



Atomic layer deposition thin film techniques and its bibliometric perspective

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Abstract

Atomic layer deposition (ALD) is known for depositing ultra-thin film materials that enable control of composition, highly conformal film, desirable thickness, self-saturating, and uniform deposition, and this review has established its evolution in recent times. The ALD techniques have made more device applications possible in energy storage, solar cells, memory storage, catalysis, sensors, and many more. Its advantages and disadvantages for different modes were emphasized and the precursors used for several ALD processes were highlighted. The bibliometric approach used in this review has also revealed how ALD has evolved through the assessment of published documents, journals, authors, organizations, sponsors, and countries. The method also revealed that ALD research is limited in Africa, however, the first two ALD facilities were confirmed to be acquired by T.C. Jen at the University of Johannesburg, which will in turn burst ALD material research in Africa. The current study has provided researchers with a choice when considering using the ALD technique and in terms of research collaborations. It concluded by highlighting the challenges and future perspectives of ALD and bibliometric technique.

Keywords ALD · Technique · Evolution · Bibliometric · Thin films

1 Introduction

Atomic layer deposition (ALD) involves the process of depositing thin films on the surface of a substrate. These films are conformal with a desirable thickness which are self-saturated due to precursors' gaseous interaction on the growth surface. The foundation of ALD started by developing atomic layer epitaxy (ALE) in 1970 by Dr. Tuomo Suntola and his associates. They used the technology in depositing thin film materials in the industry and was first used in electroluminescent (TFEL) displays [1]. After Intel introduced ALD into their industry in 2007, it quickly became the focus of the semiconductor production industry [2, 3]. Beyond microelectronics, the choice of applications for ALD may have increased, including catalysis [4], energy storage [5], phase-change memory materials (PCMMs) [6],

photovoltaics [7], ovonic threshold switches (OTSs) [8], and many more.

Several researchers have written different reviews that covered the ALD fundamentals, scientific growth, and applications [1, 2, 9–13]. Also, ALD's historical evolution has been studied. For example, Puurunen focused on the early years of the development of ALE [1]. As the development of ALE continues, reactors were designed to integrate metals and their oxides, numerous thin films were fabricated non-epitaxially, and the adoption of a common name ALD was established [11]. In the twentieth century, Parsons and co-workers described the backgrounds of ALD and also reported its growth in the American Vacuum Society [14]. Nevertheless, the ALD techniques and its research evolution is ripe for a review.

The evolution of ALD thin film research can also be established by exploring the use of bibliometric analysis, which can provide information about the ALD community. Bibliometric is a powerful tool that offers the opportunity for reviewing the outputs of science, structure, and dynamics of scientific disciplines [15]. Content is one of the primary methods used in bibliometric analysis [16]. The content review tried to pinpoint the most current hotspots by examining the frequency of the authors' keywords and other

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dispersions. In general, a bibliometric study is a useful tool for connecting authors and organizations to various countries around the world. By using such an approach, it is also possible to establish the research areas and current topics in the study area [17–21].

However, this review focused on the ALD techniques and how its research has evolved in recent times. In this present study, we have utilized both qualitative and quantitative approaches (two key factors of literature review) to establish a broad perspective about ALD technique in terms of history, processes, research, and collaborative network. To the best of my knowledge, no work has presented the ALD technique with such a prospect. It also concluded by emphasizing the challenges and future perspectives of advancing the ALD technique.

2 Atomic layer deposition (ALD) techniques

2.1 Brief history of ALD

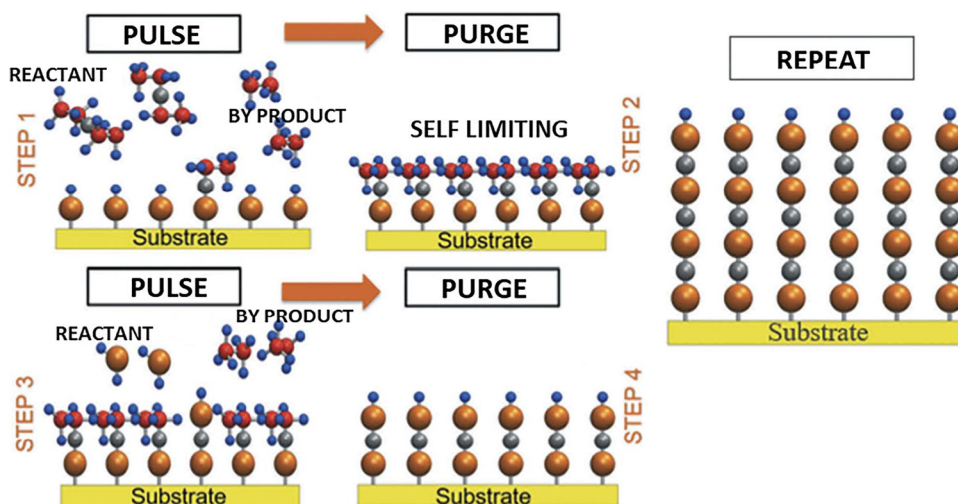
The concept of ALD was initially presented by Prof. Valentin Aleskovsky during his doctoral research [22]. Thereafter, he was able to publish an article based on ALD principles titled “Molecular Layering.” The study was carried out with Prof. Stanislav Koltsov’s at Leningrad Technological Institute (LTI) in 1960 [22]. In 1970, the creation of atomic layer epitaxy (ALE) technique was established by Dr. Tuomo Suntola and his associates. The technique was used in the industry to develop thin film [1]. The technique was initially employed electroluminescent (TFEL) displays [1]. Thereafter, this technology was presented in 20 nations [1]. The technology was ground-breaking when Suntola and co-workers came up with an inert gas reactor that enables more inclusion of compound reactants such as water vapor (H_2O), metal chloride (MCl_3), and hydrogen sulfide (H_2S) in the

process of ALE [1]. The research started at the Helsinki University of Technology in 1980; the ground-breaking technology was presented at the SID conference [1]. Subsequently, in Microchemistry Ltd, the ALE technology was developed for other applications such as heterogeneous catalysts and photovoltaic devices by Suntola. The ALD technology and manufacturing company Microchemistry Ltd was sold and became ASM Microchemistry Oy in 1999, which was the only manufacturing company of ALD reactors commercially. In the early stage of the 2000s, Picosun Oy and Beneq Oy manufacturing companies sprout up their expertise in Finland based on ALD reactors. The manufacturers of ALD reactors started increasing rapidly and the ALD technology became the industrial revolution of semiconducting applications, as a furtherance of Moore’s law [1]. This innovation led to the European SEMI award and Millennium Technology Prize received by Tuomo Suntola in 2004 and 2018 [1], respectively.

2.2 General process of ALD

The ALD procedure is known as a bottom-up method. As seen in Fig. 1, the necessary ALD thin film can be created in two or more procedures with various purge stages. Subjecting a substrate to a precursor or reactant is the first phase in the four-step ALD process (i.e., reactant 1). Reactant 1 is injected, generating chemical absorption on the surface of the substrate, and surface contacts between the substrate’s functional group and reactant 1 is being initiated. The method is repeated till all the functional groups on the substrate have been utilized. Surface contacts cause by-products to be released from the process of the first step. The by-products and unreacted reactant 1 are eliminated in the second purge phase. In the third phase, chemical absorbed reactant 1 and the co-reactant are involved in a self-limiting surface reaction on the surface of the substrate. In the

Fig. 1 Schematic of ALD general process [24]



second purge phase, the elimination of the by-products and unreacted co-reactants occurred, followed by the fourth step. This results in the formation of new generated active sites with functional groups on the substrate surface. Repeating this method on the substrate's surface results in layer-by-layer film deposition. This layer-by-layer process is shown in the right side of Fig. 1. The ALD cycle can be repeated countless of times to achieve the required thickness. As a result of its capacity to generate uniform deposition, desired thickness, and very conformal coatings on the surface of substrate, ALD has become a viable process for fuel cells, electronics, catalysis, and other applications [23]. Figure 1 depicts the general process of the ALD deposition technique.

To be as effective as feasible, ALD should be operated within a propitious temperature window to minimize sluggish thermal disintegration, growth rates, and inadequate reaction precursor condensation [2]. As a result, the phrase “temperature window” refers to the range of temperature where the film growth is saturated (see Fig. 2). ALD techniques include plasma-enhanced (PE) ALD, flash-enhanced ALD, thermal (T) ALD, and photo-assisted ALD. TALD and PEALD are the most frequent ALD techniques among these ALDs.

2.2.1 Thermal ALD (TALD)

TALD is a surface reaction method that allows for conformal, precise thickness control and substrate geometry. It also permits conformal coating in complicated structures with large aspect ratios. The temperature process of TALD runs from 150 to 350 °C and is considered a somewhat high-temperature technique, which limits its applicability. PEALD was created in order to solve this constraint [24].

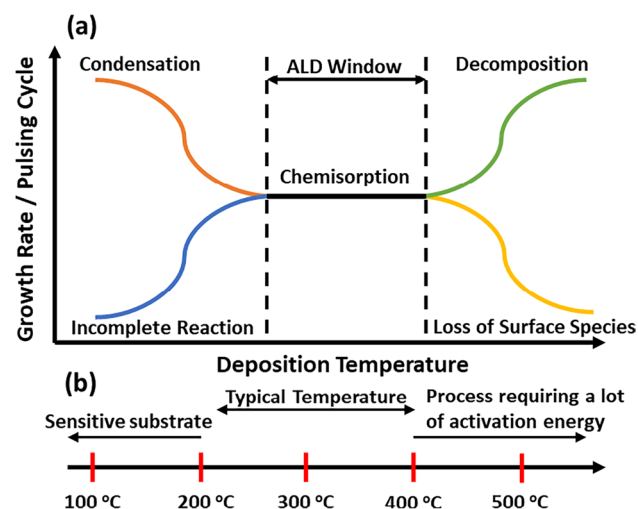


Fig. 2 Schematic of **a** ALD growth rate as a function of deposition temperature, and **b** temperature range of precursors

An example of TALD film deposition was Al_2O_3 fabrication from trimethylaluminum (TMA) and water (H_2O) as precursor and reactant, respectively. While introducing TMA, the process showed dissociative chemisorption of TMA on the substrate surface, pumping out the leftover TMA from the compartment and allowing AlCH_3 to cover the surface. Then, H_2O vapor is injected to react with the surface, thereby forming a reaction by-product of CH_4 and leading to a hydroxylated Al_2O_3 surface [24]. The $\text{Al}(\text{CH}_3)_3$ and H_2O ALD reaction is displayed in Fig. 3.

2.2.2 Plasma-assisted ALD (PA-ALD)

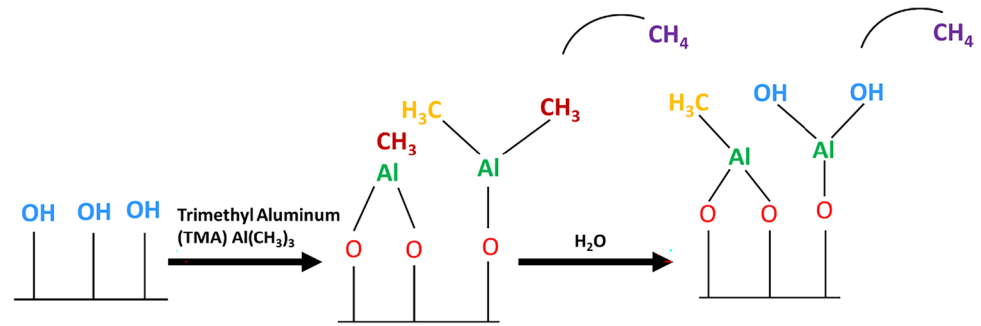
Another term for PA-ALD is plasma-enhanced ALD (PEALD). This type of ALD technique comprises plasma species with strong reactivity, allowing the deposited film temperature to be reduced without impacting the film's quality. As a result, PEALD is another process mode for improving a material's quality and attributes. Furthermore, PEALD offers a diverse spectrum of precursors that may be aided by plasma and enables for deposition of material that is difficult for TALD [25].

PEALD permits for comparatively deposition of low-temperature, and the film qualities are improved over TALD-deposited films [26]. PEALD also produces highly reactive catalysts or species at lower temperatures to boost the process. This reaction opens up a broader range of deposition precursors and substrates [24]. PEALD may therefore be used to build films on a heat-sensitive substrate. Furthermore, PEALD can generate more pure films than TALD [24]. Again, PEALD has the benefit of working at a faster pace of growth because to a diverse method. Again, PEALD has the benefit of working at a faster pace of growth through a diverse method [27].

Despite its benefits, PEALD has several unfavorable side reactions. These drawbacks include restricted conformal with a high aspect ratio or surface area, low throughput, difficult reaction chemistry, possibly poor conformal, extra growth factors, and complicated reactor designs [24]. There are four (4) types of low-pressure configurations of PEALD, namely, radical enhanced, direct plasma, remote, and direct plasma with mesh (see Fig. 4). The advantage of radical enhanced configuration over other configurations (direct and remote plasma) is because energetic plasma species do not damage the substrate or film growth and in the process of deposition, merely neutral plasma species are involved [24]. The direct plasma with mesh was developed to control the plasma surface interaction [12].

PEALD technique has been previously reported for the depositions of several nitride compounds, metal, and metal oxide films [28].

Fig. 3 Mechanism of ALD $\text{Al}(\text{CH}_3)_3$ and H_2O reaction to produce Al_2O_3 films on the surface of the substrate using TALD



2.2.3 Spatial ALD (SALD)

When the volume and surface area of an ALD reactor increases, this results in lower throughput of purge time and longer pulse. The lower throughput can be controlled by the use of spatial ALD (SALD), which addresses the quality of films produced by spatially resolving the head by exposing the substrate to a precise precursor based on position and eliminating the pulse-purge compartment [29]. SALD method is capable of reaching the rate of deposition of about 3600 nm/h [2].

The working principle of SALD is different from that of TALD and PEALD. In TALD and PEALD processes, the precursors are introduced in sequential pulses separated by purge steps and therefore involve time separation, whereas SALD involves the introduction of precursors through the different inlets into the reactor as depicted in Fig. 5. In this process, the substrate moves to the different inlets for film deposition. The process also reaches surface limiting and self-terminating due to chemical reaction which makes it

equivalent to ALD. Thus, SALD guarantees a high-quality film deposition at low temperatures, a specific growth per cycle, and a high aspect ratio conformal coating. SALD can be 10^2 magnitudes faster and it retains the distinctive properties of ALD. Also, SALD is a cheaper process due to the possibility to fabricate thin films at atmospheric pressure (AP-SALD) and it is easy to scale up because it does not involve an expensive and complex vacuum process. This process also faces some challenges as there is no wide range of precursors to be used [29]. Also, there are challenges such as impurity and non-desirable gas-phase reaction.

2.2.4 Photo-assisted ALD (PA-ALD)

Photo-assisted ALD, also known as ultraviolet (UV)-enhanced ALD was recently developed. As compared to PEALD, UV-enhanced ALD has a weak activation, but it can be controlled at ease when the intensity, illumination timing by UV light, and wavelength are been modified [24]. Exposure to UV does speed up the surface reactions on the substrate [24] and thereby enhancing the quality of the film [24, 25]. In another word, UV supplies energy to deposition reactions. More recently, a similar PA-ALD was developed called flash-enhanced ALD (FEALD). The only difference is that the light is used to transport heat that is of short pulses on the substrate surface and therefore drives the reactions on the surface of the substrate [30]. PA-ALD has been previously reported to deposit Al_2O_3 , ZrO_2 , TiO_2 , ZnO , BN , Ta_2O_5 , and GaAs using alkoxide precursors [31], and some of these precursors are tabulated in Table 2. Limited articles have reported PA-ALD research due to the challenge of constructing photo-ALD reactors [31].

2.2.5 Flash-enhanced ALD (FEALD)

FEALD comprises the process of exposing a substrate to a precursor. The molecules of the precursor chemically or physically absorb on the substrate surface. The substrate is next exposed to a flashlight (an arrangement of xenon flash lamps) of high intensity. This light's spectral

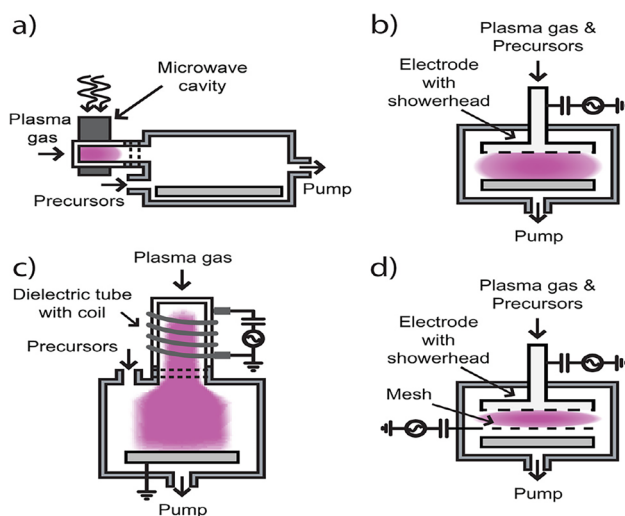
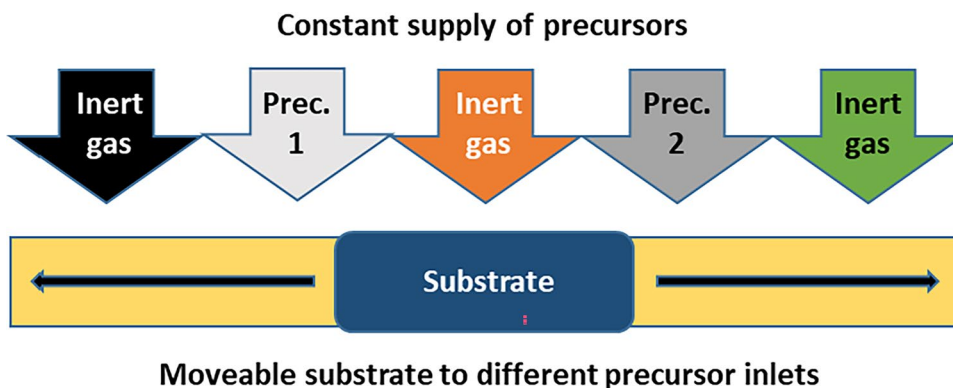


Fig. 4 Schematic of various forms of plasma configurations **a** radical, **b** direct, **c** remote, and **d** direct with mesh [27]

Fig. 5 Schematic of SALD process with a moveable substrate



scattering is primarily visible and near-infrared, with merely a small portion extending into the near-ultraviolet range [32]. As a result of direct interactions, the photolytic effects between the precursor molecules and light are negligible, and the flash treatment merely causes brief heating of the illuminated surface, resulting in activating the chemical reaction between the molecules of the adsorbed precursor and the second reactant or the thermally decomposing the molecule of the adsorbed precursor. Atoms from the precursors or compounds produced from the reactions form a layer on the substrate surface, followed by the purging of the volatile by-product from the compartment where the reaction took place. This process operates sequentially and it is also a layer-by-layer deposition process. Furthermore, the periodic annealing of the previously developed film caused by flash heating in each cycle may result in better film quality [30]. This technique has been reported for the depositions of Al_2O_3 , Si, and Ru [31]. Figure 6 displays the configuration process of FEALD.

The advantages and disadvantages of different modes of the ALD process are presented in Table 1.

3 ALD precursors

Solids, gasses, and volatile liquids can all be used as ALD precursors. For effective mass passage, the vapor pressure must be high enough; some liquid and solid precursors must be heated. Because of the self-limiting growth, the requirement for consistent evaporation rates is less severe than in other chemical vapor deposition (CVD) processes, making solid precursors easier to use. The inert-gas valving solves the difficult problem of pulsing precursors evaporating at high temperatures [10, 33]. The precursors need to be thermally stable at the growth temperature so that the decomposition of the precursor will not take away the self-saturated growth process and the benefits that come with it. Only mildly self-saturated growth can be achieved for thermally unstable precursors, that is, if the decomposition is delayed and contributes just a minor amount to total growth.

The surface sites must adsorb or react with the precursors. The adsorption type, whether dissociative or molecular, as well as reaction mechanisms, has been investigated recently in situ using data from mass spectrometry and quartz-crystal microbalance. For various precursors, different mechanisms

Fig. 6 Schematic of FEALD configuration process with flash lamp unit and cross-flow deposition reactor [30]

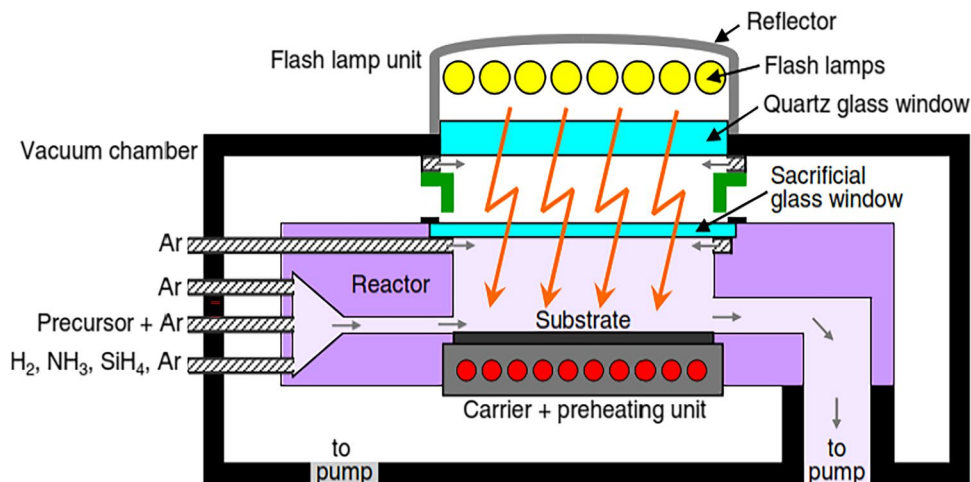


Table 1 Advantages and disadvantages of different ALD process modes

ALDs	Advantages	Disadvantages
Thermal ALD	Conformal coating in high aspect ratio and complex structure	High deposition temperature and slow deposition process
Plasma-enhanced ALD	Low deposition temperature, low impurity, and a wide choice of precursors	Low throughput and possibly poor conformal on the large surface area
Spatial ALD	The fast deposition rate and low deposition temperature	No wide range of precursors. Impurity and non-desirable gas-phase reaction
Photo-assisted ALD	High growth rate	Materials for photo-ALD are scarce. Challenge in construction of the photo-ALD reactors
Flash-enhanced ALD	Fast surface reaction	High deposition temperature

have been discovered [10, 33]. Halides, particularly chlorides, alkyl compounds, and alkoxides are common metal precursors utilized in ALD (see Table 2). Organometallic compounds like silyl and alkyl amides and cyclopentadienyl have recently received increased consideration [10, 34] (see Table 2).

Water (H_2O), hydrogen peroxide (H_2O_2), di-oxygen (O_2), and ozone (O_3) are utilized as non-metal precursors for oxygen; ammonia (NH_3), hydrazine (N_2H_4), and amines as nitrogen precursors and hydrides as the precursor for chalcogens [10, 33] (see Table 3).

It was realized that precursor combination for novel material with improved properties is very important. Table S1 presents the precursors and reactants used in the previous study for ALD deposition. More of these used precursors and their reactants can also be found in previous reports [12, 23, 36–43]. Most of these depositions were achieved employing thermal and plasma ALDs.

4 Bibliometric analysis of ALD thin film research

In this notion, bibliometric study entails the collection of methods for looking into the technical and scientific analysis of various published documents [67]. Through a series of procedures, including various publishing reports, organizations, authors, countries, and classification methodologies, the method examines several scientific journals with a focus on evolution and analysis [68].

H-index and impact factor (IF) are two (2) key metrics used to evaluate a journal's quality and are related to bibliometric analysis. IF is a method for measuring how frequently a typical journal work has been cited over a given year. The Institute of Scientific Information (ISI) recognized this instrument as a standardized technique for assessing high-quality journals [69, 70]. Hirsch introduced the h-index in 2005 as a quantitative method for evaluating the overall influence of academic's performance and output. This index can be used by both institutions and periodicals [71, 72].

Although Alvaro and Yanguas [3] have studied the research growth of ALD using the bibliometric technique, but their findings was not up to date. The authors investigated the growth of ALD up to 2015. Hence, this study demonstrates how rapidly ALD has expanded recently (2016–2021).

4.1 Methodology and data collection

4.1.1 Methodology

To assess ALD published literature, including reviews, book chapters, conference papers, journal articles, books, conference reviews, and other mediums of communication, this part used the bibliometric analysis method [73]. To complete the bibliometric assessment, R-studio from the R-package software was used along with the command languages bibliometric and biblioshiny. By providing details about a published document such as authors' names, journals, affiliations, nations, h-index, and publication year, the software makes it possible to do in-depth analyses.

4.1.2 Data collection

Numerous data were retrieved from the Scopus database, a recognized repository for academic publications. The data from 2016 to 2021 is retrieved using the keyword TITLE-ABS-KEY [atomic-layer-deposition]. We then analyzed the published documents based on the retrieved data.

4.2 Result and discussion

4.2.1 Published documents

Figure 7 presents the number of documents published in a year from 2016 to 2021. From the same figure, an increase in publication was observed in 2017, which later decreases in 2018 and further decreased in 2019. This decrease in publication may be because of understudy

Table 2 Types of metal precursors for ALD [35]

Precursor family name	Element	Precursor	Advantage	Disadvantage
Halides	B, C, Cd, Al, P, Cu, Ti, Ta, V, Sn, Cr, Mn, Zn, Pb, Ga, Hf, Ge, In, Sb, W	WF ₆ , TiCl ₄ , HfCl ₄ , SnCl ₄ , VOCl ₃ , CrO ₂ Cl ₂ , GeCl ₂	Thermally stable Usually inexpensive	Halogen impurities in films Corrosive by-products Some elements have low volatility
Alkoxides	B, Ge, Al, Ti, Nb, V, Ni, Zr, Pb, Si, Gd	Al(OEt) ₃ , Hf(O ⁱ Bu) ₄ , B(OMe) ₃ , AlMe ₂ (O ⁱ Pr), Hf(mmp) ₄ , Ni(dmamp) ₂ , Nb(OEt) ₅ , Ti(OEt) ₄ , Pb(O ⁱ Bu) ₂ , Si(OEt) ₄ , VO(O ⁱ Pr) ₃ , Ti(OMe) ₄ , Si(O ⁱ Pr) ₃ , Ta(OEt) ₅ , Ti(O ⁱ Pr) ₄ , Si(O ⁱ Bu) ₃	Reactive to water vapor => oxides	Thermal stability is limited Inappropriate for nitrides Inappropriate for pure metals
Beta-diketone	Mg, Zr, Ca, Sc, Cu, Ti, Sr, V, Cr, Ba, Mn, Fe, Co, Pt, Ni, Ga, Y, Ru, Pd, In, La, Sm, Hf, Re, Os, Ir, Er, Pb, Ce, Rh, Nd, Eu, Gd, Dy, Ho, Tm	Ba(thd) ₂ , Ni(acac) ₂ , Co(acac) ₂ , Dy(thd) ₃ , Pb(thd) ₂ , Cu(hfac) ₂ , Co(acac) ₃ , Pd(thd) ₂ , Ru(thd) ₃ , Ce(thd) ₄ , Pd(hfac) ₂ , Sr(methd) ₂ , Ru(od) ₃ , Nd(thd) ₃ , Ni(thd) ₂ , Pt(acac) ₂ , Mn(thd) ₃ , Pb(thd) ₂ , Pt(acac) ₂ , Mg(thd) ₂	Non-reactive to ambient air High thermal stability	Low reactivity to water vapor Low vapor pressure except for Cu(hfac) ₂ Not suitable for making nitrides Solids with high melting points
Alkylimides and alkylamides	Ti, Be, Cr, Sc, Al, Si, V, Mn, Fe, Co, Se, Ni, Cu, Zr, Ga, Ge, As, Pb, Sn, Y, Ta, Nb, Pr, Hf, Mo, Sb, Te, La, W, Bi	Al(NMe ₂) ₃ , Hf(NMe ₂) ₄ , Zr(NMe ₂) ₄ , Zr(NEt ₂) ₄ , Ti(NMe ₂) ₄ , Ta(NMe ₂) ₅ , Bi[N(SiMe ₃) ₂] ₃ , Zn[N(SiMe ₃) ₂] ₂	Highly reactive Suitable for oxides and nitrides	Thermal stability is limited Silylamides contain impurity of silicon
Amidates	Li, Mg, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Ga, Ge, Sr, Y, Zr, Ru, Ag, In, Sn, Ba, La, Hf, Pb, Bi, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb	Ag(ⁱ Bu ₂ -amd) ₂ , Co(ⁱ Pr ₂ -amd) ₂ , Co(ⁱ BuEt-amd) ₂ , Cr(Et ₂ -amd) ₂ , Cu ₂ (ⁱ Pr ₂ -amd) ₂ , Er(ⁱ Bu ₂ -amd) ₂ , Fe(ⁱ Pr ₂ -amd) ₂ , Fe(ⁱ BuEt-amd) ₂ , Ga(Et ₂ -amd) ₃ , Hf(Me ₂ -fmd) ₄ , Lu(Et ₂ -fmd) ₃ , Mn(ⁱ Bu ₂ -pemd) ₂ , Ti(ⁱ Pr ₂ -amd) ₂ , Zr(Me ₂ -pmd) ₄ , Zr(Me ₂ -bmd) ₄	High reactive to H ₂ O => oxides High reactive to NH ₃ => nitrides High reactive to H ₂ S => sulfides Reactive to hydrogen gas H ₂ => metals	Several different ligands are needed Some are solids, not liquids
Alkyls	Cd, B, Al, Te, Si, Zn, Ga, In, Bi, Ge, Sn, Sb, Hg	(CH ₃) ₃ Al, (CH ₃ CH ₂) ₂ Zn, ⁱ Pr ₂ Te	Volatile, highly reactive in ALD	Hazardous, burst into flame in the air (pyrophoric)
Cyclopentadienyls	Mg, Ca, Co, Sc, Ba, Ti, Sr, Mn, Ru, Fe, Ni, Y, Zr, In, La, Hf, Pt, Pr, Er, Lu	Cp ₂ Ni, (EtCp) ₂ Ru, (Me ₅ Cp) ₂ Sr, (ⁱ PrCp) ₃ La, Cp ₂ MeZr, (MeCp)(Me) ₃ Pt	Thermally stable	Elements like Ni and Ru have low reactivity La, Ni, In, Mg, and Sr are solids Low volatility is attribute of some elements (La, Sr, Mg)

Cp cyclopentadienyl, *MeCp* methylcyclopentadienyl, *Me₅Cp* pentamethylcyclopentadienyl, *EtCp* ethylcyclopentadienyl, *ⁱPrCp* isopropylcyclopentadienyl, *OⁱBu* isobutoxy, *OEt* ethoxy, *OⁱBu* tert-butoxy, *OⁱPr* isopropoxy, *mmp* 1-methoxy-2-methyl-2-propoxy, *dmamp* 1-dimethylamino-2-methyl-2-propoxy, *acac* acetylacetonate, *hfac* 1,1,1,5,5,5-hexafluoro-acetylacetonate, *thd* 2,2,6,6-tetramethyl-heptane-3,5-dione, *od* octane-2,4-dione, *NⁱBu* tert-butylimino, *NMe₂* dimethylamino, *N(SiMe₃)₂* bis(trimethylsilyl)amido, *NEt₂* diethylamino, *NEtMe* ethylmethyl-amino, *ⁱPr₂-amd* N,N'-isopropyl-acetamidinate, *Bu-amd* N'-butyl-acetamidinate, (*Me₅fmd*) N,N'-dimethylformamidinate, (*BuEt-amd*) N'-tert-butyl-N-ethylacetamidinate, (*ⁱBu₂amd*) N,N'-dimethylacetamidinate, (*Et₂fmd*) N,N'-diethylformamidinate.

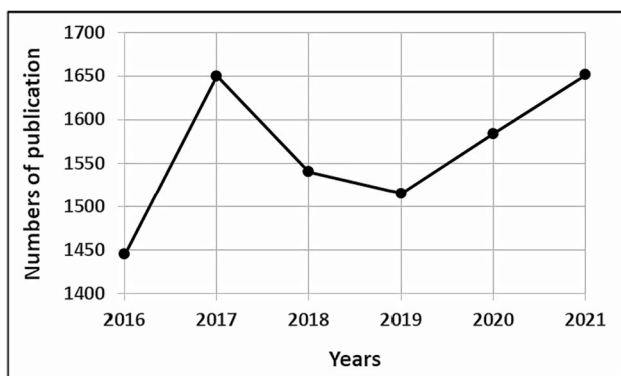
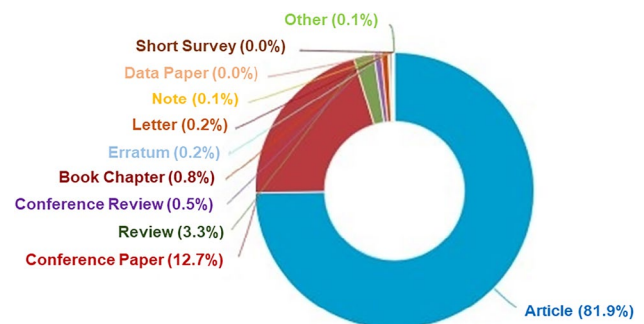
Table 3 Types of non-metal precursors for ALD [35]

Reactant	Type	Quality and toxicity
O	H ₂ O vapor	
	O ₂	Form of oxygen in the air
	O ₃	It is made in plasma; it is more reactive and always supplemented by O ₂
	NO ₂	Always accompanied by its dimer N ₂ O ₄
	H ₂ O ₂	At times, it is more reactive compared to H ₂ O (always accompanied by water)
N	NH ₃	
	N ₂ H ₄	It is more reactive compared to NH ₃ It is also toxic and explosive
	Plasma-activated NH ₃	It is more reactive compared to NH ₃
	N ₂	Usually not reactive under ALD situations
	Plasma-activated N ₂	It is more reactive compared to N ₂
	Nitric oxide (NO)	Could be utilized for nitrogen-doping of oxides
C	Acetylene gas (C ₂ H ₂)	Not particularly toxic, but toxic when produced from calcium carbide Highly flammable
S	Sulfur vapor, Sn	
	Hydrogen sulfide gas (H ₂ S)	Poisonous
Se	Hydrogen selenide gas H ₂ Se	Very poisonous
	Bis(triethylsilyl)selenium ((Et ₃ Si)Se ₂)	
Te	Bis(triethylsilyl)tellurium ((Et ₃ Si)Te ₂)	
P	Phosphine gas (PH ₃)	Very poisonous
As	Arsine gas (AsH ₃)	Very poisonous
Sb	Antimony trichloride (SbCl ₃)	
F	Hydrogen fluoride gas (HF)	Highly corrosive and powerful contact poison

the ALD thin films research development leading to an increase in the publication in 2020 and 2021. Generally, it was observed that the highest publication was in 2021 and 2017 with 1668 and 1653 published documents, respectively. Followed by 2020, 2018, 2019, and 2016 were the least with the numbers of publications of 1597, 1520, 1540, and 1448, respectively.

Scopus has shown 9426 published documents in which seven thousand seven hundred and twenty-two (7722)

articles were peer-reviewed, one thousand one hundred and ninety-seven (1197) conference papers, three hundred and fifteen (315) review articles, forty-four (44) conference review paper, eighty (80) book chapter, twenty (20) Erratum, seventeen (17) Letter, fourteen (14) Note, four (4) data paper and short survey each, three (3) book, two (2) editorial, one (1) retracted, and 3 others. Figure 8 displays the percentage distribution of these articles. The documents were also found to cross a variety of academic fields, as shown in Fig. 9.

**Fig. 7** Numbers of articles published per year**Fig. 8** Published documents percentage distributions

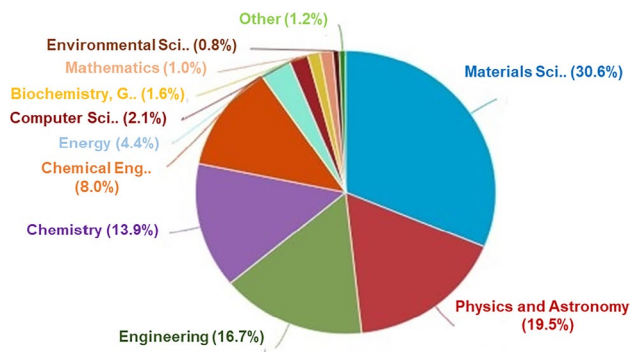


Fig. 9 Published documents percentage distributions across several academic fields

4.2.2 Journal source growth and performance

Figure 10 portrays the top 5 journals publication source growth of the ALD research. From the figure, *American Chemical Society (ACS) Applied Materials and Interfaces* had its highest publication in 2018 with 89 published documents, followed by *Journal of Vacuum Science and Technology A Vacuum Surfaces and Films* in 2020, *Chemistry of Materials* in 2019 and 2020, *Applied Surface Science* in 2017, and *Applied Physics Letters* in 2016 with 88, 55, 53, and 43 published documents, respectively. Generally, the lowest published document (21) was found in 2019 by *Applied Physics Letters*. Based on the result presented in Fig. 10, it can be said that more consideration is given to *ACS Applied Materials and Interface* and *Journal of Vacuum Science and Technology A Vacuum Surface and Films* has been displayed by several authors, which translates to their activeness in publishing the ALD research.

A sum of 154 journals has published several investigations from 2016 to 2021 based on different materials that used the ALD technology for various applications. Table 4 shows the evaluation of the top twenty (20) of these journals based on several performance indicators. It was noted that

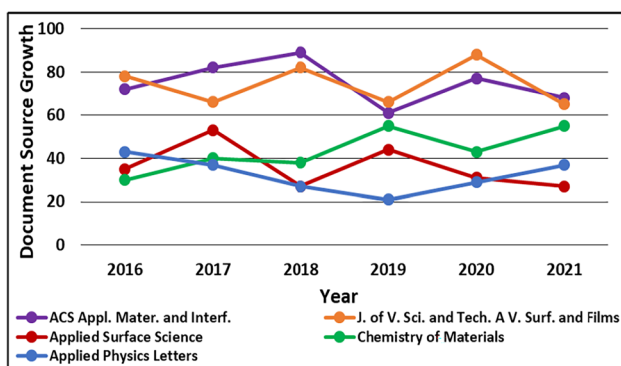


Fig. 10 Top 5 journals publication source growth per year

ten (10) of these journals are from the USA and 6 from the UK, indicating the most considered by several authors. Of the top 20 presented journals, the first five (5) most relevant journals are *ACS Applied Materials and Interfaces*, *Journal of Vacuum Science and Technology A Vacuum Surfaces and Films*, *Applied Surface Science*, *Chemistry of Materials*, and *Applied Physics Letters*. Among the first 5 journals, *ACS Applied Materials and Interfaces* has the highest number of articles, came third in IF, and fifth in h-index with values of 451, 9.229, and 228, respectively, indicating that a larger percentage of ALD material research was published in a high IF journal. *Journal of Vacuum Science and Technology A Vacuum Surfaces and Films* came second (445) in numbers of publications with a low IF of 2.427 and h-index of 112, followed by *Applied Surface Science* with values of 261 (numbers of publications), but ranked fifth in IF and ninth in h-index. It was observed that *Applied Physics Letters* has the highest h-index of 442 but came fifth in the numbers of documents with a low IF of 3.791. The table also revealed *Journal of Materials Chemistry A* had the highest IF of 12.732, but ranked fourteenth in numbers of documents (106) with an h-index of 212, implying that the journal has an IF quality. *Chemistry of Materials* also has an IF quality of 10.159 as well as an h-index of 375 and 218 for the total document. Other journal and their ranking information can also be found in the table.

Considering the work of Alvaro and Yanguas [3], *Thin Solid Films*, *Applied Surface Science*, *Journal of Vacuum Science and Technology A Vacuum Surfaces and Films*, *Chemistry of Materials*, *Journal of Physical Chemistry C*, and *ACS Applied Materials and Interfaces* were still considered by authors based on the number of published documents as they are listed among the top 10 in Table 4. *ACS Applied Materials and Interfaces* was ranked tenth in Alvaro and Yanguas's previous study, but came first in this current investigation, implying more consideration of the journal by authors, translating to the improvement of the quality of the journal from 2016 to 2021.

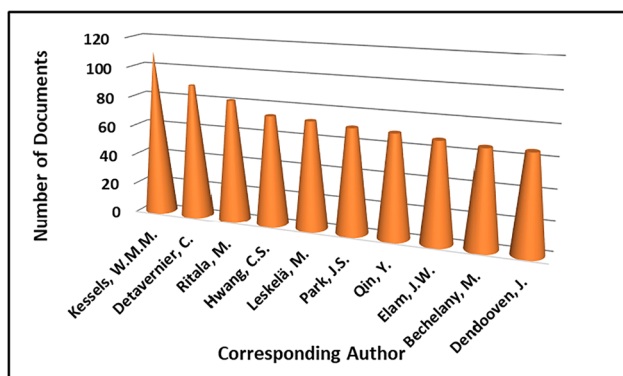
4.2.3 Authors and institution performance

ALD material research was analyzed based on corresponding authors of published documents from 2016 to 2021 and 159 authors were determined. Figure 11 demonstrates the number of documents published by the top ten (10) productive authors. From the figure, it was noted that Kessels WMM. and Detavernier C. have the highest numbers of articles 110 and 89, respectively. The observed highest publication translates to the activeness and effectiveness of the author in the field of ALD research. It also indicates that Kessels WMM. is the most productive author among the top 10 authors. Ritala M. and Hwang CS. were ranked third and fourth with 81 and 73 documents, respectively. Followed

Table 4 Twenty most relevant sources and their country, IF, h-index, and numbers of documents

S/N	Journal	Country	Impact factor	h-index	Document
1	<i>ACS Applied Materials and Interfaces</i>	USA	9.229	228	450
2	<i>Journal of Vacuum Science and Technology A Vacuum Surfaces and Films</i>	USA	2.427	112	445
3	<i>Applied Surface Science</i>	Netherlands	6.707	188	261
4	<i>Chemistry of Materials</i>	USA	10.159	375	216
5	<i>Applied Physics Letters</i>	USA	3.791	442	194
6	<i>Journal of Physical Chemistry C</i>	USA	4.126	289	163
7	<i>Nanotechnology</i>	UK	3.874	203	150
8	<i>Proceedings of SPIE The International Society for Optical Engineering</i>	USA	0.45	176	148
9	<i>Thin Solid Films</i>	Netherlands	2.183	192	134
10	<i>ECS Transactions</i>	USA	0.521	52	127
11	<i>Journal of Applied Physics</i>	USA	2.546	319	118
12	<i>Advanced Materials Interfaces</i>	UK	6.147	65	117
13	<i>RSC Advances</i>	UK	3.361	148	116
14	<i>Journal of Materials Chemistry A</i>	UK	12.732	212	106
15	<i>IEEE Transactions on Electron Devices</i>	USA	2.917	186	98
16	<i>Journal of Alloys and Compounds</i>	Netherlands	5.316	172	88
17	<i>IEEE Electron Device Letters</i>	USA	4.221	154	87
18	<i>Japanese Journal of Applied Physics</i>	Japan	1.480	67	85
19	<i>Journal of Materials Chemistry C</i>	UK	7.393	128	84
20	<i>Nanoscale</i>	UK	7.790	244	84

by Leskela M., Park JS., Qin Y., and Elam JW. with 72, 70, 69, and 67 documents, respectively. Bechelany M. and Dendooven J. were the least with published documents of 65 each. Considering the work of Alvaro and Yanguas [3], in terms of the productive authors, it was observed that Leskela M. and Ritala M. are still at the forefront of ALD research as they fell into the first ten productive authors in this present review (Fig. 11). Kessels WMM. came tenth most productive author in their work, which was ranked the first in this current review. Combining the previous work of Alvaro and Yanguas in 2018 and this current study, Ritala M. is the most productive author.

**Fig. 11** Performance of top 10 active authors

A total number of 160 affiliations were revealed by Scopus and Table 5 displays the number of publications and the most pertinent institutions in their country. The top 20 most productive institutions are spread throughout the top 7 most populous nations, according to the table. Additionally, it was discovered that South Korea and the USA each had five institutions, indicating that these countries are actively conducting research using the ALD technique. China also accounted for 4 institutions, while Finland and Belgium accounted for 2 each, and others (France and Netherlands) accounted for one each. Even though France and Netherlands showed the least activity among the 20 most active institutions, they publish more and are ranked among the top 8 countries for the number of papers published, with 257 and 177 articles, respectively. However, China also held the top spot in terms of publications (389) and appeared in the top 5 most-read articles. The results obtained above indicate that the Chinese researchers focus more on research using ALD technology owing to their exceptional film depositions, followed by the French and the Americans.

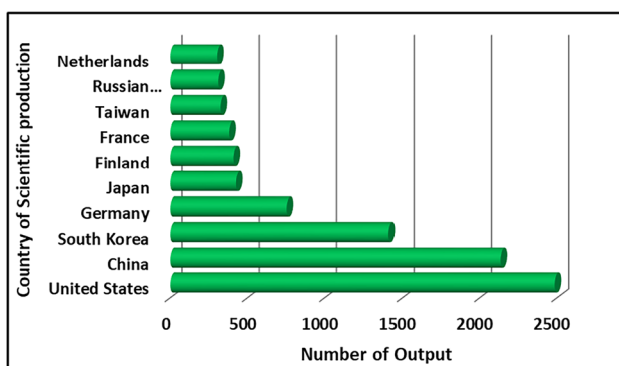
4.2.4 Countries and sponsors of scientific research production

A total number of 167 countries of scientific research production were identified and the top 10 were demonstrated in Fig. 12. The figure also displayed the frequency of each

Table 5 Twenty most relevant affiliations

S/N	Affiliation	Country	Document
1	Chinese Academy of Sciences	China	389
2	Ministry of Education China	China	270
3	CNRS Centre National de la Recherche Scientifique	France	257
4	Argonne National Laboratory	USA	211
5	Hanyang University	South Korea	205
6	Aalto University	Finland	186
7	Stanford University	USA	182
8	Technische Universiteit Eindhoven	Netherlands	177
9	Fudan University	China	168
10	Seoul National University	South Korea	157
11	University of Chinese Academy of Sciences	China	154
12	Yonsei University	South Korea	153
13	Sungkyunkwan University	South Korea	137
14	Helsingin Yliopisto, University of Helsinki	Finland	132
15	Interuniversity Micro-Electronics Center at Leuven	Belgium	130
16	Northwestern University	USA	119
17	Universiteit Gent	Belgium	112
18	University of Colorado Boulder	USA	105
19	Seoul National University of Science and Technology	South Korea	100
20	The University of Texas at Dallas	USA	98

nation's research output. The USA was found to have the highest output (2496), suggesting greater attention in the field of research than other nations. China, South Korea, and Germany were the next three countries, with 2142, 1404, and 761 research outputs, respectively. The outputs for Japan, Finland, and France are 427, 411, and 382, respectively. However, Taiwan, Russia, and the Netherlands exhibited the least countries as shown in the figure. It was also confirmed from the accounted 167 countries of scientific research production that African countries were lagging far behind in ALD thin film research (South Africa 45, Egypt 33, Tunisia 11, Algeria 5, Nigeria 4, Cameroon 1, and Ghana 1).

**Fig. 12** Countries of scientific production

Based on ALD research from 2016 to 2021, 159 sponsors were identified, and the top 20 sponsors have been displayed in Table 6. From the same table, the top 20 sponsors came from China, the USA, South Korea, the European Union, Germany, and Taiwan. The National Natural Science Foundation of China was found to have sponsored 1366 ALD research, implying their great interest in the ALD community. National Science Foundation in the USA also sponsored 838 research documents. South Korea was ranked third with sponsored ALD research of 672 documents, followed by the U.S. Department of Energy (fourth) and European Commission (fifth) with sponsored research of ALD deposited films of 599 and 394 documents. As seen in the table, the USA, China, and the European Union appeared five times each, followed by South Korea (appeared three times), Germany and Taiwan appeared once each among the top 20 sponsors. These results have shown that the USA, China, and the European Union sponsored most of ALD thin film research.

5 Conclusions, challenges, and perspectives

The ALD techniques have been studied, its advantages and disadvantages for different modes have been established, and the precursors used for several ALD processes have been highlighted. Currently, thermal and plasma ALD are the most used techniques due to their large availability of precursors. The plasma ALD has the limitation of low throughput and possible poor conformal that can be achieved on a

Table 6 Top 20 research sponsors, their countries, and published documents

S/N	Sponsor	Country	Document
1	National Natural Science Foundation of China	China	1366
2	National Science Foundation	USA	838
3	National Research Foundation of Korea	South Korea	672
4	U.S. Department of Energy	USA	599
	Office of Science	USA	394
5	Ministry of Science, ICT and Future Planning	South Korea	372
6	European Commission	European Union	370
7	Ministry of Trade, Industry and Energy	South Korea	348
8	Horizon 2020 Framework Programme	European Union	251
9	Basic Energy Sciences	USA	249
10	Fundamental Research Funds for the Central Universities	China	234
12	U.S. Department of Defence	USA	212
13	Ministry of Science and Technology of the People's Republic of China	China	210
14	National Key Research and Development Program of China	China	195
15	Deutsche Forschungsgemeinschaft	Germany	188
16	European Research Council	European Union	177
17	Ministry of Education of the People's Republic of China	China	169
18	Ministry of Science and Technology	Taiwan	161
19	European Regional Development Fund	European Union	152
20	Seventh Framework Programme	European Union	144

substrate with a high aspect ratio or high surface area. Thermal ALD on the other hand is a technique of choice based on temperature but can deposit on such substrate. Despite the advancement of ALD in recent years, there remain some challenges to its future improvement. Fundamental research on precursor chemistry (especially on non-toxic precursors) and film deposition will continue to be crucial, particularly as ALD's application to novel materials and technologies is expanding. Precursor combination is also an important perspective; therefore, a thorough review should be done to have a great knowledge based on the choice of precursors or considering simulations using both density functional theory (DFT) and Ansys Fluent (prediction of future outcome) to avoid trial and error which may lead to wastage and to reduce cost and adverse environmental impacts. Though the simulation challenges have been pointed out by Sibanda et al. and the most crucial is the complexity of the simulation software and the simulation robust structure [74] which needs simplicity and high computing power, respectively, to avoid time wastage. Furthermore, the development of new reactors for optimization of ALD processes is essential as conventional ALDs have different disadvantages limiting material production and applications.

The evolvement of ALD research was also investigated using bibliometric analysis in recent times. From 2016 to 2021, the technique revealed a total number of 9426 published documents based on ALD research. It was confirmed that 154 journals (sources) published the above number of documents. It was also affirmed that most of these journals

are from the USA and the UK, indicating their consideration by authors and the quality of the journals. The Chinese Academy of Sciences and the National Natural Science Foundation of China were the most productive institution and sponsors of ALD research, respectively, suggesting that they focus more on ALD research. Considering the work of Alvaro and Yanguas [3] and this current review, it was noted that Ritala M. with a University of Helsinki affiliation from Finland is the most productive author. The bibliometric analysis also revealed that ALD research is limited in Africa, nevertheless, the first two ALD facilities (thermal and plasma ALD) have been acquired by Professor T.C. Jen at the University of Johannesburg, South Africa in 2019 [75], which will, in turn, burst ALD material research in Africa. The bibliometric method has been demonstrated to be a crucial instrument for the quantitative study of ALD research, but the method still faces some challenges which lie in the author's names and initials. Authors with the same name and initial may be presented as one and vice versa. Again, content evaluation is necessary as some articles could be listed in the investigation in which their main text might not complement the interested keywords, but because their abstract quoted these keywords. Thus, there is a need to investigate and resolve these issues for an excellent presentation of authors and documents.

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Author contribution James Ayodele Oke: conceptualization, methodology, investigation, formal analysis, data curation, software, investigation, visualization, writing—original draft. Tien-Chien Jen: conceptualization, project administration, funding acquisition, writing—reviewing and editing.

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Data availability The data used in this study are available on Scopus at www.scopus.com.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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