Thin Film Deposition Techniques and Nanofabrication: A Comparative Analysis vis-à-vis Recent Advances

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Abstract: Present manuscript is a comprehensive analytical review of prevailing techniques used for thin film deposition in materials science. To keep this review abreast with latest advancement in the discussed field, about 60% of the referred literature belongs to last 5 years and about 85% of the referred literature belongs to last 10 years. The detailed comparative analysis of physical vapour deposition, chemical vapour deposition, atomic layer deposition, sputtering method and liquid-liquid-interface transfer methods is provided, focusing on their key parameters, advantages, and limitations. The applications of these techniques in semiconductor technology, photodetectors, emerging two-dimensional materials and heterostructures, and solar cells are also discussed. Furthermore, the role of machine learning in optimizing thin-film deposition processes is highlighted, and potential future research directions are outlined to address existing challenges and gaps in the field.

Keywords: Thin films, Deposition techniques, Nanofabrication, Semiconductor devices, 2D materials.

1. INTRODUCTION

The technique of coating one substance with a layer of another material in order to achieve certain well-defined objectives can be traced way back to the period of ancient history. For example, the use of plating techniques and plated materials to serve a wide array of purposes ranging from aesthetic ones like mere decoration to more consequential ones like reducing corrosion have been reported since the Etruscan, Greek and even the Iron age as evident from the antiquities of these eras [1,2]. With the advent of structured modern societies and ever-increasing degree of sophistication in prevailing technologies, that finds manifestation in various scientific developments, depositing thin-film of a material over some substrate has evolved as a technique to facilitate application of electrical, optical and other properties in a controlled manner. This makes the technique of thin film deposition a cornerstone of modern materials science and nanotechnology. In order to exercise greater control over the relevant properties of a certain material, the thickness of thin films needs to be carefully regulated. Thin film deposition techniques make it possible to modulate and control the thickness of the material being deposited with overwhelming precision spanning over a few nanometers to several micrometers. This feature of the thin film deposition technique that facilitates deposition of precisely controlled functional layers of materials over the substrate in question encompasses far-reaching applications in a wide range of processes and product-designs like integrated circuits, semiconductor devices, solar cells, sensors, and micro-electro-mechanical systems [3].

In order to keep pace with advancements related to miniaturization and novel functionalities of material properties in general and also adequately address requirements of the fields of electronics and optoelectronics in particular, the thin film deposition technique has evolved commensurately with time. The general umbrella term 'thin-film-deposition' encompasses a diverse array of techniques; each of which is unique in terms of characteristics like physical and chemical properties, advantages, limitations, time-taken for deposition, and also extent of control over thickness of the layer being deposited. The most common methods of thin film deposition of materials that find application in research and industrial settings include physical vapour deposition (PVD), chemical vapour deposition (CVD), atomic layer deposition (ALD) and sputtering [4-6]. The liquid-liquid interface transfer (LLIT) is a relatively recent addition to the list of deposition methods. Furthermore, the deposition mechanisms envisaged by these different techniques can range from purely physical processes like transport of material in vacuum to complex chemical reactions at the substrate surface and also to utilisation of effects like immiscibility and surface tension observed in different liquids [7-11].

Another factor that has significantly contributed to the remarkable progress registered by the thin film deposition paradigm can be identified as the recent advancement in domains like next-generation

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semiconductors, advanced photodetectors and energy conversion devices [4,12]. It is also expected that deposition techniques can be instrumental in achieving parameters like atomic-level precision and scalability of industrial production. This is also likely to result in development of novel hybrid materials such as transition metal dichalcogenides (TMDCs), metal oxides, and other complex heterostructures [4,13]. This paper analyses various thin film deposition techniques with reference to their fundamental mechanisms, comparative advantages as also limitations, and their applications in emerging two-dimensional (2D) materials and their heterostructures. The paper focuses on broad perspectives of deposition techniques with respect to energy devices, optical coatings, functional materials in general and semiconductor device fabrication, photodetectors and solar cells in particular. The paper draws on inputs based on an extensive review of recent literature in this area with the aim of highlighting prevailing state of art approaches and exploring prospective research possibilities.

2. FUNDAMENTAL PRINCIPLES OF THIN FILM DEPOSITION: AN OVERVIEW

2.1. Physical Vapour Deposition (PVD)

As evident from the name itself, PVD broadly refers to the technique of thin film deposition wherein the material is transferred from a source to a substrate through certain well-defined physical processes, generally carried out in vacuum conditions. The fundamental mechanism of deposition under this method sequentially involves following three stages beginning with vaporization of the source material, then upward transport of the vapour through the vacuum chamber, and finally condensation of vapour on the surface of the substrate which is placed at the top of the vacuum chamber in an inverted position [14-16] (Figure **1**).

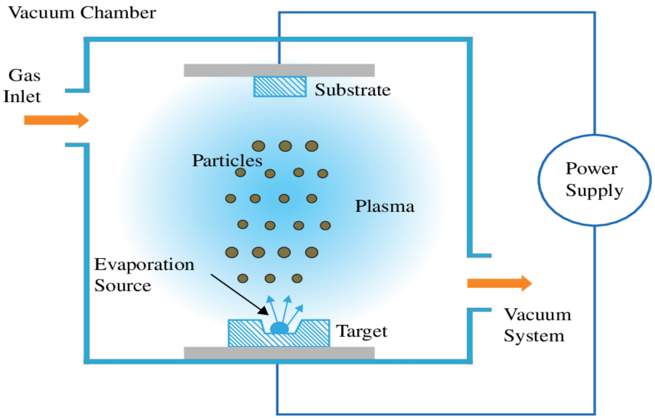


Figure 1: Schematic diagram of the fundamental mechanisms of physical vapour deposition (thermal evaporation). (Reproduced with due permission [16]).

The PVD technique relies primarily on physical processes like evaporation/sublimation, or momentum transfer knocking process to generate a suitable vapour-phase. Certain studies [14-17] suggest that the kinetic energy of the depositing vapour-phase significantly influences the morphological distribution of the thin film with respect to parameters like film density, adhesion, and microstructure. For instance, high -energy particles like ionized and accelerated atoms are observed to improve adhesion and density through increased surface mobility, ion implantation, and atomic peening effects. Conversely, low-energy deposition causes reduced atomic movement which in turn leads to shadowing effects, increased intra-column gaps, and creation of porous, column-like structures. Different methods like thermal evaporation, electron beam evaporation (E-beam), pulsed laser deposition (PLD), and molecular beam epitaxy (MBE) techniques [4-6,18-20] that are used for obtaining a suitable vapour-phase lead to different variants of PVD techniques.

Each of these methods encompass the following specific advantages [4-6, 18-20]:

- The thermal evaporation method is relatively simple in operation and cost-effective as compared to other methods. This technique is prominently employed for deposition on substrates in the nature of metals and simple compounds.
- The E-beam method is most effective for deposition of materials that have high melting points.
- The PLD method entails the advantage of being most effective in stoichiometric transfer of complex materials.
- The MBE method has the ability to deliver epitaxial growth with atomic-layer precision in case of deposition of materials with high melting points.

It needs to be noted here that selection of the most appropriate method of deposition from among those discussed above is ultimately based on a consideration of factors like desired film quality, properties of the material in question, and other requirements of the specific application. Furthermore, specific properties of the substrate also play a deterministic role in the choice of appropriate deposition method. For instance, an important parameter that needs to be considered for applying the PVD method is the temperature of the concerned substrate. This is because temperature of the substrate influences parameters like mobility of surface adatoms, nucleation density, and grain growth kinetics which in turn determine film morphology and crystallinity [14,17].

2.2. Chemical Vapour Deposition (CVD)

The basic difference between the PVD and CVD deposition techniques is that while the former focusses on physical processes, the latter depends on chemical reactions to form thin films. The modus operandi of the CVD technique revolves around introduction of gaseous precursors into a reaction chamber where they are subjected to chemical reactions like among others, thermal decomposition and reduction or oxidation aimed at deposition of the desired materials. [3,21-24]. This endows the CVD technique with certain unique advantages like conformity, control over composition, and the ability to accurately deposit materials such as complex compounds and alloys. The CVD process involves the following steps in a sequential manner-The process is initiated with transport of the precursor to the surface of the substrate followed by processes like adsorption of precursor molecules, surface diffusion, chemical reactions and film formation, and finally desorption of by-products generated by the chemical reaction [3,25,26] (Figure **2**).

It is important to note here that the overall rate of film-growth is constrained by the rate-limiting step which is observed to vary with variations in characteristics of the physical environment like temperature and atmospheric pressure, ultimately manifesting in emergence of different deposition regimes. For instance, it has been found that at low temperatures, parameters like surface reaction kinetics have a dominating role to play while parameters like mass transport are found to assume a deterministic role at high temperatures. It thus follows that key elements of the physical environment like temperature can be instrumental in choice of the appropriate process based on specific objectives of the application under consideration.

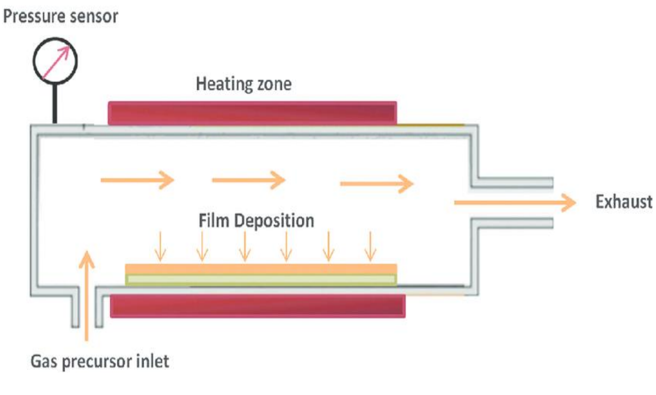


Figure 2: Schematic showing the fundamental mechanisms of chemical vapour deposition. (Reproduced with permission [26]).

Furthermore, it may be noted here that multiple variants of the CVD process have been developed to address specific requirements of different deposition processes which have notably enhanced the applicability of this technique. This is briefly discussed below-

The Plasma-Enhanced CVD (PECVD) method relies on specific properties of plasma substances to initiate/catalyse chemical reactions at lower temperatures which facilitates deposition on substrates that are sensitive to variations in temperature. [24].

The Metal-Organic CVD (MOCVD) method employs organometallic precursors for effecting growth of compound semiconductors. Such precursors hold particular significance in case of materials that belong to the III-V and II-VI categories [4].

The Low-Pressure CVD (LPCVD) is observed to operate with relatively greater efficiency under conditions of reduced atmospheric pressure which ensures enhanced levels of uniformity and conformality.

The Atmospheric Pressure CVD (APCVD) entails advantages like higher rate of deposition and relative operational simplicity of equipment required for using this technique [3].

The conformality of films deposited using the CVD technique serves as one of the most significant advantages of this technique. Unlike line-of-sight PVD methods, CVD techniques can be employed to coat complex three-dimensional structures like high-aspect-ratio trenches and vias essential for modern integrated circuits in a uniform manner [24]. The presence of this inherent capability can be attributed to the possibility of transporting precursors through gas-phase chambers. This property in turn makes deposition possible across all exposed surfaces regardless of structural and geometric complexities.

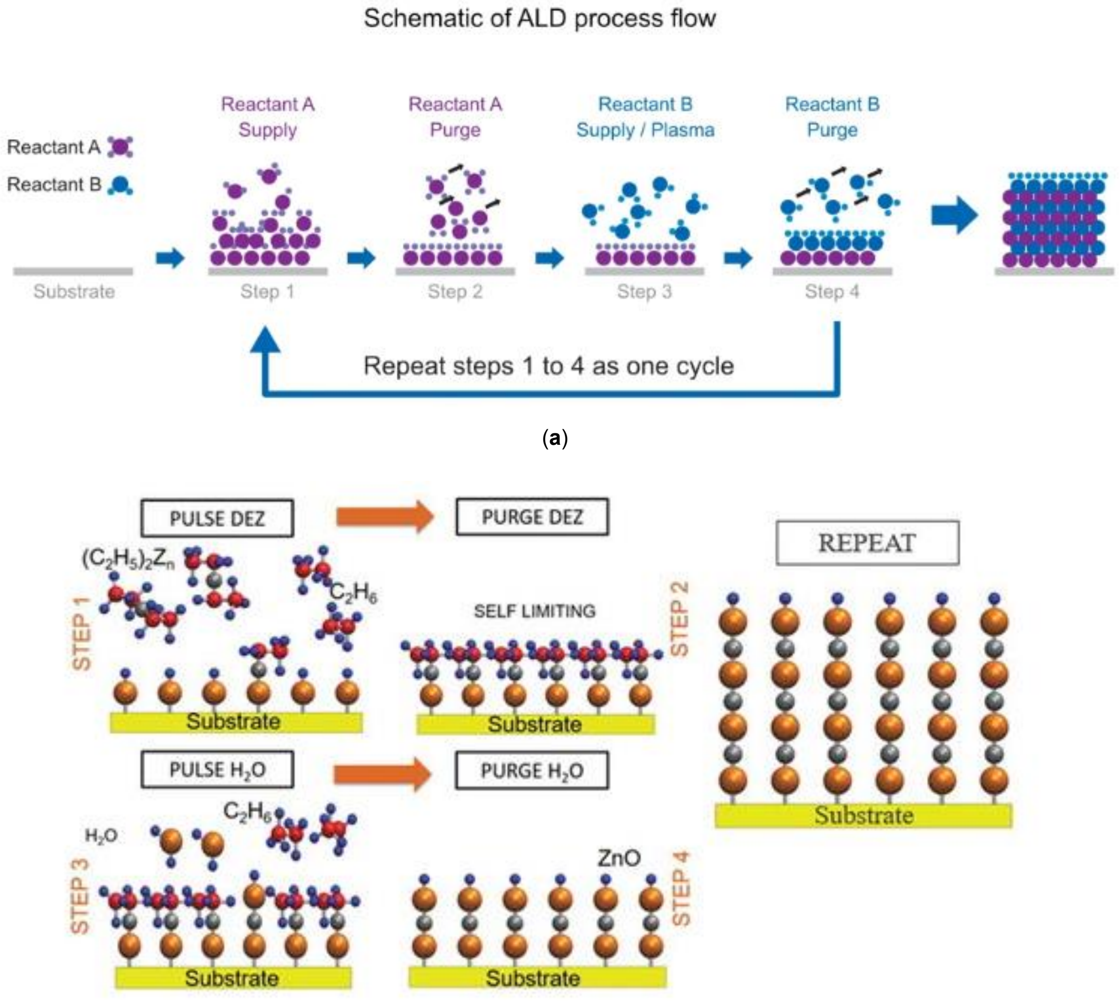
2.3. Atomic Layer Deposition (ALD)

The ALD technique is a precise method of vapour-phase thin film deposition that builds materials atom-by-atom (or monolayer-by-monolayer) using self-limiting surface reactions in a sequential manner. This makes the technique one of the most efficient methods of thin film deposition with respect to parameters like enhanced control over the deposition process. The greatest advantage of this technique is that it facilitates layer-by-layer growth of the thin-film material with atomic-level precision [3,12,13,27,28]. This technique encompasses unparalleled control over parameters like film thickness, composition, and conformality which distinguishes it from conventional CVD and PVD techniques. ALD may be described as a subset of the CVD technique wherein two precursors, introduced alternately to the substrate, are separated by inert gas purges in order to prevent gas-phase reactions [3,12,27,28] (Figure **3**).

The process of inert gas purging is critical for removing excess, unreacted precursors and by-products and thereby for ensuring thin-film deposition in a controlled manner. This step prevents gas-phase reactions and thus checks inappropriate growth, a feature common to application of CVD techniques. This in turn ensures growth of high-quality and conformal thin films with atomic-level precision and control. The ALD technique may be briefly described as follows- Consider a thin-film deposition process involving two precursors A and B. In the first part of the reaction, precursor A reacts with surface-level functional groups and forms a chemisorbed monolayer. The self-limiting nature of this reaction ensures that only one layer of the film is formed, regardless of the time duration of the exposure, more than a well-defined minimum threshold. At the next step purging is done with an inert gas following which precursor B is introduced allowing its reaction with the surface species that has evolved from the first part of the reaction to complete the deposition cycle. This binary sequence is applied repetitively to construct the desired level of thickness in the film with atomic-level precision [12,13,27,28]. This method provides exceptional conformality, uniformity, and thickness control, even on complex three dimensional nanostructures.

The self-limiting nature of ALD reactions encompasses several critical advantages that include the following-

- Since thickness of the film is determined solely by the number of deposition cycles, it enables precise control independent of variations in precursor flux and presence of non-uniform elements in the structure and geometry of the concerned reactor [3,12]. This characteristic also ensures remarkable conformality, even on structures involving extremely high-aspect-ratios as for instance ratios exceeding even levels of



(**a**)

(**b**)

Figure 3: Schematic showing the fundamental mechanisms of Atomic Layer Deposition (**a**) (Reproduced with permission [27]) and (**b**) (Reproduced with permission [28]).

1000:1; something that is unattainable with other techniques [12].

- The ALD technique enables deposition of ultra-thin films with thicknesses as small as a few angstroms, an essential requirement for advanced semiconductor devices whose dimensions are measured in terms of nanoscales.
- This technique facilitates not only deposition but also etching, surface modification, and area-selective deposition [13].
- Also, layer-engineered functional multilayer structures can be fabricated by alternating different ALD chemistries, creating heterostructures with precisely controlled interfaces and composition profiles [12].
- Molecular Layer Deposition, a variant of the ALD technique that uses organic precursors, enables the fabrication of hybrid materials encompassing organic and inorganic elements exhibiting properties tailored to meet the specific requirements of different deposition processes [12].

2.4. Sputtering Techniques

Sputtering refers to a PVD technique wherein atoms are ejected from a solid target material through bombardment of energetic ions, typically argon ions, generated in a plasma (Figure **4**) [29-31,32]. The simplest description of this deposition technique is that the sputtered atoms (that have ejected from the solid material post-bombardment by energetic ions) travel through the vacuum chamber and get deposited on the substrate, forming a thin film. This technique encompasses several advantages like the ability to deposit a wide range of materials on the substrate, excellent adhesion, and good control over film composition and properties in the deposition process [30].

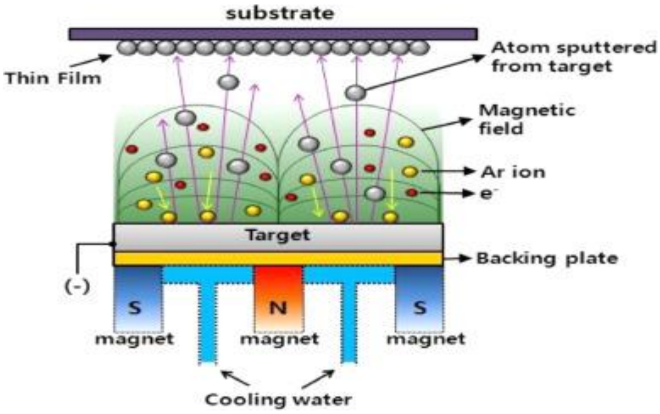


Figure 4: Schematic showing the fundamental mechanisms of sputtering method (magnetron sputtering). (Reproduced with permission [32]).

The basic operational mechanism of sputtering revolves around transfer of momentum from incident ions to target atoms. When an energetic ion strikes the target surface, it initiates a collision cascade in the region proximate to the surface. If atoms near the surface receive a level of energy that is sufficient to outweigh the energy level required to bind them to the surface, they get ejected into the gas phase [15]. It would be pertinent to note here that the sputtering yield, defined as the number of atoms ejected per incident ion, depends on factors like ion energy, mass of the ion, properties of the target material, and the angle of incidence. Sputtering yields are typically observed to range between 0.1 to 10 atoms per ion for commonly used materials and under commonly prevalent conditions.

Among the different variants of sputtering methods, magnetron sputtering is the most commonly used variant encompassing wide applications in industrial processes. In this configuration, permanent magnets are positioned behind the target material to create a magnetic field that seeks to effect ejection of electrons near the surface [29,30,32,33] (Figure **4**). The deliberate confinement of electrons increases ionization efficiency of the sputtering gas, enabling higher deposition rates at lower operating pressures. The enhanced plasma density near the target surface also notably improves quality of the film and also its level of uniformity. Recent advances in the realm of magnetron sputtering have sought to improve the technique's level of uniformity further, develop new target materials, and optimize process parameters so as to address requirements of specific applications of this technique [29,33].

The method of High-Power Impulse Magnetron Sputtering (HiPIMS) has emerged as a significant advancement in sputtering technology. This technique applies high power in short pulses (impulses) within a few microseconds at low duty cycle (on/off time ratio of < 10%) to the target, generating extremely dense plasmas with high degrees of ionization of the sputtered material [17]. Unique features of HiPIMS that distinguish it from other sputtering techniques include a high degree of ionisation of the sputtered metal and a high rate of molecular gas dissociation which essentially result in high density of deposited films. This high ionization fraction enables better control over the level of energy and direction of depositing species in turn resulting in formation of denser films that exhibit improved properties. This technique has been successfully applied in deposition of hard coatings, optical films, and functional materials with relatively greater efficiency as compared to conventional magnetron sputtering methods [17].

Reactive sputtering is another variant of the sputtering technique which extends its capabilities further by introducing reactive gases like oxygen and nitrogen into the sputtering chamber. The sputtered metal atoms react with the reactive gas to form compounds such as oxides, nitrides, or oxynitrides [30,31]. This approach enables the deposition of a wide range of compound materials while maintaining the advantages of sputtering at the same time. However, reactive sputtering mandates careful control of the reactive gas to prevent poisoning of the target material and also to ensure maintenance of stability in deposition conditions. Recent research in this area has focused on understanding and controlling the complex dynamics of reactive sputtering processes [30].

2.5. Liquid-liquid Interface Transfer (LLIT)

As evident from the name itself, LLIT method is a deposition technique wherein a thin-film of desired material is synthesized, rather self-assembled, at the interface of two immiscible liquids like for instance water and oil. This method also allows synthesis of 2D materials and ultrathin films using the boundary effects of immiscible liquids. Choice of the appropriate material and liquids needs to be made such that the desired material settles at the interface of the two immiscible liquids and is therefore critical to the efficiency of this method. This is shown in Figure **5** [34]. After the upper layer of the chosen liquid is removed either through evaporation or by flushing it out using a micropipette, the thin film is stamped on the substrate. The biggest advantage of the LLIT technique vis-à-vis other deposition methods is its cost-effectiveness. Furthermore, it also facilitates creation of an orderly range of nanocrystallines and enables transfer of functional materials between different phases. This technique relies primarily on two factors- first, exercising control over interfacial tension and second, leveraging on solubility of particles. The modus operandi of this technique involves transfer of 2D materials like Graphene, Molybdenum disulphide (MoS_2), Tungsten disulphide (WS_2), Indium selenide (InSe) from an aqueous surfactant-stabilized suspension onto a substrate by passing the material in question through the interface of two liquids resulting in formation of thin-films with high conductivity. This method is highly effective for creation of self-assembled, large-area, and high-quality 2D materials and nanomaterials at room temperature [11,34].

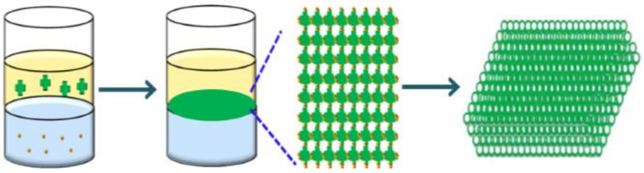


Figure 5: Schematic of liquid-liquid interface transfer method. (Reproduced with permission [34]).

Thienoacene-based organic small molecular semiconductors, such as DNTT and BTBT, have long been recognized for their exceptional air stability, high crystallinity, and impressive hole mobility. However, their insolubility in organic solvents has hindered their application in organic electronics [35-37]. In response to this challenge, numerous research groups have sought to enhance solubility by incorporating alkyl and phenolic groups into these small molecules. A significant contribution to this field was made in 2007 by Hideaki Ebata and his team, who conducted a comparative analysis of the solubility and electronic properties of n-alkyl derivatives of BTBT [38]. Their findings revealed that the dioctyl derivative exhibited outstanding characteristics, including high solubility and mobility, leading to the recognition of benzothiophene (C8BTBT) as a leading material for organic printed electronics [38]. Theoretical investigations have indicated a record high mobility of 180 cm²/Vs for C8BTBT [39,40]; however, experimental measurements have shown a considerable discrepancy, with reported mobilities ranging from 0.03 cm²/Vs to 43 cm²/Vs [41-44].This divergence between theoretical predictions and experimental results suggests potential overestimations of mobility or variations in the quality and nature of the films studied.

The discovery of graphene has led to the identification and processing of numerous layered crystalline two-dimensional materials for use in transparent and flexible devices using the LLIT method. However, the fabrication techniques employed remain either non-scalable, such as top-down mechanical exfoliation, or necessitate sophisticated instrumentation, which significantly increases costs [45]. In the context of bottom-up approaches, producing films of inorganic compounds across various substrates poses challenges in that only substrates with crystal lattices that align with those of the compounds can be utilized [46,47]. Consequently, there is a pressing need for a thin film fabrication method that operates at low processing temperatures, is compatible with a wide range of substrates, and is cost-effective. LLIT serves as an effective method to achieve these specific objectives. Researchers have explored alternative methods, such as liquid phase exfoliation, to create films on substrates. Various techniques exist for exfoliating bulk layered crystals or powders into solutions, including solvent-assisted exfoliation, alkali ion intercalation, ammonium ion intercalation, and polymer-assisted exfoliation, among others. Some of the initial studies on organic solvent-based liquid phase exfoliation using simple sonication methods date back over a decade [48,49]. While this approach has yielded highly pure monolayer to few-layer 2D nanosheets, it has been hindered by significant issues, including a wide size distribution, low yield, and extended processing times. Subsequently, evenly sized flakes enabled the formation of well-connected films through spin coating [50,51]. However, a significant limitation of this method was the difficulty in removing THA ions, which hindered the achievement of complete phase purity and consequently distorted the electronic properties. In contrast, liquid phase exfoliation, which produces smaller flakes, offers higher purity and has been widely adopted by researchers for the fabrication of solution-processed devices [52].

3. COMPARATIVE ANALYSIS OF DEPOSITION TECHNIQUES:

3.1. Process Parameters and Control

A comparative analysis of the thin film deposition techniques discussed above reveals some fundamental differences with respect to factors like process parameters, control mechanisms, and operational requirements in the deposition process encompassed therein. These differences in turn are observed to have a direct impact on characteristics of the end-product (the thin film) with respect to factors like film quality, reproducibility, and suitability for specific applications. Understanding the nature of these differences is of central importance in selection of the most optimal technique for a given application. This is discussed below.

3.1.1. Deposition Temperature

Deposition temperature is one of the most critical parameters for distinguishing between the suitability of various deposition techniques across different applications. For instance, CVD processes typically require elevated temperatures, often ranging from 400°C to 1000°C or even higher ones in certain applications, in order to initiate the required chemical reaction [3,53]. However, operating under conditions involving temperatures of such a high magnitude can be detrimental for substrates like polymers, flexible electronics, or devices with pre-existing structures that are temperature-sensitive in nature and are at a high risk of disintegrating at elevated temperatures. Using PVD techniques that generally operate at lower substrate temperatures, often below 200°C, could therefore serve as a better option in such cases [14,15]. In this context, the ALD method represents an intermediate position, with process temperatures typically ranging between 100°C and 400°C, even as recent explorations in this area have enabled operation of ALD processes at temperatures as low as room temperature for specific applications [13].

3.1.2. Pressure Requirements

Pressure requirements essential for application of different deposition techniques are also observed to vary significantly. For instance, PVD methods, like evaporation and sputtering methods, typically operate at pressures ranging from 10^{-6} to 10^{-2} Torr, and therefore require systems encompassing conditions of high-vacuum [15,54]. On the other hand, CVD processes can operate over a wider range of pressure as evident from the ability of the APCVD method to operate at relatively high levels of atmospheric pressure and that of the LPCVD method to operate at relatively low levels of pressure in the range of 0.1-10 Torr [3]. The ALD technique typically operates at a low level of pressure, similar to the LPCVD method, in order to facilitate precursor transport and purging [12,13]. The variable pressure regime entails major implications not only for factors like equipment requirements and operating costs but also for film properties like density, conformality, and incorporation of impurities.

3.1.3. Deposition Rate

The variable rate of deposition prevalent across different techniques is another crucial parameter for distinguishing between suitability of different techniques for specific applications. For instance, CVD and PVD methods typically encompass high deposition rates, ranging from tens to hundreds of nanometers per minute, enabling rapid film growth and high throughput [3,14]. On the other hand, deposition rates in case of sputtering methods depend on factors like power density, properties of target material, and substrate structures, but are generally observed to vary within a range of 10-100 nm/min in case of conventional magnetron sputtering [30]. In contrast, the ALD method is inherently a slow process, with typical growth rates of 0.1-1 nm per cycle and cycle times also ranging from seconds to minutes, with the net result that deposition rates can attain a maximum value of a few nanometers per minute [12,13]. However, it also needs to be noted that this slow growth rate is effectively the cost incurred for attaining atomic-level precision and conformality.

On the whole, the quality of films produced by these techniques depends primarily on factors like the choice of substrate, conditions of deposition temperature, range of temperature window, and properties of the precursor used [3]. CVD and ALD are observed to encompass superior control over film composition and stoichiometry, given their ability to exercise precise control over precursor ratios and reaction conditions [3,12]. PVD techniques entail advantages with respect to deposition of pure elemental films and simple alloys but confront challenges in maintaining stoichiometry for complex compounds [14,15]. In this scenario, the sputtering technique, particularly reactive sputtering, serves as a middle ground by enabling deposition of compound materials while simultaneously maintaining good control over composition [30,31].

3.2. Film Quality and Properties

Film quality encompasses multiple attributes like uniformity, conformality, density, crystallinity, composition, and defect density. Different deposition techniques exhibit variable strengths and weaknesses across these quality metrics, rendering the choice of an optimal technique dependent on specific requirements of the application in question. Conformality, that may be commonly defined as the ability to coat non-planar surfaces and high-aspect-ratio structures in a uniform manner, may be identified as one of the most significant parameters for distinguishing between different techniques. Given the unique feature of the ALD technique to self-limit the magnitude of surface reactions, it provides unparalleled conformality as evident from its ability to achieve uniform coating of structures with aspect ratios exceeding 1000:1 [3,12]. Although not as efficient as the ALD technique the CVD method also encompasses a high degree of conformality and depends on factors like the specific CVD variant being applied and the nature of the process conditions [24]. In contrast, PVD techniques are generally found to be line-of-sight processes which result in poor levels of conformality, particularly in case of complex three-dimensional structures [14,15]. The sputtering technique is capable of achieving a higher level of conformality in comparison to evaporation processes given its inherent ability of gas-phase scattering, but cannot attain the same level of conformality as CVD or ALD for coating high-aspect-ratio features [30].

Film density and microstructures are also found to significantly influence the mechanical, electrical, and optical properties. Sputtering and other deposition techniques involving high energy levels typically produce denser films as compared to thermal evaporation methods due to the higher kinetic energy present in the depositing species [15]. The energy distribution of sputtered atoms and ions is observed to play a critical role in determining film density, with higher energy particles found to be capable of producing denser and more adherent films [15]. Films produced by the CVD technique can achieve high density if deposited at appropriate temperatures and pressure levels. However, it needs to be noted here that even though CVD processes applied at low temperatures produce films with less density, they encompass a high level of hydrogen content [24]. Films produced by the ALD technique are typically very dense due to operation of the layer-by-layer growth mechanism and the chemical nature of the deposition process [12].

Crystallinity and grain structures are also found to vary significantly across deposition techniques and conditions. For instance, CVD processes conducted in conditions involving high temperatures are found to produce highly crystalline films with large grain sizes while CVD processes conducted in conditions involving low temperatures are generally found to yield amorphous or nanocrystalline materials [3,24]. PVD techniques are capable of producing a wide range of films that can be amorphous or highly crystalline depending on factors like temperature of the respective substrate and the relative rate of deposition [14,15]. Films produced by the ALD technique are generally amorphous in nature on completion of the deposition, primarily due to the relatively low process temperatures involved. However, it needs to be noted that these films can be subsequently crystallized through post-deposition annealing or by elevating deposition temperatures within the ALD window [12,13].

Impurity incorporation and film purity are other important areas of concern related to many applications, particularly with respect to semiconductor device fabrication. PVD methods are generally found to produce films with a high level of purity since they do not involve chemical reactions that can generate unwanted elements [14,15]. However, it also needs to be noted here that residual gases in the vacuum chamber can subsequently get incorporated into the growing film thereby undermining the initial level of purity achieved. Films produced by the CVD and ALD techniques are generally observed to contain impurities generated by decomposition of the precursor, typically involving elements like hydrogen, carbon, or halogen species depending on the chemical composition of the precursor [3,12]. It therefore follows that selection of an appropriate precursor alongside process optimisation is essential for minimising impurity levels in films produced through chemical deposition techniques.

3.3. Scalability and Industrial Viability

Factors like scalability and viability in industrial applications are important considerations for evaluating the relative efficiency of different deposition techniques. These elements in turn depend on multiple factors like equipment cost, throughput, process complexity, compatibility with materials, and scalability to large substrate sizes. In this context it would be pertinent to note that the CVD and sputtering techniques are generally understood to be workhorses of industrial thin film deposition due to their inherent advantages like reasonable deposition rates, good film quality, and proven scalability [3,30,53]. On the other hand, CVD techniques have been extensively deployed in manufacturing of semiconductor devices, particularly for depositing dielectric layers, polysilicon, and epitaxial films [3]. The sputtering technique is observed to dominate the deposition of metal interconnects, barrier layers, and optical coatings across industries engaged in manufacture of microelectronics and architectural glass [30,33]. Both these techniques have been successfully scaled to handle large substrates like 300 mm semiconductor wafers and multi-meter glass panels and solar cell applications.

The ALD technique has transitioned from being a subject limited to research curiosity to emerging as an essential manufacturing technology for manufacturing advanced semiconductor devices. Industrial ALD systems address the limitation of being inherently slow through introduction of batch processing, spatial ALD configurations, and optimized time-cycles. However, despite these modifications at the operational level, it needs to be noted that throughput remains lower as compared to that attainable through application of CVD or PVD techniques [13]. Equipment costs and complexity are found to vary significantly across different deposition techniques. For instance, simple thermal evaporation systems can be identified as the most economical option while sophisticated ALD, MOCVD, and advanced sputtering systems require substantial capital investment [3,12,30]. As far as operating and maintenance costs associated with consumables like precursors and target materials are concerned, it may be noted that CVD and ALD precursors can be expensive. Likewise, target materials deployed in sputtering techniques entail significant costs of a recurring nature [12,30].

Process complexity and control requirements are other important factors that are found to influence industrial adoption of different deposition techniques. For instance, it is generally observed that the ALD technique requires precise control over precursor pulsing, purging, and timing, which mandates the use of sophisticated process control systems [12,13]. On the other hand, factors like the rate of precursor flow, temperature profiles, and pressure need to be carefully managed in the use of CVD processes in order to produce films encompassing desired properties and uniformity levels. [3,24]. The effective deployment of sputtering systems requires control over power, pressure, and gas composition. Furthermore, in case of reactive sputtering additional measures need to be taken in order to prevent target poisoning and hysteresis effects [30,31].

The detailed comparative analysis of different deposition techniques in terms of their relative advantages and disadvantages discussed above can be summarised in the following Table **1**.

Table 1: Comparison of Different Methods of Thin-film Deposition

Method → Feature ↓	Physical Vapour Deposition (PVD)	Chemical Vapour Deposition (CVD)	Sputtering Method	Atomic Layer Deposition (ALD)	Liquid-Liquid Interface Transfer Method (LLIT)
Process parameters	High Vacuum, Thermal/ E-beam, Temp: Low (<200°C)	Vacuum/ Atmosphere, Gas flow, High Temp: 500-1100°C	High Vacuum, Plasma, Target voltage, Temp: 350-600°C	Vacuum/ Flow, Sequential Precursors, Temp: Moderate/ Low	Room Temperature, Atmospheric Pressure, Solution chemistry
Control of parameters	Limited thickness control, manual, lower precision	Good for thickness; high temperature control needed	Good for alloy composition and thickness	Exceptional atomic-level precision (self-limiting)	Precise stoichiometric control in wet phase
Quality of film	Moderate, line-of-sight, lower density, potential impurities	High quality, excellent stoichiometry, good step coverage	Good, high purity, good adhesion, dense coatings	Highest quality, pinhole-free, very dense, uniform	High-quality, thin monolayers, often amorphous/ low-defect
Scalability	Moderate (limited by vacuum chamber size)	Good, widely used in industry for mass production	Excellent, high for large area substrates	Low to Moderate (Slow process), batch-dependent	Excellent, capable of large area, low-cost
Industrial viability	High for optics, decorative, cutting tools	Very high, standard for semiconductors, hard coatings	High, standard for electronics/ semiconductors	High (for high-end logic/ memory)	Moderate, emerging in flexible electronics/ solar
Specialities	Excellent adhesion of metallic layers	Excellent conformal coating on complex geometries	Superior for non-conductive and oxide targets	Unmatched conformal coverage on 3D structures	High-purity, low-cost synthesis of nanostructures
Advantages	Low temp, fast, low damage (thermal)	High deposition rate, versatile material, good for complex shapes	Uniformity, wide material choice, good for Large Area	Perfect conformality, uniform, precise thickness, pinhole-free	Low cost, low temperature, simple setup
Limitations	Poor step coverage, line-of-sight, high temp	High temp, toxic precursors, high power usage	High target cost, lower rate than evaporation	Slow, high cost, limited precursor availability	Long processing time, sensitive to impurities

Following are some key takeaways that can be inferred from the above table-

- Best for low-cost/ flexible substrates: LLIT (wet chemical techniques) since they are found to be suitable for application under conditions involving low temperature levels and large areas.
- Best for complex 3D structures: ALD is found to encompass best conformal coverage, followed by CVD
- Best for high-quality (comparatively 'thick') films: CVD is generally preferred in such cases given its suitability to processes requiring high-strength and hard industrial coatings
- Best for high-rate metallic coatings: Sputtering/ PVD is considered ideal in such cases given qualities like speed and adhesion associated with these techniques.

4. APPLICATIONS OF DEPOSITION TECHNIQUES

Deposition techniques encompass multiple applications in diverse technology-intensive fields, some of which are discussed below:

4.1. Semiconductor Device Fabrication

Thin film deposition techniques have enabled the creation of complex multilayer structures with dimensions as minute as those that can be measured using only nanoscales which endows them with a deterministic role in modern semiconductor device fabrication. The continuous scaling up of semiconductor devices in accordance with the basic postulations of Moore's Law has emerged as a strong driver of innovations in deposition technologies, with different techniques finding specific niches based on their unique capabilities. Modern fin-field-effect-transistors and gate-all-around transistors require conformal deposition of high-k dielectrics, metal gates, and spacer materials on complex three-dimensional structures which has been made possible by thin-film deposition techniques [3]. Likewise, the ALD technique has become indispensable for deposition of high-k gate dielectrics such as HfO_2 and ZrO_2, which has replaced SiO_2 in order to reduce gate leakages while maintaining capacitance [12,13]. The unique properties of ALD enable uniform coating of the intricate structures of modern transistor architectures. Interconnect metallization is yet another critical domain where multiple deposition techniques are simultaneously deployed to achieve synergetic outcomes. For instance, copper interconnects, which replaced aluminium ones during the late 1990s, require barrier and seed layers in order to prevent copper diffusion and enable subsequent electroplating [3]. PVD sputtering is widely used to deposit tantalum (Ta) and tantalum nitride (TaN or Ta_2N) barrier layers, as well as ultra-thin (typically ~50nm) copper seed layers [30]. The transition to smaller feature sizes at the application-level over the years has undermined the usefulness of the conventional PVD-based deposition methods, which has generated interest in alternative techniques like CVD and ALD [12,13].

Epitaxial growth of semiconductor materials is another specialized application where deployment of CVD techniques, particularly MOCVD and MBE methods, have been found most suitable. These techniques enable the growth of single-crystal semiconductor layers with precise control over elements of the deposition process like composition, doping, and thickness [4,54]. Specifically, the MOCVD method is being used extensively for fabricating compound semiconductor devices such as light emitting diodes, laser diodes, and high-electron-mobility transistors based on III-V group materials [4]. Since the MBE method encompasses a higher degree of control as compared to other deposition methods, its applicability is higher in fabrication of more sophisticated designs like quantum wells, superlattices, and other heterostructures that entail atomically abrupt interfaces [54].

4.2. Photodetector Technologies

Photodetectors are another critical domain where thin film deposition techniques encompass wide applications given their ability to facilitate fabrication of devices spanning across the electromagnetic spectrum, from the ultraviolet to the infrared. The performance of photodetectors depends critically on factors like the quality of active layers, contact materials, and passivation films, all of which are deposited using various deposition techniques. Metal oxide semiconductors have emerged as materials that are highly conducive for manufacture of photodetectors due to their unique properties like wide bandgaps, high electron mobility, and environmental stability [19]. Specifically, materials like ZnO, SnO_2, TiO_2, and related oxides can be deposited using multiple thin-film deposition techniques like sputtering, CVD, ALD, and also solution-based methods like liquid exfoliation [19,20]. Sputtering appears to be a particularly suitable option for manufacture of photodetectors that are based on metal oxide materials due to its unique ability to deposit high-quality films at relatively low temperatures with good control over stoichiometry and doping [19,30]. Furthermore, the optical and electrical properties of sputtered metal oxide films can be tailored to suit specific requirements of different applications by exercising control over conditions like oxygen partial pressure, substrate temperature, and post-deposition annealing [19].

2D materials, that have exhibited unique optoelectronic properties in turn attributable to quantum confinement and strong light-matter interactions, have emerged as the latest and a highly promising frontier in photodetector technology, [4]. For instance, TMDCs exhibit layer-dependent bandgaps which makes them a suitable option for wavelength-selective photodetection.

Heterojunction photodetectors, which combine multiple materials with different bandgaps and work functions, benefit from the ability of various deposition techniques to deposit dissimilar materials in sequence and at the same time maintain sharp interfaces. The ALD technique has been considered particularly suitable for fabricating heterojunction structures due to its ability to control film thickness with precision and deposit conformal layers on pre-existing structures [12,13]. Layer-engineered functional multilayer structures created through ALD enable the design of photodetectors encompassing unique properties like tailored spectral responses, enhanced quantum efficiency, and reduced dark current [12].

Transparent conducting oxides (TCOs) like indium tin oxide (ITO) and aluminium-doped zinc oxide serve as essential components of photodetectors, providing transparent electrodes that allow light to reach the active layer while simultaneously collecting photogenerated carriers [18,19]. Sputtering has been identified as a highly suitable technique for deposition of TCO films due to its unique ability to produce highly conductive and transparent films while maintaining uniformity over large areas at the same time [18,30]. Needless to say, processes need to be optimised to enable seamless application of this deposition technique to photodetector technology. [11,18,19].

4.3. Emerging 2D Materials and Heterostructures

The pursuit of cost-effective, high-performance, and flexible alternatives has driven extensive research into organic and hybrid materials [55]. Organic semiconductors offer advantages such as ease of processing, structural flexibility, and chemical tunability [55,56]. Inorganic semiconductors, particularly nanomaterials, contribute high charge carrier mobility and tunable optoelectronic properties [55]. Combining these two material classes leads to hybrid organic-inorganic materials that can potentially merge the benefits of both [55,56].

Among these hybrid materials, two-dimensional (2D) materials have garnered significant attention [57]. These materials, characterized by their layered structure with strong in-plane and weak out-of-plane bonding, exhibit unique electronic and optoelectronic properties that are tunable based on their thickness and composition [57]. Graphene, transition metal dichalcogenides (TMDs), and perovskites are examples of 2D materials that have shown promise in various applications [57-61]. For instance, TMDCs, hexagonal boron nitride (h-BN), black phosphorus, MoS_2, InSe and other 2D materials exhibit unique electronic, optical, and mechanical properties that make them highly conducive for next-generation electronic and optoelectronic devices [4,11,31,60,61]. Furthermore, the integration of organic components into these 2D structures can lead to novel heterostructures with enhanced properties and functionalities [57,60,61].

The CVD method, which enables the growth of large-area, highly crystalline films by exercising control over factors like precursor chemistry, temperature, and growth kinetics, has emerged as the leading technique for scalable synthesis of 2D materials, particularly TMDCs [4]. Sulfurization or selenization of pre-deposited metal films has emerged as an alternative CVD approach that is capable of producing uniform TMDC films over large areas.

Given its unique properties, ALD technique encompasses unique advantages for synthesis of 2D materials, particularly in case of materials where CVD methods do not appear to be very effective. Recent advances in ALD chemistry have enabled direct growth of materials like TMDCs and metal oxides with improved quality and uniformity but the scope for further exploration remains vast [13].

Van der Waals heterostructures, which vertically stack different 2D materials to create artificial materials with novel properties have emerged as a frontier application on the thin film deposition landscape. The fabrication of high-quality heterostructures requires sequential deposition of different 2D materials while simultaneously maintaining atomically sharp interfaces and also preventing occurrence of interlayer contamination [4,12]. ALD and MBE techniques are generally understood to encompass the precise control required for heterostructure fabrication.

4.4. Applications in Solar Cells

Thin film solar cells represent a major application domain where multiple deposition techniques are employed. Cadmium telluride (CdTe) and copper indium gallium selenide (CIGS) solar cells utilize sputtering, CVD, and co-evaporation to deposit absorber layers, transparent conducting oxides, and buffer layers [3,26]. Sputtering is particularly useful for large-area TCO deposition on solar cells due to its unique properties of scalability and uniformity. [26]. Emerging perovskite solar cells employ solution-based and vapor deposition techniques to create high-efficiency absorber layers, with ALD showing suitability for depositing conformal buffer and passivation layers that enhance stability [12,13].

Furthermore, super capacitors benefit from vacuum-assisted thin film deposition techniques that enable precise control over electrode materials and architectures [33]. Sputtering and PVD techniques are used to deposit metal oxide and conducting polymer films with high surface areas and excellent electrical conductivity [33]. The ability to create nanostructured electrodes through controlled deposition enhances charge storage capacity and power density. Fuel cells and electrolysers utilize thin film catalysts and membranes deposited by sputtering, ALD, and other techniques. Platinum and platinum-alloy catalysts for oxygen reduction and hydrogen evolution reactions can be deposited as ultra-thin films, reducing precious metal loading while maintaining catalytic activity [12,26]. Protective coatings deposited by ALD can enhance the durability of catalysts and membranes in harsh electrochemical environments [12,13].

5. CURRENT CHALLENGES AND FUTURE DIRECTIONS

As discussed above, the thin film deposition techniques have registered remarkable progress over the years with respect to advent of innovative techniques, horizontal and vertical explorations in existing areas and also in terms of the ever-increasing applicability of these techniques over a wide range of research and industrial processes. However, the deposition paradigm has come with its own set of challenges that are manifested in certain specific areas of their current applications as also limit the scope of their application in hitherto unexplored domains. It is important to identify these challenges and address them in order to enhance the efficiency of these techniques and expand their applications in a manner commensurate with their capabilities. Some specific areas of concern in this respect are discussed below:

- Concerns related to scalability and throughput remain a persistent challenge, particularly in case of ALD and other inherently slow deposition techniques. Despite important advantages like unparalleled precision and conformality associated with the ALD technique, its low deposition rate limits the degree of throughput and increases manufacturing costs [12,13]. Although innovations like the spatial ALD technique, which separates precursor exposure zones spatially rather than temporally, batch processing and optimized time cycles encompass the potential for attaining a higher level of throughput while retaining the core advantages of the ALD technique have been developed, a research gap does exist in this area that needs to be addressed.
- Challenges like identification of suitable precursors encompassing the required degree of volatility, reactivity, and thermal stability [12,13,24] and also high cost are a critical bottleneck for enhancing accessibility of materials collected in the pallet while applying techniques like ALD and CVD. Future research in this area needs to focus on areas like creation of more versatile, cost-effective, and environmentally sustainable precursors [13].
- Challenges related to uniformity over large areas and complex structures are a major concern in case of techniques like sputtering where uniformity is contingent on substrate properties with non-uniformities becoming more evident in case of larger substrates [30] and CVD where factors like temperature gradients, nature and rate of precursor depletion, and gas flow patterns determine uniformity-levels [24]. Future research needs to address these specific concerns.
- Integration of dissimilar materials and heterostructures into the deposition process in a seamless manner remains a formidable challenge with respect to factors like interface quality, thermal budget compatibility, and process contamination. This calls for research in specific areas like plasma-enhanced processes, photo-assisted deposition, and development of other approaches that enable film growth at reduced temperatures [13,17,24].
- Challenges related to defect control and film quality optimization continue to be an important area of concern and also active research across all deposition techniques. Defects such as pinholes, particles, and grain boundaries can significantly undermine the performance of the device and thereby its reliability [3,12,30]. Understanding the origins of such defects and developing strategies to mitigate their impact requires application of advanced characterization techniques and a comprehensive study of growth mechanisms.
- Several precursors used in the application of CVD and ALD techniques have been reported to be toxic, corrosive and damaging for the environment. Energy consumption for vacuum pumping, heating, and plasma generation that are an integral part of the deposition process can have extremely harmful implications like carbon emissions and environmental footprints. Future research therefore needs to focus on

development of greener precursors, ensuring reduced energy consumption, and implementing the principles of circular economy in the manufacturing of thin films [13].

5.1. The Road Ahead for Thin-film Deposition Techniques

Simulators, machine learning algorithms and artificial intelligence are emerging as powerful tools for optimizing thin film deposition processes [25,60-62]. Application of relevant principles of the physical sciences like for instance, the domain of neural networks can enhance our understanding of the operational mechanism of deposition techniques. For instance, it may be possible to embed basic physical laws into machine learning algorithms that could facilitate more accurate prediction of film properties and process outcomes [25,60-62]. These approaches can accelerate process development, reduce experimental iterations, and enable real-time process optimization. However, challenges persist in areas like acquisition of sufficient training and data and also ensuring generalizability of models across different materials and equipment [25,60-62].

6. CONCLUSION

This comprehensive review has examined the fundamental principles, comparative advantages, and applications of major thin-film deposition techniques like physical vapour deposition, chemical vapour deposition, atomic layer deposition, and sputtering methods. The paper has also reviewed the liquid-liquid Interface Technique that is a recent development on the deposition landscape; highlighting its unique properties and scope for future explorations. It can be inferred from the comparative analysis that no particular technique can be adjudged as superior to others in absolute terms. As evident from the discussion in Sections 2 and 3, each method encompasses its own set of possibilities, advantages and opportunities that are unique to it. Similarly, each technique also has its own set of challenges that undermine its effectiveness in specific applications. The application of thin-film deposition techniques in the manufacture of semiconductors, photodetectors, and energy systems like solar cells exemplifies the critical role of these techniques in development of next-generation electronic and optoelectronic devices. The integration of novel materials such as transition metal dichalcogenides, metal oxides, and complex heterostructures has driven innovation in deposition technologies, with multiple techniques often employed synergistically to achieve desired levels of device performance. The continued scaling of semiconductor devices and the emergence of new device architectures will require further advances in deposition techniques, particularly in areas like conformality, selectivity, and low-temperature processing. Improving throughput while maintaining precision, developing new precursor chemistries for expanded access to materials, achieving better uniformity over large areas and complex structures, and reducing environmental impact represent critical research directions in this area. The integration of machine learning and artificial intelligence into process development and control encompasses major and hitherto unexplored possibilities for accelerating innovation in this area and also enabling real-time optimization. In sum, continued advances in thin film deposition techniques has been deemed essential for realising next generation electronic, optoelectronic, energy, and functional materials and technologies.

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CONFLICTS OF INTEREST

Authors declare that there are no known conflicts of interest.

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