, D: Deposition Rate, R: Roughness, Lc: Critical Load, COF: Coefficient of Friction

5.4 Features of non-conventional methods for DLC deposition

The key features of the non-conventional family, particularly micro-plasma and DBD discharge is the capability to perform surface treatment, single-step synthesis of multi-nanomaterials, homogeneous coatings, and the potential to deposit a hybrid coating, i.e. embedded with nanomaterials or a layered structure. In a sequence, they can also perform post-treatment or post-nanofabrication on coatings within a single process. For example, the surface etching or nano-structuring of the DLC surface, which is emerging for self-lubrication application, can be performed in a single-step process. The DBD systems can efficiently coat inner surfaces [138] of narrow tubes having an internal diameter as low as 2 mm, which is not feasible with conventional PVD techniques. Intrinsically, these setups support small area or localized depositions. However, they have great flexibility to increase the projected area by adding arrays [169] of micro-plasma sources in parallel, according to the desired geometry. Hence, they can be practically implemented for large area deposition of coatings, or nanomaterial synthesis or surface treatment of textiles and other roll-to-roll technologies. Similarly, these non-conventional methods can play an important role in the electronic industry. Referring to electrical batteries, graphene-coated nickel foam saturated with paraffin has shown [170] improved performance, but since the paraffin has a lower melting point of ~ 60 ^{o}C, it is not possible to deposit it using conventional PVD systems. However, the micro-plasma or DBD discharge processes have the potential to deposit graphene and nickel composites on paraffin in a single step process while controlling temperature effects either with distancing or nanomaterials transportation. Similarly, graphene sheets [171] and carbon dots are receiving increasing attention in wearable [172], bio-imaging [173], and energy efficiency [174], thus these systems can do a selective deposition of surface plasmon, nanostructures, or homogeneous coatings made with the assembly of nanomaterials. In the same way, flexible plastics and polymer composites usually have rough and porous surfaces which may act as defects depending on the application. Thus, the surface performance of such polymers can be improved significantly with the application of DLC coatings. DLC coatings deposited with sputtering not only transform the hydrophilic surface (76^{o} contact angle) of polyester composites [175] into hydrophobic (~ 98^{o} contact angle), but also improve their friction behaviour and surface strength. However, not all polymers can withstand thermal depositions. Therefore, the significance of cold and atmospheric plasma increases for the deposition of DLC coatings for a wide range of polymers that cannot be produced with conventional PVD and PECVD systems. Thus, the non-conventional deposition methods have huge potential to grab a significant share of the growing DLC markets.

5.5 Future outlook of non-conventional methods for DLC deposition

The setup cost of non-conventional DLC deposition systems is lower when compared with conventional PVD systems. Normally a standard sputtering system costs at least \$250,000, whereas, a high specification micro-plasma or DBD systems cost between \$5,000 to \$10,000. The portability of micro-plasma and DBD instruments due to their small size and simple configurations is another key feature. Instruments are simple to build and can be configured on remote spots for coating or recoating of immovable structures. The non-conventional instruments such as micro-plasma, DBD, or microwave resonators have not received widespread attention for DLC deposition yet. There are a few challenges to be resolved for their adaptability. The physical outputs of non-conventional DLC deposition methods, such as amorphous material, macro, micro, or nanoparticles, grains, agglomerations, or dust formation are highly sensitive to the operational parameters. Therefore, precise control is essential to fabricate the desired physical and architectural design. Similarly, these methods coat a relatively small surface area per plasma source, therefore, they may yield gradient properties (like hardness) from source to source. A uniform distribution of nanomaterials flux is essential either for a single source or multiple arrays to deposit a uniform coating with homogeneous properties. In the same way, preliminary investigations with a microwave resonator [154] have reported high coating throughput of 4-7 μm/min but with a low hardness of 1.5 ± 0.3 GPa and Young's modulus of 60 ± 15 GPa. Such a high throughput is desirous, but the corresponding low hardness limits the usability for mechanical applications. However, the low hardness is suitable for textile, electronic industry or other applications which do not require load-carrying functions. Hence, intensive parametric investigations are required either around system design or operational parameters to improve the mechanical properties of DLC coatings deposited with non-conventional methods. Similarly, the standard protocol, ethics, and safety standard need to be established for DLC deposition by non-conventional methods since they involve potentially hazardous nanomaterials handling and high voltage electricity.

6. Concluding remarks

Diamond-like carbon (DLC) coatings are being deposited mainly with physical vapour deposition and plasma-enhanced chemical vapour deposition methods. Some of the established DLC markets, such as fuel engines, are shrinking while others like electronics, textile, biomedical, additive manufacturing etc. are evolving with the industrial transformation. Therefore, the conventional DLC deposition methods may not perform the same function for new products due to technical limitations, such as higher deposition temperatures of 100 ^{o}C to

300 ^{o}C, high vacuum requirements and limitations in bulk scale or roll-to-roll throughput capacity. Therefore, non-conventional methods should be investigated which can deposit good quality DLC coatings at low-temperature, atmospheric pressure with large scalability. Micro-plasma, dielectric-barrier-discharge, microwave resonators and plasma guns are potential non-conventional methods that can be used for such specialized purposes but they currently have higher deposition cost per unit article when compared to the well established conventional PVD and PECVD methods. However, these non-conventional methods provide more flexibility for the simultaneous or sequential process of surface treatment, nanomaterials/nanostructures synthesis, homogeneous coating deposition, and their combinations by materials or physical architectures. There are only a few published works to date that demonstrate DLC deposition with non-conventional methods, which can be assumed as a proof-of-concept. Therefore, methodological investigations with system properties and operational parameters are essential to evolve high-quality DLC deposition with non-conventional methods.

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Permissions Summary

Sr. No.	Figure number in this article	Figure number in source	Source	Status
1	2A	2	Y. Kang, B. Li, J. Zhao, B. Ge, M. Weng, Z. Shi b, Y. Zhao, Effect of structure on the secondary electron emission of tetrahedral amorphous carbon films, Vacuum 172 (2020) 109043.	Granted
2	2B	4a	V.S. Yadav, D.K. Sahu, M. Singhc, K. Kumar, D.C. Dhubkarya, Y. Singh, Characterization of Nano-Crystalline Diamond like Carbon (DLC) Films with Substrate Temperature Using Dense Plasma Focusing Method, AIP Conf. Proc. 1247 (2010) 363 373.	Granted
3	2C	3d	Y. Lei, J. Jiang, Y. Wang, T. Bi, L. Zhang, Structure evolution and stress transition in diamond-like carbon films by glancing angle deposition, Appl. Surf. Sci. 479 (2019) 12-19.	Granted
4	2D	2b	F. Li, S. Zhang, J. Kong, Y. Zhang, W. Zhang, Multilayer DLC coatings via alternating bias during magnetron sputtering, Thin Solid Films 519 (2011) 4910–4916.	Granted
5	2E	4b	H. Kinoshita, I. Ippei, H. Sakai, N. Ohmae, Synthesis and mechanical properties of carbon nanotube/diamond-like carbon composite films, Diam. Relat. Mater. 16 (2007) 1940–1944.	Granted
6	2F	2	C.Q. Guo, Z.L. Pei, D. Fan, J. Gong, C. Sun, Microstructure and tribomechanical properties of (Cr, N)-DLC/DLC multilayer films deposited by a combination of filtered and direct cathodic vacuum arcs, Diam. Relat. Mater. 60 (2015) 66–74.	Granted
7	2G	1c	W. Yue, C. Liu, Z. Fu, C. Wang, H. Huang, J. Liu, Effects of Tungsten Doping Contents on Tribological Behaviors of Tungsten-Doped Diamond-Like Carbon Coatings Lubricated by MoDTC, Tribol. Lett. 58 (2015) 31.	Granted
8	2H	8d	Y.T. Pei, D. Galvan, J.Th.M. De Hosson, Nanostructure and properties of TiC/a-C:H composite coatings, Acta Mater. 53 (2005) 4505–4521.	Granted
9	2I	3b	X. Xu, Y. Zhou, L. Liu, P. Guo, X. Li, K.R. Lee, P. Cui, A. Wang, Corrosion behavior of diamond-like carbon film induced by Al/Ti co-doping, Appl. Surf. Sci. 509 (2020) 144877.	Granted
10	3A	1e	D. Ding, X. Dai, C. Wang, D. Diao, Temperature dependent crossover between positive and negative magnetoresistance in graphene nanocrystallines embedded carbon film, Carbon 163 (2020) 19-25.	Granted
11	3B	1f		
12	3C	2A	A.W. Zia, Z. Zhou, L.K.Y. LI, Detailed study of structural, mechanical and tribological characteristics of ~100 nm sized carbon particles embedded amorphous carbon coatings, Surf. Coat. Technol. 357 (2019) 313-321.	Granted
13	3D	2E		
14	4A	2	D. Liu, S. Yu, Y. Liu, C. Ren, J. Zhang, T. Ma, Deposition of diamond-like carbon films by barrier discharge plasma with 1.4 and 20 kHz power sources, Thin Solid Films 414 (2002)163–169.	Granted
15	4B	2 (left)	R. Pothiraja, N. Bibinov, P. Awakowicz, Amorphous carbon film deposition on the inner surface of tubes using atmospheric pressure pulsed filamentary plasma source, J. Phys. D: Appl. Phys. 44 (2011) 355206.	Granted
16	4C	1e	Q. Zhang, Y. Wang, W. Wang, N. Mitsuzak, Z. Chen, Low voltage and ambient temperature electrodeposition of uniform carbon films, Electrochem. Commun. 63 (2016) 22-25.	Granted
17	4D	1a	A. Habibi, S.M.M. Khoie, F. Mahboubi, M. Urgen, Raman spectroscopy of thin DLC film deposited by plasma electrolysis process, Surf. Coat. Technol. 309 (2017) 945-950.	Granted
18	4E	4a	X. Bian, Q. Chen, Y. Zhang, L. Sang, W. Tang, Deposition of nano-diamond-like carbon films by an atmospheric pressure plasma gun and diagnostic by optical emission spectrum on the process, Surf. Coat. Technol. 202 (2008) 5383-5385.	Granted

19	4F	4c	A.A. Abbass, S,J. Kadhem, Preparation and characterization DLC thin films using atmospheric pressure plasma Jet, IOP Conf. Ser.: Mater. Sci. Eng. 454 (2018) 012065.	CC 3.0
20	4G	7b	H.C. Thejaswini, A.R. Hoskinson, B. Agasanapura, M. Grunde, J. Hopwood, Deposition and characterization of diamond-like carbon films by microwave resonator microplasma at one atmosphere, Diam. Relat. Mater. 48 (2014) 24-31	Granted
21	4H	1a	K.H. Lee, S.H. Lee, R.S. Ruoff, Synthesis of Diamond-Like Carbon Nanofiber Films, ACS Nano 14 (2020) 13663-13672.	Granted
22	4I	4a	N. Argibay, T.F. Babuska, J.F. Curry, et al., In-situ tribochemical formation of self-lubricating diamond-like carbon films, Carbon 138 (2018) 61-68.	Granted