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Optimizing diamond-like carbon coatings - From experimental era to artificial intelligence

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ABSTRACT

Diamond-like carbon (DLC) coatings are widely used for numerous engineering applications due to their superior multi-functional properties. Deposition of good quality DLC is governed by energy per unit carbon atom or ion and plasma kinetics, which are independent parameters. Translating independent parameters to dependent parameters to produce a best DLC is subjected to deposition method, technology, and system configurations which may involve above 50 combinations of bias voltage, chamber pressure, deposition temperature, gas flow rate, etc. Hence DLC coatings are optimized to identify the best parameters which yield superior properties. This article covers DLC introduction, the role of independent parameters, translation of independent parameters to dependent parameters, and discussion of four generations of DLC optimization. The first-generation of DLC optimization experimentally optimizes the parameter-to-property relationship, and the second-generation describes multi-parameter optimization with a hybrid of experimental and statistical-based analytical methods. The third generation covers the optimization of DLC deposition parameters with a hybrid of statistical methods and artificial intelligence (AI) tools. The ongoing fourth generation not only performs multi-parameter and multi-property optimization but also use AI tools to predict DLC properties and performance with higher accuracy. It is expected that AI-driven DLC optimizations and progress in virtual synthesis of DLC will not only assist in resolving DLC challenges specific to emerging markets and complex environments, but will also become a pathway for DLC to enter a digital-twin era.

1. Introduction of diamond-like carbon coatings

Diamond-like carbo (DLC) coatings are used for broad industrial applications as they possess high hardness [1], good wear resistance [2], low friction coefficient [3], chemical inertness [4], antireflection [5], biocompatibility [6], permeability [7], electrical insulation [8], and thermal stability up to ~ 400 °C [9]. DLC coatings are well adopted in automotive [10], aerospace [11], cutting tools [12], mechanical components [13], and optical devices [14]. Their market is continuously expanding for new applications such as fusion reactors [15], digital screening equipment [16], multispectral interference [17], biomedical [16,18] and dental [19] implants, energy storage devices [20] etc. Fig. 1 shows the number of scientific documents recorded on sciencedirect. com in past 25 years. The accelerated trend is attributed to their superior multi-functional performance and extended product life..In addition, the global revenue of DLC coatings was worth USD 1.7 bn [21] in

2020, which is estimated to increase by USD 2.6 bn [22] in 2027 at a compound annual growth rate of 6.2%.

The DLC coating is now more than 40 years old. Originally, the soft carbon coating was developed in Schmellenmeier Experiments in 1954 [23]. Whereas in 1970's, the diamond-like features emerged in carbon coatings after Eisenberg and Chabot experiments with the application of bias voltage [24]. Comprehensive studies have been performed on various aspects of DLC coatings either by deposition parameters [25], substrate system [26] testing conditions [27], and currently in continuation with doping [28], and microstructure [29] etc. DLC has been delivering superior performance for numerous mechanical applications such as fuel engines, cutting tools, die and molds etc. Whereas, new applications of DLC coating are now emerging in various sectors such as medical and health care [30,31], textiles [32], pipeline [33] etc. where DLC coatings are expected to perform in complex environments and working conditions. For example, load-carrying artificial orthopaedic
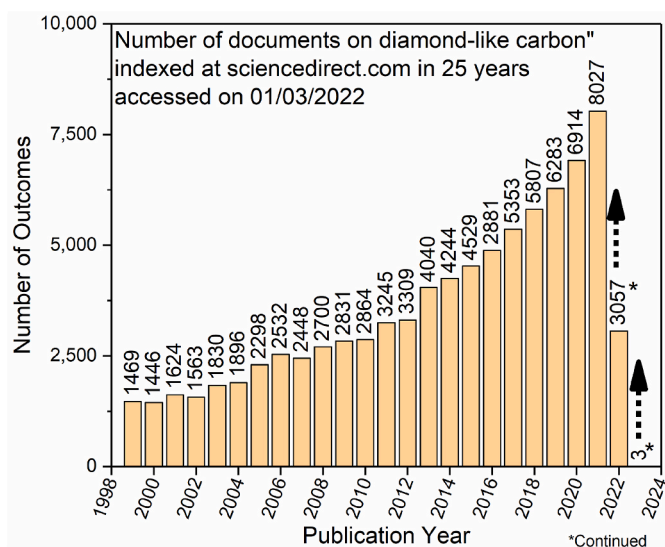
Fig. 1. Number of scientific articles on "diamond-like carbon" documented at sciencedirect.com in 25 years.

implants, where DLC coatings are supposed to simultaneously deliver a combination of at least mechanical, tribological, and biological properties. Hence, DLC coatings with multi-functional properties have a potential to improve product life and performance in numerous industries. Therefore, transforming from the typical application of fuel engines, cuttings tools, die and molds to new applications, especially working in a complex environment needs DLC optimization. Hence, the significance of optimization to reduce pre-deposition experiments, time, and resources still prevails in meeting emerging applications of DLC coatings.

1.1. Why deposition of good DLC is complex – role of independent parameters

Carbon coatings exhibit varying compositions of sp^2 and sp^3 atomic bonds [34] between carbon atoms, and this atomic composition categorizes them as diamond-like (higher sp^3 bonds) or graphite-like (higher sp^2 bonds) coatings [35]. DLC coatings are desirous with a higher sp^3 fraction that regulate atomic structure [36] and ensure dense coating [37] with high hardness [38], superior tribological behaviours, adhesion strength [39], the optical gap [40], haemolysis [41] and thrombus [42] regulation. DLC coatings are typically deposited with physical vapor deposition (PVD) [43] and plasma-enhanced chemical vapor deposition (PECVD) methods [44] where the carbon atoms received

from solid or gaseous precursors are ionized with energies provided to plasma (by electric, magnetic, etc. sources), and further bombarded at the substrate to grow a coating. The composition of sp^2 and sp^3 bonds regulate their physical [45], mechanical [46], thermal [47], electrical [48], optical [49], biomedical [50], etc and tribological [51] performance. The formation of sp^3 bonds and their proportion with sp^2 bonds in DLC coating are associated with the ion energy of carbon atoms. Referring to the theoretical model proposed by Robertson [52] in Fig. 2A and experimental results mapped by Hofsäss [53] in Fig. 2B suggests that the higher sp^3 fraction is obtained at a certain ion energy of carbon atoms, that is around 100 eV irrespective to hydrogen-free [54] or hydrogenated DLC coatings [45].

The carbon atoms are connected through single, double, and triple bonds and require disassociation energies between 160 and 230 kcal/mol. Hence, a sufficient amount of energy (such as sputtering power) is supplied to the system to produce plasma by disassociating process gas and carbon atoms. The plasma is the complex environment where carbon atoms are charged after receiving free electrons and transforming into carbon ions. Further, other types of energies are added to the plasma with an application of gas kinetic, thermal, magnetic, and electric potential. Hence, the carbon atoms and ions have a certain amount of energy which defines their behaviour inside plasma and their ability to grow coatings and induce specific characteristics. Referring to Fig. 2A, a theoretical model presents that once the carbon ions have an energy of around 100 eV, they have maximum potential to grow a diamond-like structure, irrespective of hydrogenated or hydrogen-free carbon coatings. Above or below the threshold value increases the probability of formation of the graphitic structure. Fig. 2B presents the experimental studies and a generic peak for ion energy could be observed, where the various deposition systems despite a diamond-like structure with the highest sp^3 bonds. The experimental studies also suggest that there is an optimum amount of ion energy which grows a diamond-like structure and sp^3 fraction reduced above or below that optimum ion energy. The formation of sp^3 bonds also increases due to structural densification up to a certain ion energy threshold and then starts declining afterwards due to relaxation [55,56]. The experimental investigations presented in Fig. 2B suggest that the optimum energy values for carbon ions are typically between 30 eV and 100 eV where highest sp^3 proportion is obtained. Jiménez et al. [57] demonstrate that the deposition rate sharply declines after increasing ion energy beyond threshold values and coating do not grow if energy increase beyond 2000 eV per carbon atom as the carbon atoms and ion either implanted in sub-surface or re-sputtered from the coating surface.

Ion energy is an independent parameter, and threshold energy values per carbon atom vary with deposition technologies, system design, and operational parameters. The optimized energy of carbon ions is recorded between 80 eV and 90 eV for pulsed laser deposition [58] and between
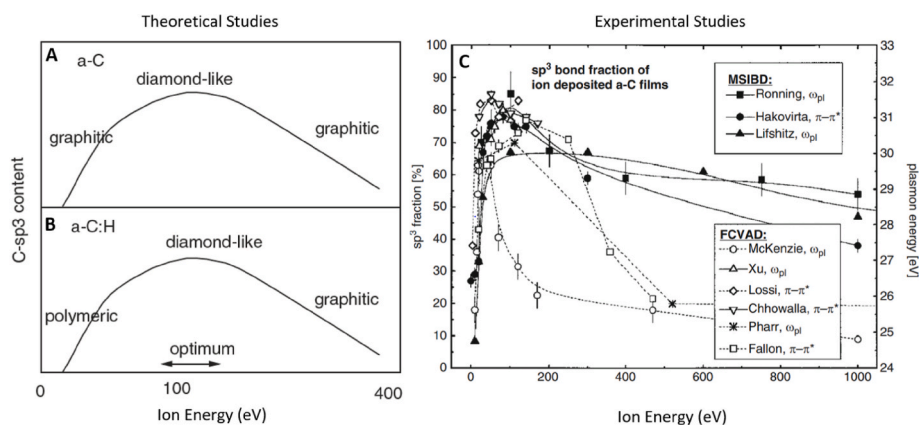

Fig. 2. (A) Theoretical model and (B) Experimental investigation suggest 100 eV as optimum energy per carbon atom to produce the highest sp^3 fractions in DLC coatings. Reproduced with permission. A from [52] Copyright 2008, Springer. B from [53]. Copyright 1969 Springer-Verlag.

100 eV and 1000 eV for mass-selected ion beam deposition [59] methods. Similarly, carbon atoms usually have ion energy of around 100 eV when DLC coatings are deposited with cathode arc systems. However, their optimized ion energy values could be lower as 30 eV [60] or high as 140 eV [61], depending on the system design, such as the usability of filter bends and the corresponding number of bends. Hence, the deposition of DLC coatings remained complex due to the random behaviour of instruments, methods, and operational parameters against ion energies of carbon which determine coating growth with sp^3 fraction and influence coating properties. Similarly, plasma kinetics, including mean-free-path, velocities, and moment of carbon atoms, are also independent parameters governing coating growth [62].

1.2. Translating DLC deposition from independent to dependent parameters

The independent parameters like the energy of carbon atoms and ions which govern DLC growth and corresponding properties are translated into dependent parameters. For example, ionization or disassociation energies required for plasma processing are an independent parameter that is translated with dependent parameters of electric power (DC/RF/Pulsed DC power of electrodes), magnetic fields, thermal environment, chamber pressure, electrostatic potentials like substrate bias. As suggested in Fig. 2, the carbon ions' energy should be around 30 eV to grow a higher sp^3 fraction which corresponds to the DLC structure. However, receiving 30 eV ion energy is a system and process-specific value that needs optimizing several parameters such as chamber pressures and pressure of process/reactive gases, bias voltage. In addition, the deposition technology such as arc, sputtering or laser, and the carbon source either a solid graphite target or various types of gaseous precursors also needs an optimized condition to assign 30 eV energy to carbon ions. Besides ion energy, the ion flux, mean-free path, projectile

velocity and momentum of carbon atoms in the plasma are also independent parameters. Referring to the dependent parameters, a bias voltage is one of the dependent parameters which significantly influence the coating growth. The other dependent parameters could be the megnatic field, deposition/substrate temperature, duty cycles, and plasma frequency used to translate independent parameters to dependent parameters. The above-described independent and dependent parameters develop several correlations among them. However, a relationship between ion energy and sp^3 fraction bonds and bias voltage and hardness are discussed here as a case study. Fig. 3 presents the translation of independent parameters, i.e., ion energy and sp^3 fraction, into dependent parameters, i.e., hardness and bias voltage. Fig. 3 A and B illustrate ion energy has a similar trend with sp^3 fraction and hardness. It can be seen from Fig. 3C and D that the bias voltage which governs ion energy also gives similar trends for sp^3 fraction and hardness. Practically, commercial coating machines and most research-grade deposition systems are seldom equipped with Langmuir probes and other sophisticated analyzers for real-time monitoring and then regulating ion energies in the desired spectrum. Hence, ion energies are controlled mainly through bias voltage. Applying a substrate bias voltage adds momentum and projectile energy of charged carbon ions and atoms and attracts them toward substrate forming a dense and disordered structure, where the probability of sp^3 formation increases due to physical and chemical interactions [63,64]. Since the bias voltage is analogous to ion energies, it also has optimum values after which the sp^3 bonds start reversing into sp^2 bonds [65]. Table 1 presents the independent parameters and translated to user/system-dependent parameters along with their potential to grow high-quality DLC coatings. The optimized dependent parameters yield a higher sp^3 fraction, which regulates hardness, toughness, friction coefficient, wear rate, residual stresses, adhesion strength, wettability, and other properties.
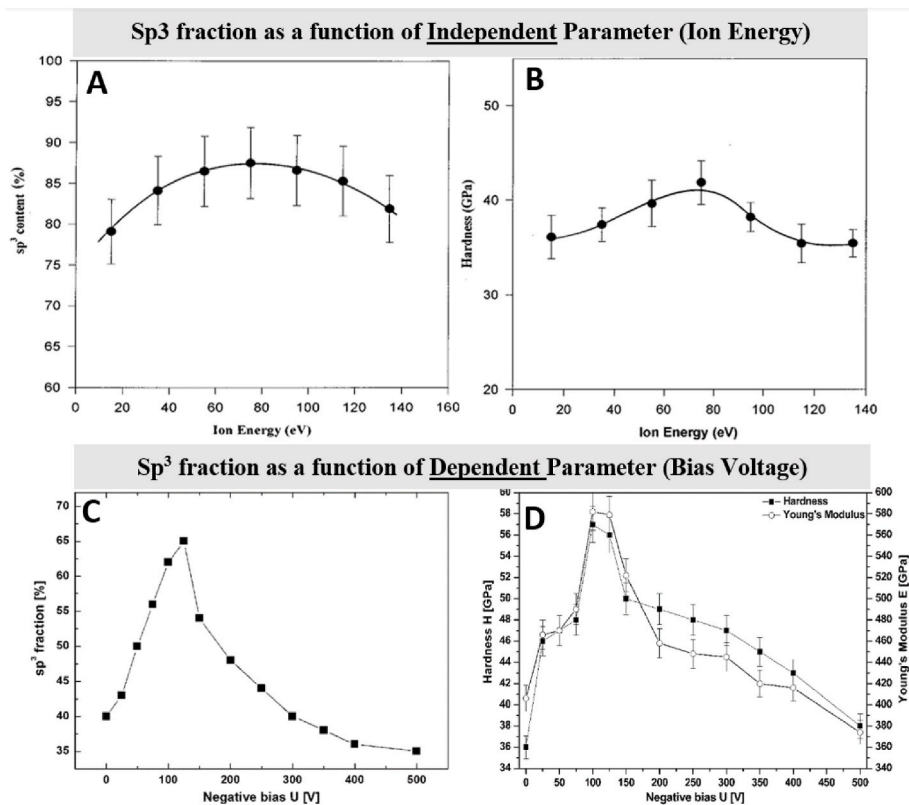
Fig. 3. Translating independent parameters i.e., ion energies, to a dependent parameter that is bias voltage. DLC coatings have similar trends of sp^3 fractions and hardness, which is demonstrated as a function of (A and B) ion energies as a independent parameter and (C and D) bias voltage as a dependent parameter. Reproduced with permissions. A and B fro [66], Copyright 1996, AIP Publishing. C and D from [64], Copyright 2021, Indian Academy of Sciences.

Table 1
Independent and translated dependent parameters to produce high-quality DLC coatings (i.e., with higher sp^3 bonds).

Independent Parameters	Dependent Parameters		*Further Study*
	Criteria	Significance of criteria	
Ion energy of carbon atoms/ions Ion Flux Mean-free-path of carbon atoms Velocity/momentum of carbon atoms	Electric Power	Producing carbon atoms/ions for coating growth	[67]
	Magnetic fields	Regulates degree of ionization and plasma density	[68]
	Plasma Frequencies	Regulates plasma kinetics and degree of ionization	[69]
	Target current/ voltage/bias	Regulates carbon atoms/ ions flux	[70]
	Substrate bias	Regulate momentum of carbon atoms	[71]
	Chamber pressure/gas flow rates	Regulate mean-free-path of carbon atoms	[72]
	Deposition temperatures	Regulates plasma kinetics and thermal migration of carbon atoms on coating	[73]
	Target-to-substrate distance	Regulates microstructural growth	[74]
	Stationary or rotating substrate	Regulates uniformity, porosity, and residual stresses	[75]
	Coating thickness	Regulates growth-dependent coating properties	[76]
	Buffer layer thickness	Regulates interface toughness adhesion	[77]

*Each parameter has a system-specific range of values. For example, a bias voltage may vary from 0 to 1000 V or 10,000 V for a certain system, where optimum values need to be identified to induce application-specific properties, such as Hard DLC or Soft DLC [78].

2. Material informatics and significance of optimizing DLC parameters

This section describes the generic role of material informatics and its significance for coatings and specifically for DLC coatings.

2.1. Overview of data analytics for material optimization

Industrial revolutions are classified from Industry 1.0 to Industry 5.0 at the transformation timeline [79]. Industry 1.0 starts from ~1784 with mechanistic approach, Industry 2.0 from ~1870 with electrification, Industry 3.0 from ~1970 with automation, Industry 4.0 from ~2011 with digitalization [80] and industry 5.0 from ~2021 for resilient, sustainable, and human-centric industrialization [81]. Correspondingly, an analogy [82] has developed between the Materials era and industrial transformation, where the bronze age (~3500 BC) transformed into Materials 1.0 as iron/steel age before the 20th century. Materials 2.0 started in the 20th century with broad coverage of prototyping, testing, life cycle assessments, and mass-scale manufacturing. Materials 3.0 emerged in 1970 with the advancement of information technology such as communication protocols, sensing, and automation which nurtured hypothetical materials research with conventional methodologies. Finally, materials 4.0 emerged in the 21st century with computation and digitalization age where big data governs the materials informatics for multi-scale, multi-agent (multi-parameters) modelling, virtual synthesis and testing of materials, virtual optimizations, and redesigning techno-economic analysis, and manufacturing procedures after all virtual validations by saving materials, energy, cost, recycling etc. resources. It is expected that Material 5.0 will foster data-driven materials

development and sustainable manufacturing by leveraging multi-disciplinary areas to enhance product life and performance while promoting United Nations Sustainability Development Goals [83]. The material informatics can help DLC coatings to meet Industry 5.0 demands. The DLC coatings deposited on basis of material informatics-driven optimization have a potential to enhance their properties, life, and performance. For example, optimizing DLC coating for durability, low friction coefficient and wear rates against high contact pressures foster their useable life. It will not only embark on the technological impact but also economic and environmental impacts due to saving of re-coating cost, recycling, waste management and circular economy.

2.2. Applications of data analytics for surface coatings

Data-driven materials analytics receives broad-spectrum data as inputs and process them with statistical and optimization tools, and recommend the best suitable parameters for desired outcomes i.e., manufacturing or performance. Materials 4.0 has boosted data analytics through artificial intelligence (AI), which comprises numerous techniques and methods such as machine learning (ML), artificial neural networks (ANN), fuzzy logic, genetic algorithms (GA), expert systems, inductive reasoning, evolutionary programming etc [84]. The data analytics under the umbrella of AI is actively supporting materials research such as underling material properties and mechanisms on the atomic scale [85], designing new bio-inspired materials [86], and shaping real world applications such as the development of energy storage devices [87] and fuel cells [88].

Likewise, artificial intelligence-based data analytics have been widely used for numerous coating systems since 2010 to predict and optimize their manufacturing parameters, performance, and properties. For example, ANN is being used to estimate deposition rate and phosphorus composition into nickel coatings [89], where the model was based on back-propagation-learning-algorithms and contained three layers of ANN. Similarly, ANN model is used to correlate hard chrome coating thickness with fatigue life of AISI 1045 steel [90]. In addition to this, the ANN model was used to optimize the cutting ability of PVD and CVD coatings as a function of micro-hardness, adhesion, grain size and coating thickness [91]. Likewise, thermal spray process parameters were optimized with multi-property genetic algorithm using the ANN model to optimize hardness, porosity, and cavitation erosion resistance of alumina-titanium oxide coatings [92]. ANN model was also used to optimize material type, loading conditions, relative velocity, and sliding distances to predict friction and wear behaviours of alloy coatings (carbon, chromium, tungsten carbide, nickel) and hard chrome coatings made with High-Velocity Oxy-Fuel (HVOF) [93]. Similarly, an ANN model was used to optimize HVOF deposition design such as coating material, type of combustible, pass number, standoff distance, combustible and oxygen flow, feed ratio, particles temperature and velocity and stoichiometry ratio to deposit binary coatings of desired features [94]. The complexity of the ANN model increases with the increasing number of hidden layers as the number of combinations between neurons residing in the anterior and posterior layers also increases. The range of validation data used in this regard becomes crucial. Furthermore, accurate and reliable models of optimization tools are desirous for accepting new data sets in parametric-space [93]. The literature also reports comparative studies to analyze the capacities of AI tools used to predict and optimize coatings. ANN and adaptive neuro-fuzzy methods have been applied for predicting the performance of intumescent flame retardant coatings [95] and have shown superior performance over the conventional Taguchi method by formulating higher mean fireproofing time. In the same way, fuzzy logics have been used to model and optimize the materials composition and corresponding deposition parameters such as deposition power, gas flow rates, temperature etc to estimate the adhesion strength of chrome-aluminium-nitride coatings [96]. The number of scientific

outputs indexed per year on sciencedirect.com for "artificial intelligence for coatings" was 68 in 2010, which increased to 1052 in 2021. This exponential trend reflects the significant role of artificial intelligence for coatings research and development. AI/ML based optimization tools are now emerging for DLC coatings and have shown good potential to optimize their deposition process and corresponding properties to uplift their performance, as detailed in the following section.

2.3. Data analytics-based optimization for DLC coatings

DLC coatings are mainly deposited with PVD and PECVD methods that more than 10 types of technologies, such as sputtering, arc methods, pulsed laser deposition, electron and ion beams, microwave plasma; their sub-variants and hybrids [97]. Each technique involves several parameters, mainly electric power inputs to the system, dimensional aspects, working pressures etc. which influence DLC growth and performance. Hence, the optimum bias that produces ~100 eV varies with system configuration and deposition method. Optimizing DLC properties and performance through optimization techniques is an integral development in this regard. However, the optimum value of a single dependent parameter, like bias voltage, does not deliver the best combination of DLC properties [98]. For example, hard DLC coatings have low friction and small wear rates but exhibit higher residual stresses [78]. Similarly, the hardness and toughness of DLC normally have inverse trends which limit their usability for high contact load applications. Prior study [99] suggests that DLC has the highest toughness and least hardness at 40 V bias while the highest hardness and poor toughness at 100 V bias voltage. There could be more than 50 combinations of parameters as described in Table 1 that need to be refined to make DLC coatings with optimum properties, i.e., achieving the best combinations of hardness, toughness, wear resistance, surface roughness, residual stress etc. or other electric, optical, biological properties depending upon the applications. The literature reports continued attempts to optimize DLC preparation and corresponding properties through experimental studies. However, the data analytics-based DLC optimization also emerged in the last decade mainly based on the Taguchi method and their combinations with fuzzy. Jean et al. improved the tribological performance of multi-layered DLC coating using the orthogonal array L18 test [100]. Similarly, Fang et al. have investigated the tribological behaviour of DLC coatings using adaptive fuzzy inference systems and presented good compliance of fuzzy with experimental results [101]. Recently, Solis-Romero et al. have used a hybrid of grey and fuzzy reasoning to optimize the working conditions of multilayer DLC coated AISI 52100 steel [102]. Similarly, metaheuristic models are recently used to study the hardness of DLC coatings and their accuracy is projected better than genetic algorithms [103].

3. Four generations of optimization for DLC coatings

The optimization of DLC coating can be classified into four regimes [104], i.e., inception regime before 2003, 1st generation between 2006 and 2010, 2nd generation from 2010 to 2014 and 3rd generation from 2014 onwards, as shown in Fig. 4. DLC coatings have been experimentally optimized for a single property such as hardness as a function of bias voltage, and this regime could be referred to as 1st generation single-parameter optimization. The 2nd generation deals with multi-parameter optimization performed with a hybrid of experimental and statistical methods that process more than one parameter like bias voltage and chamber pressure for a single property like hardness. Multi-parameter optimization for DLC coatings using statistical methods in conjunction with optimization algorithms was observed in the 3rd generation, where the Taguchi method [105] remained most popular for optimization studies. Multi-parameter optimization of DLC coatings is now evolving in an ongoing 4th generation optimization regime where data analytics and AI tools for multi-parameter prediction and optimization studies. The four generations of optimization for DLC coatings are
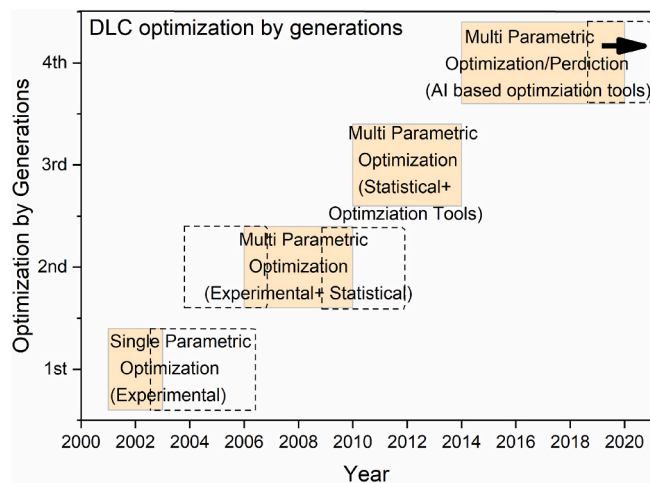

Fig. 4. DLC optimization regimes by generation.

described in detail in the following subsections.

3.1. 1st generation: Single parameter optimization of DLC coating - experimental

DLC properties are sensitive to deposition methods and parameters, system and environmental conditions. Therefore, these coatings have been optimized for 20 years (1980–~2000) primarily for single parameter-to-property relationships such as hardness, friction coefficient, wear rate, roughness etc., as a function of bias voltage. Fig. 5 presents the nano hardness of DLC coatings as a function of bias voltage [49]. It can be observed that DLC coatings derived from methane have given the highest hardness at ~200 V, whereas benzene-derived DLC coatings need more energy and have demonstrated the highest hardness at ~900 V bias. The associated reason could be dissociation energies of C–H, which are higher for benzene as 112.59 kcal/mol compared with methane, whose C–H bond dissociation energies are 105 kcal/mol. The optimum bias can also vary for Induvial DLC properties. For example, DLC coatings may have the highest hardness at bias voltage 100 [99], and they may also have the highest residual stresses at 100 eV bias [106] which promotes coating delamination. Hence, DLC optimizations have been performed for single-property relationship like friction coefficient, wear rate [107], surface roughness [108,109], and other properties as a function of coating thickness [110], bias voltage [111], deposition
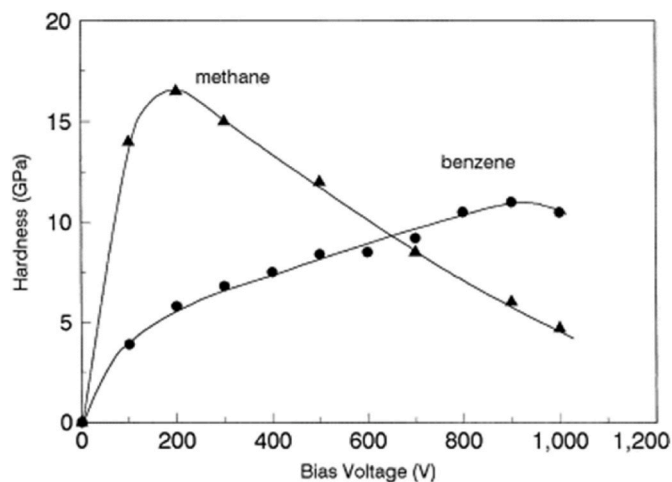

Fig. 5. Nano Hardness of DLC coatings deposited as a function of bias voltage with Methane and Benzene as carbon precursor. Reproduced with permission from [49], Copyright 2002, Elsevier Science B.V.

pressure [112] and temperature [113], gas flow rates [114] etc.

3.2. 2nd generation: Multi-parameter optimization of DLC coatings – hybrid: experimental and statistical methods

The progress in research and development of DLC coatings has demonstrated the significance of multiple deposition parameters such bias voltage, working pressures, flow rate of processing and reactive gasses, and likewise, buffer layer or coating thickness influence DLC properties. Pancielejko et al. [115], presented in Table 2, demonstrates multi-objective parametric studies of DLC coatings involving 64 combinations (4^3) of bias voltage, process gas, and DLC thickness and buffer layer thickness. Where, 64 combinations were reduced to 09 combinations with Taguchi method and further processed by depositing DLC coatings on those 09 refined parameters to identify 01 best value from each parameter, which has the potential to yield superior singular property. The benefits of 2nd generation multi-objective optimization were to identify best deposition parameters for a single property specific to certain applications such as hard DLC for mechanical components, wear resistant DLC for cutting tools, and DLC with good adhesion for optical devices etc. The same approach is also reported to optimize deposition parameters of DLC coatings doped with tungsten material for cutting tool applications [116]. More studies on multi-parameter optimization for DLC deposition specific to coating design and application can be found in the literature [117,118]. Where hardness, Young's modulus, and residual stresses were optimized as a function of gas flow rate, bias voltage, and annealing temperature [117] or deposition temperature and pressures for dielectric properties [118].

With progress in statistical methods, Jatti et al. [119] optimized deposition parameters with the Taguchi method in conjunction with ANOVA [120] and validated their outcomes with experiments. Their hydrogenated DLC coatings were made with inductively coupled plasma enhanced chemical vapor deposition (IC-PECVD). The parameters of interest in their studies were bias voltage and bias frequency, working pressure which influences plasma kinetics, and composition of CH_4 precursor gas diluted with hydrogen. The multi-variable optimization based on statistical methods [119] not only identified best parameters for carbon bonds arrangements (ID/IG ratios), hardness and Young's modulus; but also suggested that the ID/IG ratios are more sensitive to bias voltage while hardness and Young's modulus are more effected by gas compositions. Table 3 gives an overview of multi-parameter optimization with the Taguchi method. This optimization generation has started to develop parametric co-relation but remained limited to identifying a combination of inputs for a desired single output.

3.3. 3rd generation: Multi-objective optimization of DLC coatings – hybrid: Statistics and artificial intelligence

The second generation of optimization refines multiple parameters to deposit DLC coatings with the optimal values for a single property, such as hardness or wear rate. However, practically DLC coatings are required to possess a combination of properties for their multi-functional usage. Some representative examples could be DLC for engine parts where the coatings should be hard, frictionless, and thermally stable; DLC for orthopaedic joints where DLC should be biocompatible as well as hard and wear-resistant DLC for textiles where the coatings should be hydrophobic [121] and have good adhesion [122] etc. The 3rd generation uses a hybrid of statistical and AI tools to model and optimize DLC coatings. The idea is to search and identify the optimal combination of parameters that would provide a DLC coating with the best combination of properties. Yang and Huang [123] used a combination of Taguchi and grey fuzzy methods to optimize zirconium doped DLC coatings. Table 4 presents the combinations matrix (3^{-5}) which were optimized with Taguchi and Grey-fuzzy Taguchi methods using L18 orthogonal array of experiments. The optimization was performed around signal-to-noise ratios, grey relational coefficienct, fuzzy interface systems and grey relations grades with fuzzy. The outcomes suggest a combination of 5 parameters to make DLC with superior properties, i.e., less friction coefficient and wear rates, high deposition rate and turning DLC coating from hydrophilic to the hydrophobic regime. It is worth noting that the Grey-fuzzy Taguchi method optimized DLC parameters and give ~35% superior properties than the optimization performed with the Taguchi method only. Similarly, Grey fuzzy logic approach has been used to optimize DLC parameters of bias voltage, bias frequency, deposition pressure, and gas compositions which have reduced 3^4 combinations into 4 parameters [124]. It can be observed that the frition coeffiicient was 0.295 with out optimziation which reduced to 0.175 with Taguchi optimizaiton and further reduced to 0.112 i.e., a 62% decline with grey-fuzzy tagichi optmization. Similarly, the Grey-fuzzy Taguchi optimziations have identifies better wear rates, deposition rate and hydrophobic DLC surface having water contact anlge above 90°. This generation was able to suggest one combination of refined parameters out of several combinsations (3^5 or more) which can induce optimal DLC properties for the desired application. However, this generation lacks in predicting DLC properties.

3.4. 4th generation: Multi-parameter optimization and prediction of DLC coatings – artificial intelligence

ML and AI tools, particularly ANN and fuzzy logic, Gaussian process regression [125] in conjunction with other optimization techniques such as genetic algorithms and particle swarm optimization are emerging to optimize DLC parameters and properties. Fig. 6 presents the

Table 2
Multi- Variables parametric optimization of DLC coatings performed with 64 experiments. Data Adopted from Pancielejko et al. [115].

Inputs Inputs = 4^3					Optimization	Outputs Best values of 4 parameters that give superior DLC properties, individually.			
Sr. Parameter		Variables			Reducing 64 combinations to 09 with Taguchi method. Further Experimental optimization with 09 combinations	Sr. Parameter	Optimized variables for DLC Properties		
		1	2	3			*Adhesion*	*Hardness*	*Wear rate*
1	Bias voltage (V)	Floating	-20	-80					
2	Gas Pressure (Pa)	0.01	0.05	0.25		1 Bias voltage (V)	Floating	-80	-80
3	DLC thickness (μm)	0.9	1.4	1.8		2 Gas Pressure (Pa)	0.25	0.01	0.01
4	Buffer layer thickness (μm)	0.03	0.1	0.3		3 DLC thickness (μm)	1.8	1.4	0.9
						4 Buffer layer thickness (μm)	0.3	0.1	0.3

Table 3
DLC parametric optimization performed with statistical methods. Data adopted from Jatti et al. [119].

Inputs Inputs = $4^{\wedge}3$					Optimization	Outputs Optimized combinations giving best singular DLC properties.			
Sr.	Parameter	Variables			L9 ($3^{\wedge}4$) Taguchi orthogonal array, Signal-to-Noise ratio, ANOVA	Sr.	Optimized Parameters	DLC Properties		
		1	2	3				*ID/IG ratio*	*Hardness*	*Young's Modulus*
1	Bias voltage (V)	-50	-100	-150		1	-50V, 40KHz, 4 µbar, 60:40	0.18	-	-
2	Bias Frequency (KHz)	0.25	6.0	40.0		2	-100V, 6KHz, 4 µbar, 60:40	-	18.29	-
3	Deposition Pressure (µbar)	2.0	4.0	6.0		3	-50V, 6KHz, 4 µbar, 60:40	-	-	192.61
4	Gas Composition		60:40	80:20	90:10					

Table 4
Hybrid of statistical and artificial intelligence-based for multi-parameter optimization of DLC coatings. Data adopted from Yang and Huang studies [123].

| Inputs Inputs = L18($3^{\wedge}5$) orthogonal array of experiments					Optimization	Outputs Optimized combinations which give best DLC properties collectively				
Sr.	Parameter	Variables			L18($3^{\wedge}5$) orthogonal array of experiments, Signal-to-Noise ratios, Estimating grey relational coefficients, Fuzzy interface system, Grey relationship grades with Fuzzy	Sr.	DLC Properties	Progress in Optimziation		
		1	2	3				Without Optim. (Parameter series 1)	Taguchi Optim. (L18 Avg.)	Grey-Fuzzy Taguchi Optim. (70V, 0.6A, 90 KHz, 6 sccm, 90 mm)
1	Bias voltage (V)	40	55	70		1	Coefficient of Friciton	0.295	0.174	0.112
2	Target current Zr (A)	0.3	0.6	0.9		2	Wear rate $\times10^5$ mm^3/Nm	0.57	3.78	0.50
3	Frequency (KHz)	70	90	110		3	Deposition rate (µm/hr)	0.99	1.18	1.23
4	Gas flow (sccm)	3	6	9		4	Water contact Angle (deg)	83.17	84.29	91.70
5	Work distance (mm)	90	120	150						
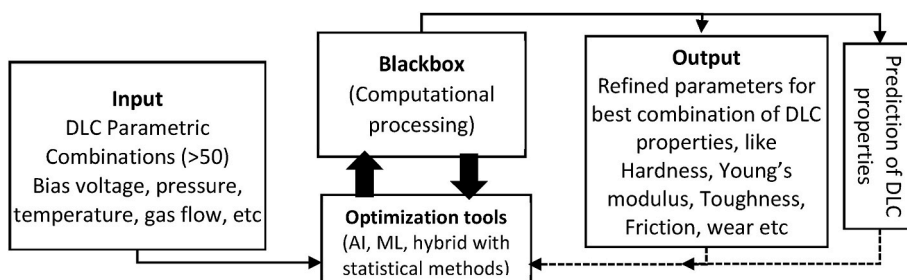
Fig. 6. Basic framework for optimizing DLC parameters, properties, and prediction using artificial intelligence tools.

fundamental understanding of DLC optimization through AI tools. The optimization outcome is the optimal set of parameters with the potential to make DLC with the desired set of properties. The optimal combination of properties refers to unique values which assure superior DLC performance. Such as, hard DLC has poor toughness, and tough DLC may not have high hardness, but their optimal combination may suggest tough-yet-hard DLC coatings. Modelling techniques such as ANN is used to predict friction behaviours of DLC coatings against engine oil additives utilizing the experimental data [126]. Sauer et al. [125] have recently reported the design of carbon coatings based on Gaussian process regression. Their data visualisation also advises on the relationships between bias voltage, sputter power, process and reactive has flow and indentation hardness. Similarly, gaussian approximation potential is used to investigate the deformation behaviours of carbon coatings [127]. Similarly, the experimental data is cross-validated with Fuzzy logic systems to analyze adhesion and the performance of

carbon/ceramic (Zr/ZrC/NC and Zr/ZrC/NZrC) multilayer coatings [128].

ML based optimization of DLC coatings has received increased attention in recent years. The GA in comparison with particle swarm optimization has refined the three best values gas flow rate, gas ratios, and deposition temperature out of $3^{\wedge}5$ that gives experimental hardness of PECVD deposited DLC [129] as 17.796 GPa, whereas the predicted hardness was 17.246 GPa. AI is being used beyond DLC optimization to predict DLC properties using the available data of parameters and properties. Fig. 7 presents the relationships between experimental and predicted hardness of DLC coatings from two different case studies, i.e., a statistical tool -central composite design-based response surface method (CCD-RSM) [129] and genetic programming (GP) [130]. Fig. 7A presents the relation between experimental hardness and the predicted hardness using the Central Composite Design (CCD) based Response Surface Method (RSM) that generates relationships between dependent
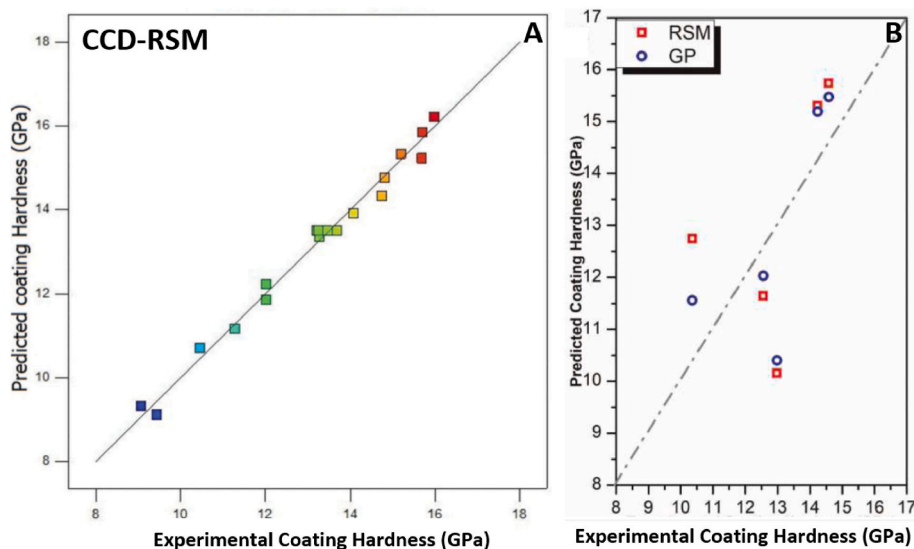
Fig. 7. Relationship between experimental and predicted hardness calculated with (A) Central Composite Design-based Response Surface Method (CCD-RSM) and (B) Genetic Programming. Genetic Programming further reduces variance, thus, improving the degree of optimization and accuracy in values. Reproduced with permissions. A from [129], Copyright 2018, Taylor & Francis. B from [130], Copyright 2021, De Gruyter Publisher.

and independent parameters [131]. The RSM predicted values have an average variance of 1.6% and a maximum variance of 3.4%. Whereas, Fig. 7B presents the relation of experimental hardness and the predicted hardness with RSM and GP. Fig. 7B also shows that the GP enhances DLC optimization and further reduces the variance between predicted values when compared with RSM. Similarly, GA has shown better metamodel prediction assessment up to 377% [130] higher than typical RSM optimization.

The ML/AI methods are evolving to predict multiple DLC properties and tribological performance [132]. For example, Fig. 8 presents experimental and predicted relations of Young's modulus and friction coefficient of PECVD derived DLC coatings [133], where DLC properties are predicted with a non-dominated sorting genetic algorithm in combination with the distance from the average solution (EDAS) technique. The DLC community is actively seeking such predictive data to develop next generation DLC coatings for emerging markets and complex working environments. Further case studies on DLC optimization with AI/ML methods can be found in the literature, which describes optimization and prediction of hardness and Young's modulus of DLC coatings with a genetic algorithm [134] and prediction of tribological behaviour of DLC coatings with the hybrid methodology of adaptive network-based

fuzzy inference systems (ANFIS) in conjunction with a genetic algorithm [135]. Furthermore [136], presented a multi-criteria decision-making approach to optimize the Zr-DLC deposition parameters using four different models.

4. Limitations and future aspects

Typical DLC remained popular for mechanical applications. However, DLC coatings are now emerging in new markets and applications such as plastics, textiles, electronics, and biomedical. DLC coatings have shown their potential for sophisticated medical applications such as dental implants [19] and vascular grafts [137], and vascular patches [138]. However, there is a need for new knowledge and tools to optimize DLC deposition and performance in compliance with new applications. Referring to foreseen challenges, the DLC coatings development is facing applications such as metallic orthopaedic implants to improve in-vivo corrosions resistance, bio-tribology, and biocompatibility. The industrial transformation has now introduced additively manufactured orthopaedic joints with the application of DLC coatings [139,140]. The optimization challenges will further increase with broader parameters, including new base-materials, non-conventional interface designs, and
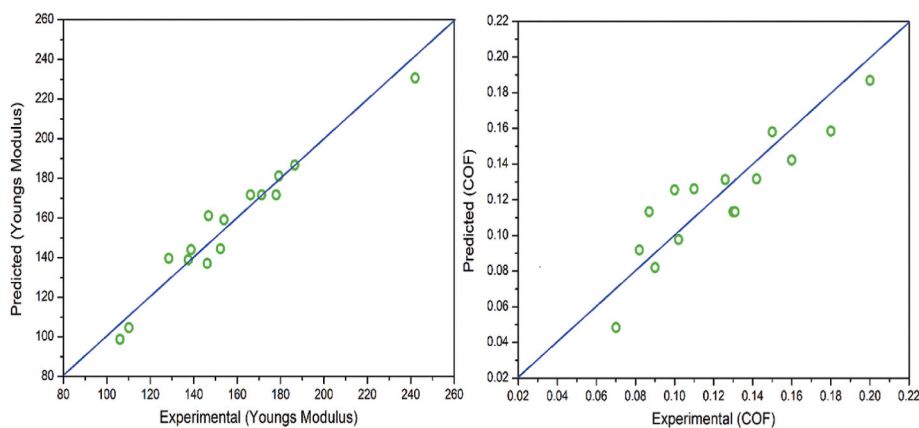

Fig. 8. Relations between Predict and experimentally measured (A) Young's modulus and (B) friction coefficient of DLC coatings made with PECVD method. DLC properties are predicted with genetic algorithm combined with distance from average solution method. Reproduced from Ref. [133], CC BY Vikram et al., 2020, Tech Science Press.

complex working environments.

The digital-twin has already evolved for coating applications [141]. The aspirations are to gain a better control over deposition system, process and anticipated results such as growth of coating architecture by thickness and structure, depositing complex morphologies [142], desired properties and performance to reduce pre-deposition experiments as much as possible. Digital-twin tools are emerging such as 'DXQ3D' by Dürr group or 'Digital Twin Coating Management Software' by Vidya Technology, Brazil for simulating coating thickness, in-situ quality assessment and interpreting results for automotive applications. Digital-Twin tools are helping to reduce maintenance [143], process time, scheduling etc. AI is well established for virtual synthesis [144,145] of other coating materials and predicting their properties and performance. Therefore, the progress in AI-based optimization of DLC coatings such as parametric optimization and properties prediction will help in reducing pre-deposition experimental and will become a pathway for DLC coatings to enter in digital-twin era.

A few things are to be critically considered while progressing with AI-based optimization of DLC coatings. We deduce that.

- The optimization is only as good as the model (developed with ML/AI/other methods) used for setting up the optimization problem. This is because the value of the parameter predicted by the model may not be accurate, which misleads the optimization outcome to suboptimal performance. Thus, model accuracy results in the reliability of decision-making with the optimization technique.
- Optimization models developed using ML/AI methods have an inherent deficiency since they rely on the range of data used for model training. The prediction performance of the models can be expected to be accurate, and that would translate into reliability with optimization action when the model is operated in a validated data range. However, outside this range of data, the model prediction will no longer be reliable.
- The importance of experimental data and utilization with advanced modelling methods becomes imperative for improving the reliability of decision-making with the optimization technique. Since DLC properties are influenced by deposition-technology, system, and parameters. Hence, DLC data is available in a broad range, but the availability of systematic and organized data is limited. The possible factors limiting the availability of experimental data is cost, time, materials, and other resources. It is expected that the Open Science campaign will make DLC data accessible across the world to foster AI investigations.
- Generic trends between DLC properties and parameters are well established experimentally in the past 30 years. However, advanced modelling methods are emerging for DLC coatings, while experimental studies of DLC coatings usually have very confined data points that could not be directly used to train AI models. Thus, training data sets are virtually produced by populating limited experimental data points. Therefore, there is a need to perform a reasonable amount of experimental studies in full spectrum to produce a significant and reliable learning dataset for validation of the model.
- The complexity of the AI model increases with the increasing number of input parameters, which accounts for the relation between parameters and their combinations. The user-defined functions also influence the outcomes. The model complexity affects the trainability of the model on the given data and reduces the model's generalizability for unseen data. In recent years, model complexity has been actively discussed for other applications and can be used to develop a reliable model for DLC optimization.

5. Conclusions

Diamond-like carbon (DLC) coatings are used for numerous industrial applications. DLC properties are governed by plasma dynamics, such as energy per carbon atom and ions, mean-free-path of carbon atoms and ions, their velocities and momentum etc. The said parameters cannot be directly controlled by users which make deposition of high-quality DLC coatings a challenging work. The above-described independent parameters are translated to dependent parameters such as bias voltage, magnetic fields, chamber pressure, gas flow rates, target-to-substrate distance, etc. which are specific to deposition method and configurations of the deposition system. Hence deposition parameters are optimized to make DLC coatings with superior properties. The input parametric of bias, target currents, chamber pressure, gas flow rates etc usually make above 50 combinations subjected to the system specifications. Therefore, there is a need to refine best deposition parameters using optimization tools. The first generation of DLC coatings deals with single property optimization as a function of single parameter variation. The second generation covers multi-parameter optimization with a hybrid of experimental and statistical methods where the combinations are significantly reduced with statistical methods, such as Taguchi method. Third generation improves statistical optimizations with integration of algorithms, such as Taguchi method hybrid with fuzzy logics to enhance optimization quality. The fourth, ongoing generation not only optimize DLC deposition parameters but also predict DLC properties using AI tools such as genetic algorithm artificial neural network etc. It is expected that AI based optimization will support in developing high performance DLC coatings for their application in complex engineering systems such as fusion reactors, robots, electric and extra-terrestrial vehicles.

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Data availability

All data associated with this publication is presented in the manuscript.

Consent for publication

All authors have consented the submission of this article to the journal.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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