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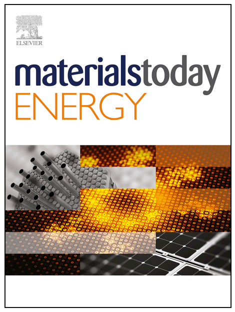

Atomic Layer Deposition of Piezoelectric Materials: A Timely Review

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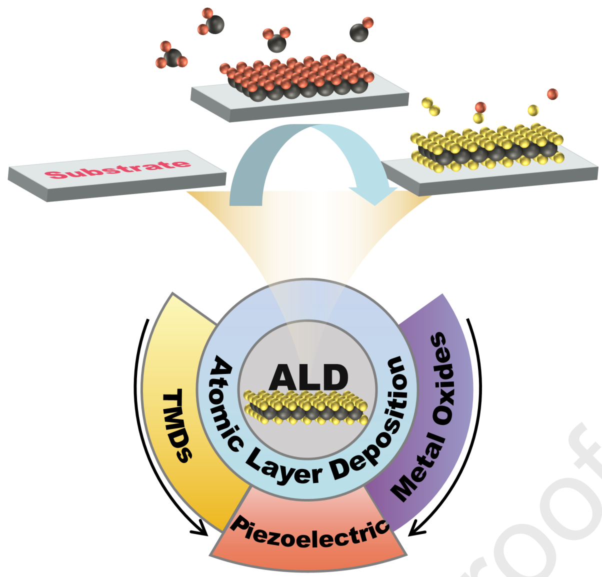
Substrate
ALD
Atomic Layer Deposition
TMDs
Piezoelectric
Metal Oxides

Atomic Layer Deposition of Piezoelectric Materials: A Timely Review

Yun Li[1,2,^], Ronn Goei[1,2,^], Amanda Jiamin Ong[1,2], Yiming Zou[1], Adva Shpatz Dayan[3], Stav Rahmany[3], Lioz Etgar[2,3], Alfred Iing Yoong Tok[1,2,*]

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Abstract

Piezoelectric effect plays an important role in a variety of applications, such as sensors, nanogenerators and piezotronics. The performance of piezoelectric device is normally enhanced with increasing dimension of the piezoelectric layer and decreasing piezoelectric layer thickness. To meet the demand for producing superior piezoelectric films (as thin as 1 nm) with precise thickness and composition control, powerful fabrication techniques are essential. Atomic layer deposition (ALD) shows exceptional potential in preparing a wide range of materials with precise thickness control (due to its self-limiting growth nature at the Angstrom level) and capability of deposition on high aspect ratio surface. Here, we provide the introduction to ALD and highlight its unique features among other fabrication techniques, with reference to the state of the art on ALD preparation of different piezoelectric materials, including novel transition metal dichalcogenides (TMDs) and traditional Metal Oxides (MOs). Different ALD-related materials preparation strategies for the improvement of piezoelectricity are also discussed, together with future perspectives on the development of ALD-prepared piezoelectric materials. We believe ALD can enable wider applications of piezoelectricity due to its unique advantages.

Keywords: Atomic Layer Deposition, Piezoelectric Materials, Transition Metal Dichalcogenides (TMDs), Metal Oxides (MOs), conformal thin film, giant piezoelectricity.

1. Introduction

Piezoelectric materials are materials that provide electrical responses when they are subjected to a mechanical stress (direct piezoelectric effect) or, reversibly, materials that demonstrate mechanical deflection or change in shape when they are subjected to an electrical stimulation (reverse piezoelectric device) [1, 2]. The direct piezoelectric effect was first discovered by Pierre and Jaques Currie in 1880. Different types of piezoelectric materials such as ceramics, perovskites, and polymers have been used in various forms of nanostructured thin films for different advanced applications such as energy harvesters, sensors, actuators, and transducers [3-8]. Furthermore, the development of materials with high piezoelectric properties combined with a flexible and/or stretchable structure opens more novel applications in soft robotics, biomedical engineering, piezotronics, and wearable electronics among other emerging technologies.

Generally, Piezoelectric materials can be catalogized into lead-based perovskites, lead-free ceramics, piezoelectric polymers and novel piezoelectric nanocomposites. Lead zirconate titanate (PZT), a type of perovskites, is the most popular piezoelectric material in the past decades due to its high output voltage (as high as 100V) and stable piezoelectric properties [9]. It is urgent to replace PZT by other lead-free ceramics due to its toxic nature. In the past few years, some lead-free ceramics attract lots of attention, such as $BaTiO_3$ (BTO) and $(Bi_{0.5}Na_{0.5})TiO_3$ (BNT), which possess relatively good piezoelectric properties [10, 11]. The conventional fabrication process is now preventing their widely applications due to the decreased piezoelectricity caused by existing non-piezoelectric phases and defects [12]. Doping is widely applied to increase and stabilize the piezoelectricity of both lead-based and lead-free ceramics [11]. However, the piezoelectric ceramics cannot meet the emerging demand for soft piezoelectric materials due to their rigid nature. Hence, polymeric piezoelectric materials are developed, in which PVDF is the most attractive one. PVDF is lightweight, flexible but with lower piezoelectric properties compared to ceramics. Its piezoelectricity is proportional to the β phase percentage. Some ceramic fillers were used to increase its β phase but destroyed its soft characteristic. Unlike ceramics, polarization is needed for piezoelectric polymers, which also limits their usage [13, 14]. Novel piezoelectric nanocomposites, such as nanowires, nanorods, nanobelts and 2D materials, are also developed to replace the ceramic materials. Metal oxides (MOs) and Transition metal dichalcogenides (TMDs) are the representative piezoelectric nanocomposites. Their piezoelectric properties are dependent on their orientation, band structures, nanostructures and thickness in film state. Due to their bandgap tunability (from 1 to 4eV) and soft nature in 2D state, they are considered as candidates for the increasing demand for portable and flexible devices. However, the piezoelectric properties of MOs and TMDs are also worse than traditional ceramics [15, 16]. Developing effective preparation methods with customizing their properties is the way towards their applications.

To satisfy the increasing demands for next-generation compact and flexible piezoelectric devices, the preparation of piezoelectric materials requires the deposition of continuous and homogeneous thin films (even down to few atomic layers) on 2D/3D substrates that would enhance the performance of the sensors [3, 5, 8, 17-22]. The most common structure is the sandwich device, which comprises of a bottom substrate, a middle active layer and a top encapsulate layer with top/bottom or side electrodes embedded inside the structure. Benefited from the developments of 3D printing and proximity-field nanopatterning, a variety of other structures can be derived from the sandwich structures, including truss structure, kirigami structure and so on [23, 24]. In all cases, the intrinsic property of the active material, device structure and the selection of electrode are the major parameters determining the device performance. Hence, we will mainly focus this review on the deposition of piezoelectric materials, which is notably related to the device performance by affecting the qualities of the materials and ceiling the tolerant temperatures of the whole structures.

Atomic layer deposition (ALD) is a thin film deposition technique with the ability to deposit a variety of materials on 2D or even high aspect ratio (as high as 4000:1) 3D features [25]. ALD has shown giant potential in solar cells, semiconductors and batteries due to its decent composition and thickness control under lower temperature [26, 27]. We will review the state of the art on ALD fabricated piezoelectric materials and use Transition Metal Dichalcogenides (TMDs) and Metal Oxides (MOs) as a case study. Interestingly, these two types of material can only exhibit good piezoelectricity when certain conditions are met. For example, MoS_2 shows piezoelectricity when its number of layers is an odd number [28]; piezoelectricity of ZnO nanowires is highly dependent on its orientation [29]. Therefore, these properties also require decent control of the deposition process, which highlights the need to summarize the correlations between deposition parameters and piezoelectric performance.

In this review, we will discuss how ALD technique is used to yield a highly uniform (conformal) piezo film growth with precise control of the resulting thin film thickness and morphology. The advantages of ALD on depositing piezo materials will be summarized by comparing with other methods. We will showcase ALD preparation of two important classes of the piezoelectric material: TMD and MO piezoelectric materials. For each material type, we will describe the predicted piezoelectric properties, summarize ALD deposition routes and discuss the strategies to improve their piezoelectricity from different directions. Strategies for enhancing piezoelectricity of TMDs by ALD can be generally categorized into: (a) 2D layer deposition on 3D substrate, (b) formation of heterostructures, multinary, and dopants in the structure of the piezo materials, (c) directly deposition of the piezo materials on the flexible substrates and (d) introduction of flexoelectricity. For the MOs piezo material, we have proposed the strategies by considering the individual preparation steps: (a) seed layer deposition, (b) 2D MO piezotronics by taking advantage of thickness control, (c) doping/composite co-deposition and (d) 3D nanostructure device integration (Fig. 1). Most of the improvement in piezoelectric properties can be obtained through precise ALD deposition controls. Finally, we will outline the prospects of ALD in depositing piezo materials to meet the increasing demand for high performance piezo devices and soft devices integration.

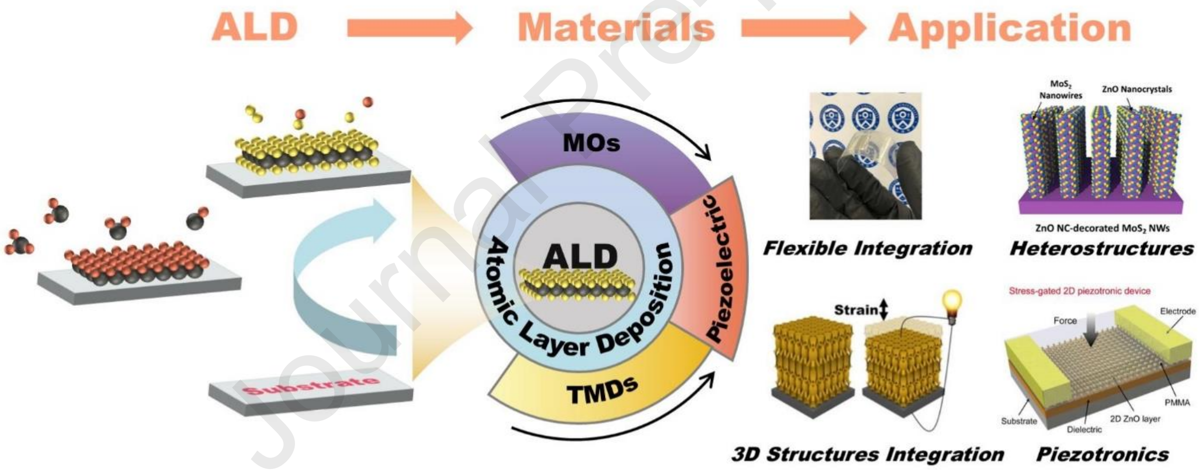

Figure 1. Typical application of ALD in TMD and Metal Oxide piezoelectric materials. Reproduced with permission from the study by Wang et al. [30]. Copyright 2019, Elsevier. Reproduced with permission from the study by Oh et al. [31]. Copyright 2020, American Chemical Society. Reproduced with permission from the study by Kim et al. [24]. Copyright 2020, Elsevier. Reproduced with permission from the study by Kang et al. [32]. Copyright 2022, Elsevier.

2. Atomic Layer Deposition Thin Film Preparation Technique

Atomic layer deposition (ALD) has been established as a thin film preparation technique that yields highly uniform growth and allows precise control over film thickness. ALD is considered an advanced development to chemical vapor deposition (CVD). In the CVD process, two vaporized precursors react in the chamber followed by the adsorption of the resulting products on the surface of the substrate. The reaction rate can be roughly controlled by adjusting the gas flow of precursors and the temperature of the reactor [33]. Due to the continuous interaction between two precursors, swift and uniform deposition cannot be easily achieved through CVD. ALD process reverses the sequence of the reaction and adsorption. The ALD precursors would adsorb and react sequentially in the self-limiting manner (i.e., the reactions would terminate once all the active sites present at the surface of the substrate have been fully occupied). A typical ALD process cycle involving two precursors (also known as an AB-type ALD) consists of four steps: (1) The vapor of precursor A is pulsed into the reactor, getting adsorbed on the surface of the substrate. The adsorption would stop when the active or anchoring sites on the substrate surface are fully occupied. (2) The residue A is then purged out from the ALD chamber by the pump. (3) Precursor B is then pulsed into the reactor reacting with the previously adsorbed precursor A. As the chemical reaction can only occur on the surface of the substrate, ALD process would result in the formation of a monolayer of product and excessively adsorbed precursor B on the newly formed layer. (4) The gaseous by-products generated by the reaction are then pumped out. As one of the deposition cycles could only contribute to a monolayer, the ALD cycle usually repeats for multiple times allowing layer-by-layer build-up of the thin film. The characteristic that limits the film growth to an atomic level is known as the self-limiting property [34]. Due to the unique growth cycle, ALD allows the formation of a highly uniform thin film with precise thickness control by tuning the number of deposition cycles. However, due to the nature of the self-limiting growth, the speed of thin film growth is generally slower than the traditional CVD or PVD (physical vapor deposition) processes.

2.1. Major parameters affecting ALD thin film growth

Even though ALD process is called “layer-by-layer” method, the growth of a thin film always starts from “nucleation points”. The nucleation first occurs on the active sites present on the surface of the substrate, forming an uniform nanoparticle layer through molecular diffusion and aggregation on the surface [35, 36]. As a result, a typical ALD process endures an initial undesirable unstable growth period before progressing to the linear growth period. The thickness error of a thick film can be negligible in ALD process with hundreds of cycles, but for many thin films with thickness less than 10 nm, such an unstable growth can be detrimental to the performance of the materials. It should be noted that the thickness of piezoelectric thin film is likely to decrease to few nanometres (even 1~3 nm for TMDs). Therefore, it is important to enhance the thickness controllability especially when the target thickness is thin. Herein, we will review two major parameters that could be tailored to improve the quality of ALD deposited thin films: surface modification and deposition temperature.

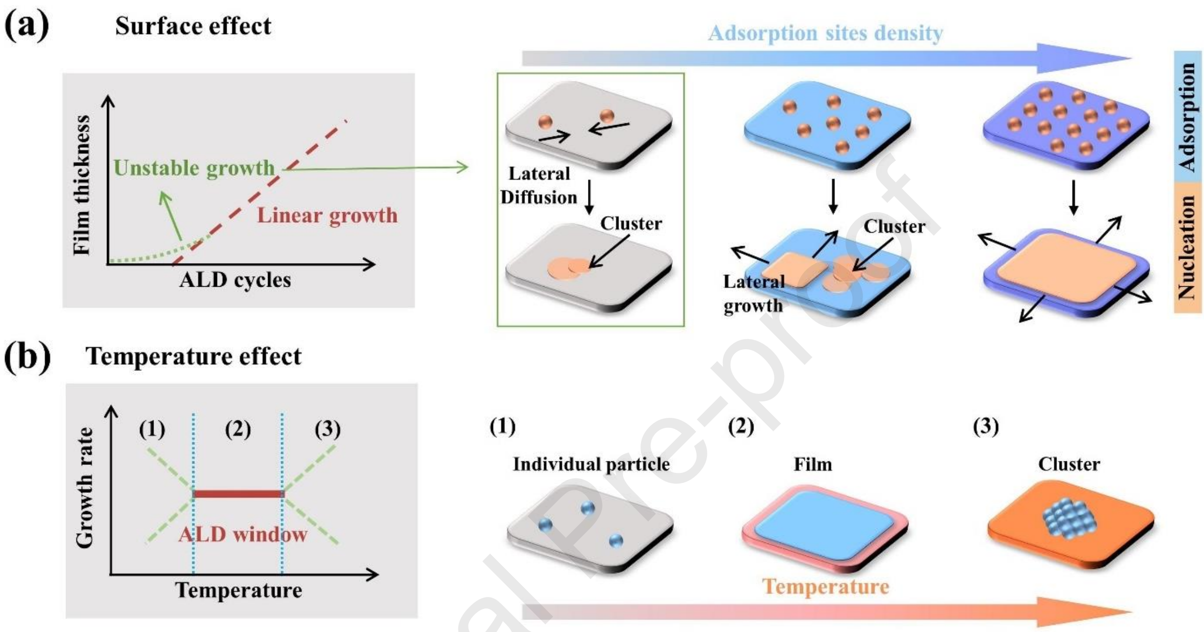

Figure 2. The effect of the major ALD parameters on the film growth. (a) Surface effect. (b) Temperature effect.

Surface modification

ALD process is a surface-dependent process, where the materials of substrates and condition of the surface largely determine the initiation of the film growth, defining the nature of film growth. A surface that favours the adsorption of vaporized precursor can shorten the initial stage of ALD process. It has been concluded from experiments that the oxide, insulating and semiconducting surfaces, such as Al_2O_3, silicon, can be more supportive to the interaction between surface and the adsorbates, as the bonding to surface may resemble the bonding in isolated compounds [37]. In addition to the substrate material selection, surface modification can be applied to further enhance the adsorption or make the adsorption viable to the precursors that are hard to get attached to any surface [38]. Simulation works have revealed the adsorption energy of adatoms to various surface sties [39-41].

Through the creation of anchoring sites on the surface of the substrate, the precursor can easily get adsorbed at those sites, thus enhance the nucleation rate and shorten the unstable growth period. For most of the metallic precursors, the hydroxyl group on the surface tends to act as the anchoring site [42], which can be promoted by chemical pre-treatment to the substrate [43] or through oxygen plasma surface treatment [44]. The anchoring sites would not only increase the adsorption of the precursor molecules, but it would also anchor them (once adsorbed) and prevent them from detachment and the subsequent lateral diffusion process [45]. As a result, more nuclei can be formed and grow independently, leading to more uniform morphology (as shown in Fig. 2a).

Deposition temperature

ALD processes are normally running at a relatively low temperature (< 300 °C), compared with other chemical deposition techniques, and each process for a specific material always has a temperature window for the deposition (see Fig. 2b). Deposition temperature determines not only the crystallinity of deposited thin films [46-48], but also the composition of them (especially when an oxidizing agent is used as the co-reactant) [46, 49, 50].

In regards of depositing thin film with low thickness and high uniformity, a lower deposition temperature is generally desired because mobility of the nanoparticles near or on the surface of the substrate is both size- and temperature-dependent. According to law of thermodynamics, the mobility of a monomer or a nanoparticle on the surface can be characterized by the diffusion rate, written as

$$D_n = D_1 n^{-s} \tag{1}$$

$$D_1 \propto e^{-E/kT} \tag{2}$$

where D_1 is the diffusion rate of a monomer, n is the number of atoms that comprise the nanoparticle, s is the constant determine by the type of diffusion, E is activation energy for adatom diffusion, k is Boltzmann's constant and T is the surface absolute temperature [36, 51, 52]. As such, at a higher temperature, atoms tend to travel on the surface and aggregate to form a bigger cluster, which is detrimental to thin layer deposition. Lu et al. proved that Pt cluster was formed at 300 °C by ALD, while dispersed single atoms were observed at 150 °C [53].

Therefore, it is important to optimize the ALD deposition temperature and balance quality and deposition rate. To enhance the crystallinity under low temperature, plasma enhanced atomic layer deposition (PEALD) is usually used by activating the reactants [44], which also provides surface modification of substrate during the deposition process. Thus, a properly treated surface and optimal deposition temperature are essential to promote the growth and uniformity of the thin films of piezoelectric materials.

2.2. ALD vs other piezoelectric material thin film deposition techniques

Along with the investigation on modifying ALD processes, various forms of nanomaterials have been realized, including a single atom [45], nanoparticles [54, 55], core-shell structures [56-58], monolayers [59], and thin films [60, 61], etc. In brief, compared with other thin film fabrication methods, ALD technique is preferred when the material design at nanoscale is required.

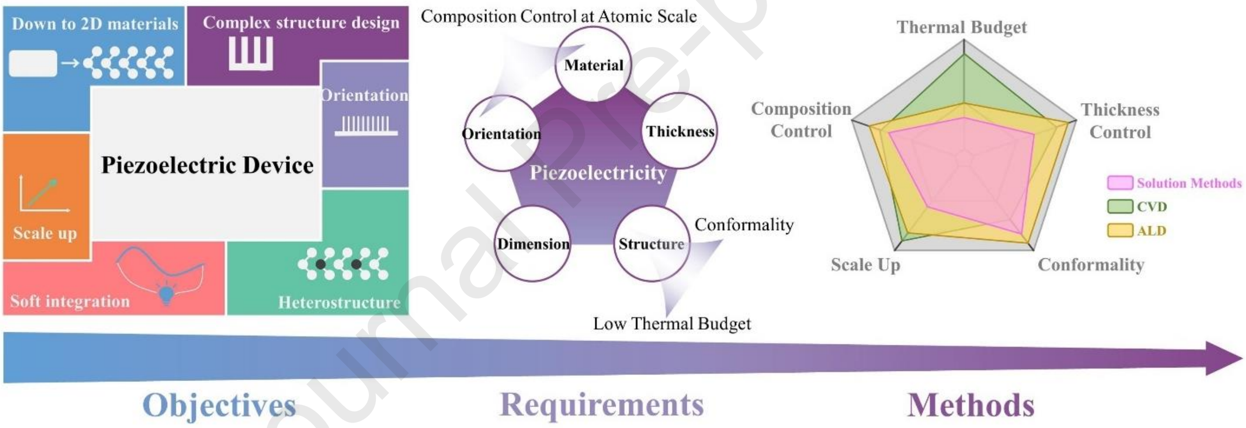

Figure 3. Relationship of objectives, requirements and methods in piezoelectric device field.

We summarize the relationship between objectives, requirements and methods of piezoelectric device in Fig. 3. The objectives of piezoelectric device can be divided into 1) down to 2D materials. To expand the piezo applications of some specific materials and piezotronics, it is important to pave the way to 2D materials; 2) scale up, i.e., increase the effective dimension of materials especially for those 2D materials. Although some of them already show decent piezoelectricity, the size can only be hundreds of square microns; 3) soft integration. Present flexible/stretchable piezoelectric devices are mainly based on transfer process. i.e., transfer the active layer to a new substrate by dry or wet transfer routes, resulting in contamination, defects and mismatch between films and new substrates [62]. This is because the high temperature of deposition process exceeds the tolerant temperature of most flexible polymers (e.g., PDMS can only work below 300 °C). Therefore, lowering the deposition temperature is the key to expand the application of flexible/stretchable piezoelectric devices; 4) doping/heterostructure. Feasible introduction of dopants and heterostructures has been proved an effective method to improve the properties of nano thin films [63, 64]. In terms of piezoelectricity, dopants and heterostructures can induce further lattice distortion to heighten the asymmetry, which is beneficial to piezoelectric coefficients. For example, piezoelectricity can be induced into graphene, a non-piezoelectric material, by adatom adsorption and/or incorporating defects [65]; 5) orientation. The piezoelectric coefficient varies with different exposed crystal orientation; 6) complex structure design. Although material plays a major role in the device performance, the structure is also a nonnegligible parameter contributing to the integrated devices [66]. For example, pyramid type contact gives five times higher output due to the decreased contact area in comparison to flat contact [67]. Hence, deposition method is required to deposit conformal films on complex 3D structures.

The main difference between ALD and other widely used thin film synthesis methods in piezoelectric device field (i.e., solution methods and CVD) is their growth mechanism. The growth rate of ALD process is linear; therefore, the thickness of the resulting thin film could be controlled by the number of deposition cycles. Other deposition methods are either time- or power- or concentration-dependent. In addition to these major parameters, some other parameters could also affect the results of solution methods and CVD, e.g., the spin speed during spin coating and the distance between sample and precursor inside CVD furnace [68, 69]. Even though these methods have been proved to synthesize high quality films such as monolayers [70-72], the parameters control is more difficult than that of ALD method [59].

The lower thermal budget is another attractive characteristic of ALD in comparison to other methods (especially CVD). This allows the direct integration of flexible/stretchable devices. It should be noted that temperature will affect the crystallinity, which is related to the piezoelectric properties. The simultaneous exposure to plasma during ALD process has been proved to be an effective method to increase the crystallinity without introducing too much thermal budget [73].

For the piezoelectric properties, which have been extensively explored in TMDs and MOs by the means of both experiments and simulations [74]. ALD with above-mentioned features is one of the best preparation methods among other techniques. Vast types of TMD and MO have been fabricated by ALD and then showed superior piezoelectric performance, such as MoS_2 [59, 75], SnS_2/SnS [74, 76, 77], and ZnO [78, 79], etc. It is known that the piezoelectric property is usually related to the thickness of the resulting thin film [79, 80].

Under this circumstance, ALD becomes a powerful tool to achieve facile deposition of thin films with various thickness by controlling corresponding number of deposition cycles.

3. Transition Metal Dichalcogenides (TMDs) Piezoelectric Material

3.1. Intrinsic piezoelectric properties of TMDs

TMD structure consists of lamellar atomic planes that interacted via the van der Waals force. TMDs have attracted considerable attention from researchers worldwide due to their remarkable potential applications in sensors, nano-generators, electronics and energy conversion [28, 81-86]. The predicted piezoelectric properties of TMDs are shown in Fig. 4a-c. As bulk materials, TMDs exhibit a minute piezoelectric response owing to the elimination effect of the oppositely directed symmetry of the adjacent atomic layers [28, 87, 88]. However, it has been proven that the two-dimensional (2D) TMDs show a significant piezoelectric effect when their thickness is thinned to a few atomic layers (especially when the number of layers is an odd number, as shown in Fig. 4d). This is due to the breaking of inversion symmetry [28, 89-91]. Furthermore, a giant piezoelectricity has been reported in monolayer MoS_2 and WS_2 [91, 92]. For example, the reported monolayer MoS_2-based piezoelectric nanogenerator exhibited a power density of about 2 mWm^{-2} and an energy conversion efficiency of about 5.08% with the output voltage and current of 15mV and 20pA, respectively [28]. Ronan et al. and Sujoy et al. reviewed the development history and principle of piezoelectric two-dimensional materials, along with the piezoelectric properties of 2D materials from both simulation and experiment [93, 94].

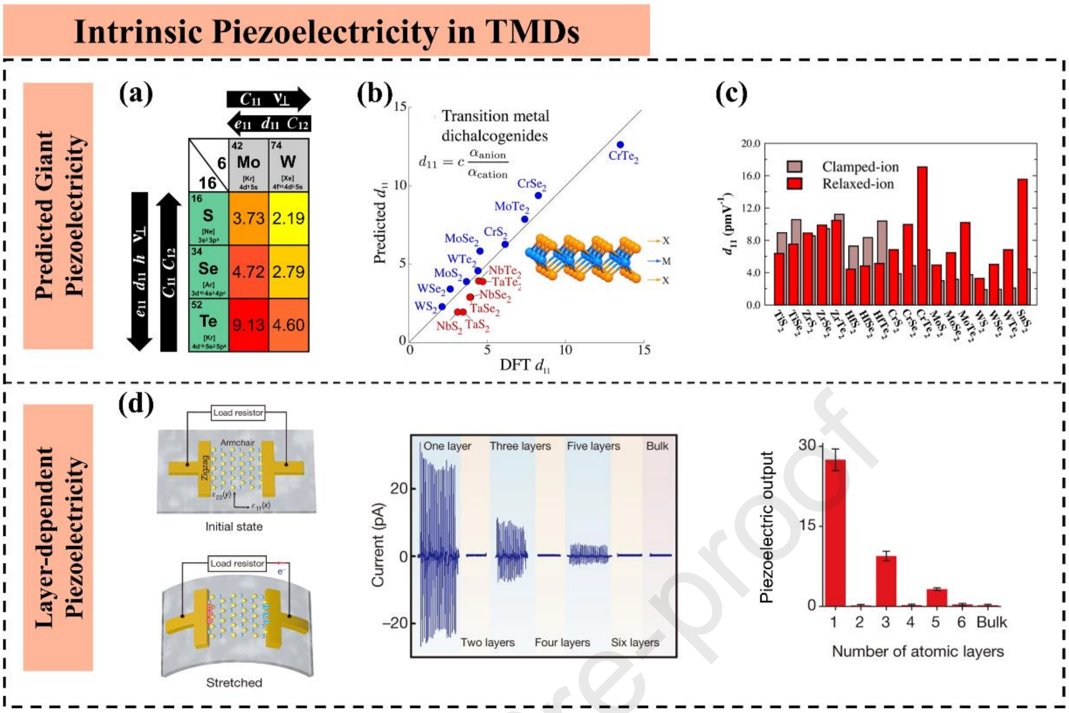

Figure 4. Summary of intrinsic piezoelectricity in TMDs. (a) relaxed-ion structural, elastic and d11 trends in 2H-MX_2, where M = Mo or W, and X = S, Se, or Te. Reproduced with permission from the study by Duerloo et al. [87]. Copyright 2012, American Chemical Society. (b) the relationship between d11 and the ratio of the anion and cation polarizabilities of TMDs. Reproduced with permission from the study by Blonsky et al. [90]. Copyright 2015, American Chemical Society. (c) d_{11} of TMDs calculated by clamped and relaxed ion. Reproduced with permission from the study by Alyoruk et al. [95]. Copyright 2015, American Chemical Society. (d) layer dependent piezoelectricity of MoS_2. Reproduced with permission from the study by Wu et al. [28]. Copyright 2014, Springer Nature.

Although these successful examples have highlighted the intrinsic piezoelectric properties of TMDs, there are several issues to be addressed, which currently limit their facile and scalable applications. One of the major issues is the preparation of a large-scale thickness controlled TMD thin film. For instance, MoS_2 with one of the highest reported piezoelectricity was prepared through mechanical exfoliation. The mechanical exfoliation technique does not allow the formation of a large-area and conformal MoS_2 layer [91]. The use of mechanically exfoliated MoS_2 flakes prevents the MoS_2 thin films from being reproduced reliably or consistently in an array, as these flakes are randomly formed and transferred. Moreover, current fabrication methods also do not allow uniform MoS_2 deposition on high-aspect ratio 3D features (or substrate) that enhance the piezoelectric effect. To date, most of the preparation work of TMDs piezoelectric material are focused on the CVD methodology [96-98]. The CVD-synthesized TMDs piezoelectric material exhibit a better adhesion to their substrate compared to mechanical exfoliation due to the thermal effect during CVD processes. Nevertheless, with the increasing demand for flexible devices, post transfer of CVD-prepared TMDs is becoming inevitable, which may introduce uneven atomic layer undermining the overall piezoelectric performance of the devices [99, 100].

The other major issue is the need for a facile and scalable method to prepare TMDs with an enhanced piezoelectric performance. Doping and heterostructures have been demonstrated as effective methods to improve the piezoelectric response of traditional piezoelectric materials, such as $BaTiO_3$ and ZnO_2 [101-104]. Compared with the zero-bandgap graphene, altering the bandgap of TMDs by changing the dopants and their concentration could easily improve their electronic and optoelectronic performance [105-107]. Hence, the preparation of TMDs layer with a controllable thickness, precise stoichiometric ratio, suitable dopant with optimum concentration, and a scalable process would path the way to wider piezoelectric applications.

ZnS, one of the TMDs, was first prepared by ALD in 1977 [108], suggesting the potential of ALD process to replace the traditionally used CVD and PVD techniques owing to its self-limited and atomic cyclic nature. Conformal thin films with controllable thickness and precise stoichiometric ratio are easily achieved via ALD due to its unique growth mechanism [26, 109, 110]. Furthermore, binary, ternary and quaternary composites or doped structures could be easily obtained by altering the cycle ratio of different precursors [111-115]. Compared to CVD, ALD process normally runs under lower temperatures [109], which provides the chance to deposit TMDs on the flexible substrate [116]. Plasma and in-situ annealing could be employed to increase the crystallinity of ALD-deposited TMDs [117-119]. Additionally, ALD technique allows the deposition of thin films on high aspect ratio (up to 4000:1) and 3-dimentional structures. These complex geometries could offer critical effect of improvements on the performance of piezoelectric devices [66, 119-121]. Arguably, ALD will be a notable preparation method to push TMD piezoelectric devices to real usage.

In the following section of this review, we will summarize the recent progress in ALD preparation of TMDs and showcase some of the exciting applications in piezoelectric field. Table. S1 lists the precursors and reaction temperature that are available for TMDs deposition via ALD technique. In general, there are two routes to prepare TMDs by ALD. The first method is post-sulfurization of ALD-deposited Transition Metal Oxides (TMOs), while the other method is single step ALD process [75, 114, 122, 123]. Due to its low reaction temperature, lower amount of precursor can be applied to ALD chamber as compared with CVD process. Therefore, suitable and active precursors are essential to ALD reaction, resulting to the extensively development of novel precursors for ALD. $MoCl_5$, MoF_6, $Mo(CO)_6$, $Mo(thd)_3$ and $Mo(NMe_2)_4$ were introduced into the ALD fabrication of MoS_2, the most representative TMDs material [25, 75, 123-126]. To date, more types of TMD can be prepared by ALD including ReS_2, HfS_2, SnSe, which could shed light on the path toward wider applications of TMDs piezoelectric material [127-129]. In addition to the stable 2D TMDs, ALD can be also applied to deposit 2D heterostructures and multi-component TMDs (summarized in Table. S2), providing possibility for further adjusting the piezoelectric properties of the resulting materials.

3.2. Common strategies to improve the piezoelectric properties of TMDs.

TMDs, especially when their thickness are over ten atomic layers, usually feature weak piezoelectricity due to the existing inversion symmetry. Therefore, it is essential to look for novel methods addressing this issue. Consequently, some strategies are proposed to improve the piezoelectric performance of TMDs. The common employed strategies are presented in Fig. 5. The major route to piezoelectric TMDs application by ALD is depositing 2D TMDs (down to monolayer or few layers) to break the inversion symmetry. Some 3D structures are also useful to enhance the piezoelectricity by reducing the contact area or inducing higher strain, such as pyramid structure, high aspect ratio substrate or even more complex design. By taking advantages of this route, a homogeneous and giant response may be realized. Apart from the usual odd atomic layer of TMDs thin film deposition, heterostructure, multinary structure and dopant have also been proven effective in prohibiting the elimination effect of the symmetric TMD structures. Recently, ALD process has been used to deposit TMD thin films on top of flexible substrates directly without any transfer process [74], paving the way to soft piezo devices. Furthermore, establishment of special structures at micro/nanoscale can also enhance the piezoelectricity of TMDs by introducing flexoelectricity [130]. With these strategies, it is promising to push their performance to the theoretical limit (Fig. 4a-c) and beyond.

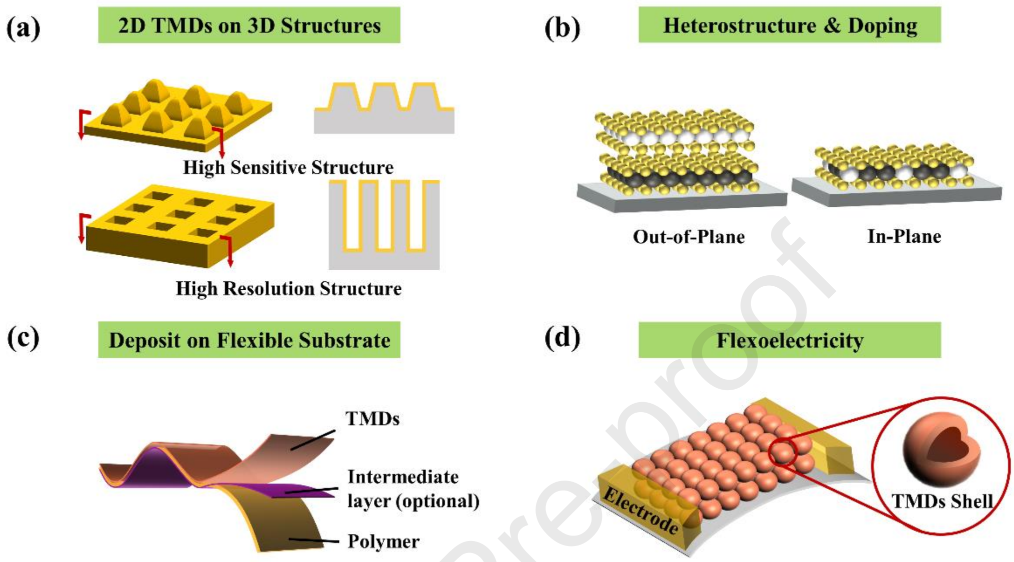

Figure 5. Common strategies to improve the piezoelectricity properties of the TMDs by ALD routes. (a) Conformal 2D TMDs deposition on 3D structures. (b) heterostructures and dopants. (c) Direct deposition on flexible substrate. (d) Flexoelectricity introduced by applying special structures.

Temperature/pulse time-influenced in-plane piezoelectricity in TMDs

Duerloo et al. predicted that giant intrinsic piezoelectricity exists in 2D materias by a density functional theory (DFT) calculation in 2012 [87]. In 2014, Zhu et al. and Wu et al. observed the piezoelectric response in a monolayer MoS_2 membrane and illustrated the high application potential towards mechanical energy harvesting device (Fig. 4d) [28, 91]. It is important to note that the reported piezoelectricity of the MoS_2 layer is from the in-plane piezoelectricity rather than out-of-plane piezoelectricity [94]. This is because two identical S layers sandwich one Mo atomic layer, constructing a hexagonal lattice structure. Blonsky et al. speculated that the in-plane piezoelectric coefficient (d_{11}) of 37 types of 2D material, including TMDs, group IIA and IIB metal oxides, group III—V semiconductors and out-of-plane piezoelectric coefficient (d_{31}) for group III—V semiconductors [90]. Most of the 2D TMDs were predicted to possess excellent in-plane piezoelectric properties (Fig. 4a-c), e.g., d_{11} of 2H-$MoTe_2$ and $CrTe_2$ are 7.39 pm/V and 13.45 pm/V, while the d_{11} of ZnO is 8.65 pm/V from the simulation results, confirming the notable potential of TMDs in piezoelectric applications [90].

To date, most of research focus on the intrinsic piezoelectric performance of TMDs, proving that the piezoelectric response only exists in an odd layer TMD (such as MoS_2, WS_2) due to the absence of inversion symmetry [87, 90]. Therefore, fabrication of a thickness controlled TMD thin film (or even monolayer TMD) is an obstacle to the wider application of TMDs-based piezoelectric devices. One idea is to use the self-limited ALD to prepare TMDs with precise atomic layer number. Song et al. developed a two-step route comprising ALD and post sulfurization process to get a conformal, crystal and continuous MoS_2 with monolayer, bilayer and triple layer thickness [114]. In addition to the two-step preparation method, ALD technique can also grow TMDs thin film directly. Lee et al. reported the success of ALD fabrication of MoS_2 using $MoCl_5$ and H_2S at 300 °C, showing a good conformity at the wafer scale with partial crystallinity [75]. Due to the erosion effect and the high reaction temperature of $MoCl_5$, $Mo(CO)_6$ was employed instead as Mo source in ALD process [124]. A conformal and layer-number controlled TMDs thin film also was prepared by this new route with better physical properties.

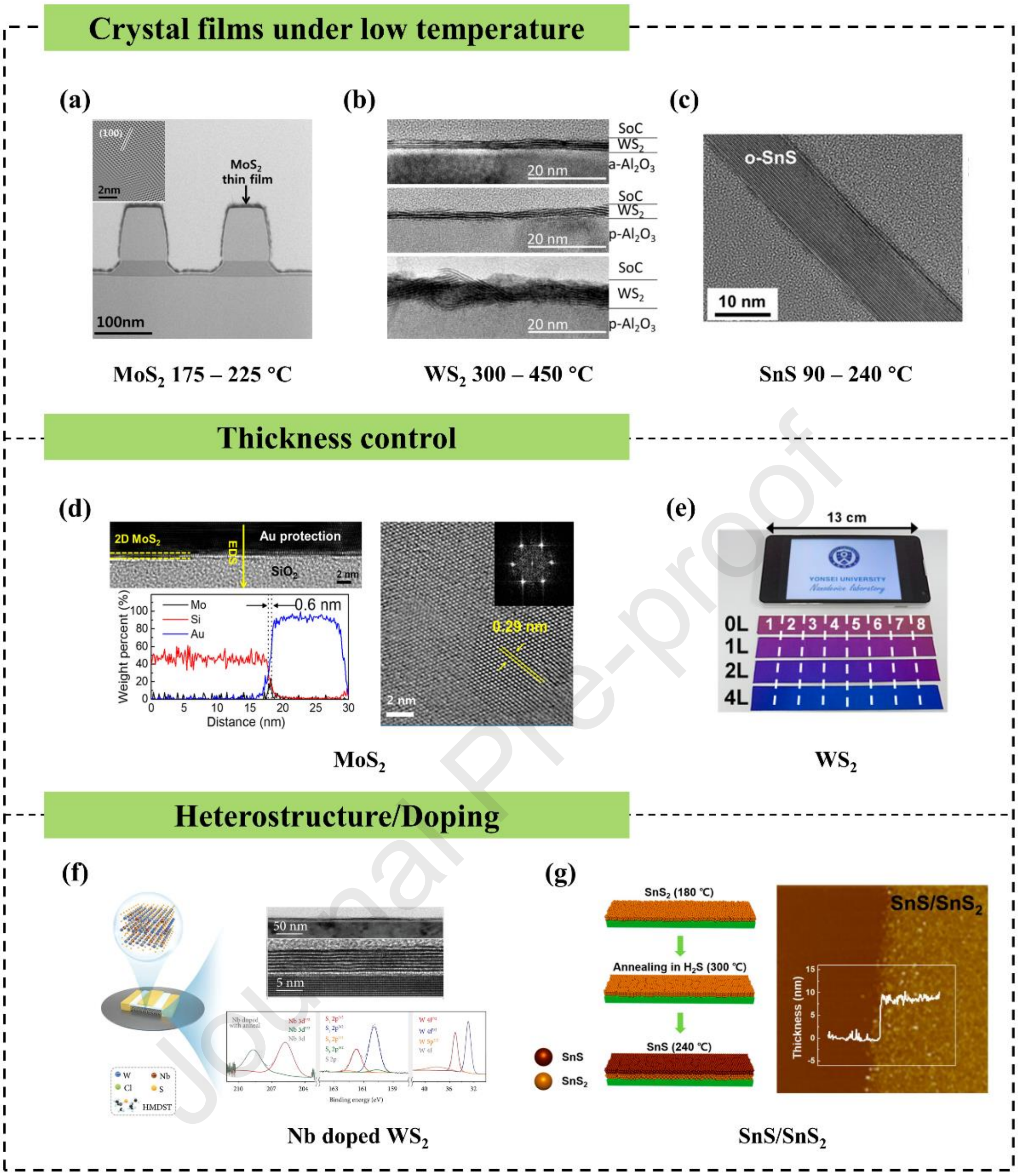

Figure 6. The capability of ALD to control the thickness, composition of TMDs under low temperature. Crystal films of MoS_2 (a) [123], WS_2 (b) [131]and SnS (c) [77] deposited under low temperature by ALD. Reproduced with permission from the study by Jang et al. Copyright 2016, Elsevier; Groven et al. Copyright 2017, American Chemical Society; Baek et al. Copyright 2017, American Chemical Society; Demonstration of monolayer MoS_2 (d) [59] and WS_2 (e) [132] deposited by ALD. Reproduced with permission from the study by Kim et al. Copyright 2021, American Chemical Society; Song et al. Copyright 2013, American Chemical Society; Demonstration of Nb doped WS_2 (f) [133] and SnS/SnS_2

heterostructure (g) [74]. Reproduced with permission from the study by Yang et al. Copyright 2021, AAAS; Cao et al. Copyright 2021, American Chemical Society.

Linear growth rate is the key of controlling the thickness in depositing TMDs by ALD or any other techniques. Linear growth rate represents the thickness controllability by adjusting one or two parameters (normally cycle number in ALD process). Although the selection of precursor determines the growth temperature and rate, the pulse time of precursor also plays an important role in controlling these characteristics. Monolayer MoS_2 was successfully deposited through only one ALD cycle by increasing the pulse number of $Mo(CO)_6$ in one cycle (Fig. 6d) [59]. The GPC was 0.03 nm/pulse while the GPC is 0.11 nm/cycle for common ALD process. The monolayer prepared in one ALD cycle was resulted from the increasing pulse number of precursor in one ALD cycle to realize saturated adsorption. The deposited MoS2 was annealed for 3mins at 900 °C via rapid thermal annealing (only 45s ramping time). Another example shows the feasible preparation of 1 layer, 2 layers and 4 layers WS_2 by adjusting the cycle number (Fig. 6e) [132]. The researchers only used common ALD route here with the GPC of about 0.08nm/cycle. The 2D crystal WS_2 was obtained by post-annealing/sulfurization at 1000 °C for 30mins. Both two examples show the thickness controllability of ALD. However, the growth temperatures of these two examples are both slightly higher than other reported works while same precursors involved in the processes, which is opposite to common sense: the chemical reaction will slow down under low temperature, prohibiting the severe aggregation of nucleation sites in initial stage and providing the possibility of lateral growth instead of vertical growth. This is because the precursors used in these two examples were metal-organic precursors, possessing larger position when adsorbed to a surface in comparison to inorganic precursors. Slightly increasing the temperature can lead to partial decomposition of these organic precursors but not affect the linear growth characteristic. Therefore, in addition to tailoring the cycle number, adjusting the pulse time and growth temperature are also essential to the deposition of monolayer or few layers TMDs. With the capability of controlling the thickness precisely of ALD, the in-plane piezoelectricity of TMDs can be approached to their theoretical limits.

Deposit crystal TMDs via ALD under low temperature

Due to the low reaction temperature during ALD process, the as-deposited TMDs tend to be amorphous, but high crystallinity is indispensable to good piezoelectric response. Depositing crystal films under lower temperature is another critical issue impeding the application of TMDs. Plasma-enhanced ALD (PEALD) is an important method to solve this problem. During thermal ALD process (TALD), a half of the cycle is mainly reactants injection, such as H_2O, O_3 (for oxide deposition) and DMDS (for sulfide deposition), where high temperatures are needed to promote the reaction between these reactants and the precursors. However, crystal products are still difficult to achieve unless a higher temperature provided during TALD. For PEALD process, reactants are replaced by O_2 and H_2S plasma, which can facilitate the deposition process. Crystal films can be deposited by PEALD due to the high reactivity of the plasma species without increasing the temperature. There are numerous demonstrations for the use of PEALD in the preparation of TMDs. For example, crystal MoS_2 can be directly deposited by $Mo(CO)_6$ and H_2S plasma at the deposition temperature of about 200 °C, which also show a conformal film prepared on a 3D substrate (Fig. 6a) [123]. Benjamin et al. developed PEALD route with WF_6, H_2 plasma and H_2S to prepare strongly textured, nanocrystalline WS_2 at 300 °C (Fig. 6b) [131]. In addition, exploration to other TMDs deposition processes is also necessary, where high temperature is not compulsory. For instance, crystal SnS can be deposited by thermal ALD under 240 °C (Fig. 6c) and shows good piezoelectric properties [74, 77].

Introduce dopant/heterostructure to TMDs by ALD

While crystal TMDs with good thickness controllability play a substantial role in approaching their simulated piezoelectric properties, further improving their performance is the third issue to be addressed. Doping and heterostructure are widely used to enhance performance in many fields and piezoelectric devices as well. For example, Yuan et al. demonstrated a novel In_2Se_3/MoS_2 heterostructure with a significant enhanced out-of-plane piezoelectric performance. The out-of-plane piezoelectric coefficient d_{33} was about 17.5 pm/V for the prepared heterostructure [134]. Similarly, Yu et al. conducted a DFT calculation on the piezoelectric response of WSe_2 /MoS_2 heterostructure and revealed that the output voltage of this heterostructure could be 0.137 eV and 0.183 eV under 4% and 8% tensile strain condition, respectively [135]. However, rare examples relate to this at film scale or even wafer scale. Most of present investigations are focused on the aforementioned two issues (thickness control and crystallinity at film scale) and doping/heterostructure at nano/micron scales (e.g., 2D TMD nano flakes) [136]. As direct bandgap semiconductor materials, the bandgaps of TMDs can be easily adjusted by introducing dopant, which is firmly correlated to piezoelectric coefficient. The efficient dopant concentration controllability in 2D TMDs film is an emerging point although there are several trials now (Nb doped WS_2 shown in Fig. 6f) [133]. On the other hand, heterostructure has been demonstrated to show better piezoelectric properties in comparison to pure TMD, e.g., the d_{33} of in-situ ALD prepared SnS/SnS_2 heterostructure is ~4.75 pm/V, while SnS is only ~1.85 pm/V (Fig. 6g) [74].

Due to the layer-by-layer growth mechanism of ALD, it is a promising way to introduce dopant and heterostructure. However, there are some scientific issues to be solved in terms of this direction: a) expand the ALD window, dopant precursor requires identical growth temperature as substrate precursor; b) simulation or artificial intelligence assisted method, versatile dopant/heterostructure can be applied to TMDs but not every route is effective; c) thickness-concentration-balance, balancing the thickness and dopant concentration is also an important point toward to further improvement due to the layer-dependent piezoelectricity of TMDs.

Direct flexible device integration

The range between 100 °C and 300 °C is the melting point and/or glass transition temperature of most of the widely used polymers in soft electronics. CVD is a general technique to deposit TMDs, but it is not suitable for those flexible substrates with a lower melting temperature (below 300 °C). To meet the high demand for the flexible and portable devices currently, CVD-prepared films must be transferred to soft substrates by wet/dry transfer methods, which will significantly undermine the conformability and lead to contamination of the resulting thin films [62]. Compared to CVD, ALD runs under a lower temperature, which is suitable for most of the flexible substrate. To keep the precise thickness of TMDs on polymers, a pre-treatment of depositing an intermediate layer could be employed on polymers to avoid apparent uneven on the surface [116]. Viet et al. demonstrated direct SnS/SnS_2 piezoelectric nanogenerator deposition on polyimide (PI) with the highest output of 60 mV, 11.4 nA/cm^2, confirming the feasibility of direct flexible device integration by ALD (Fig. 7a) [74]. For the direct deposition on polymers by ALD, more attention should be given to the functional group of polymers and the possible chemical reaction between the precursors and polymers.

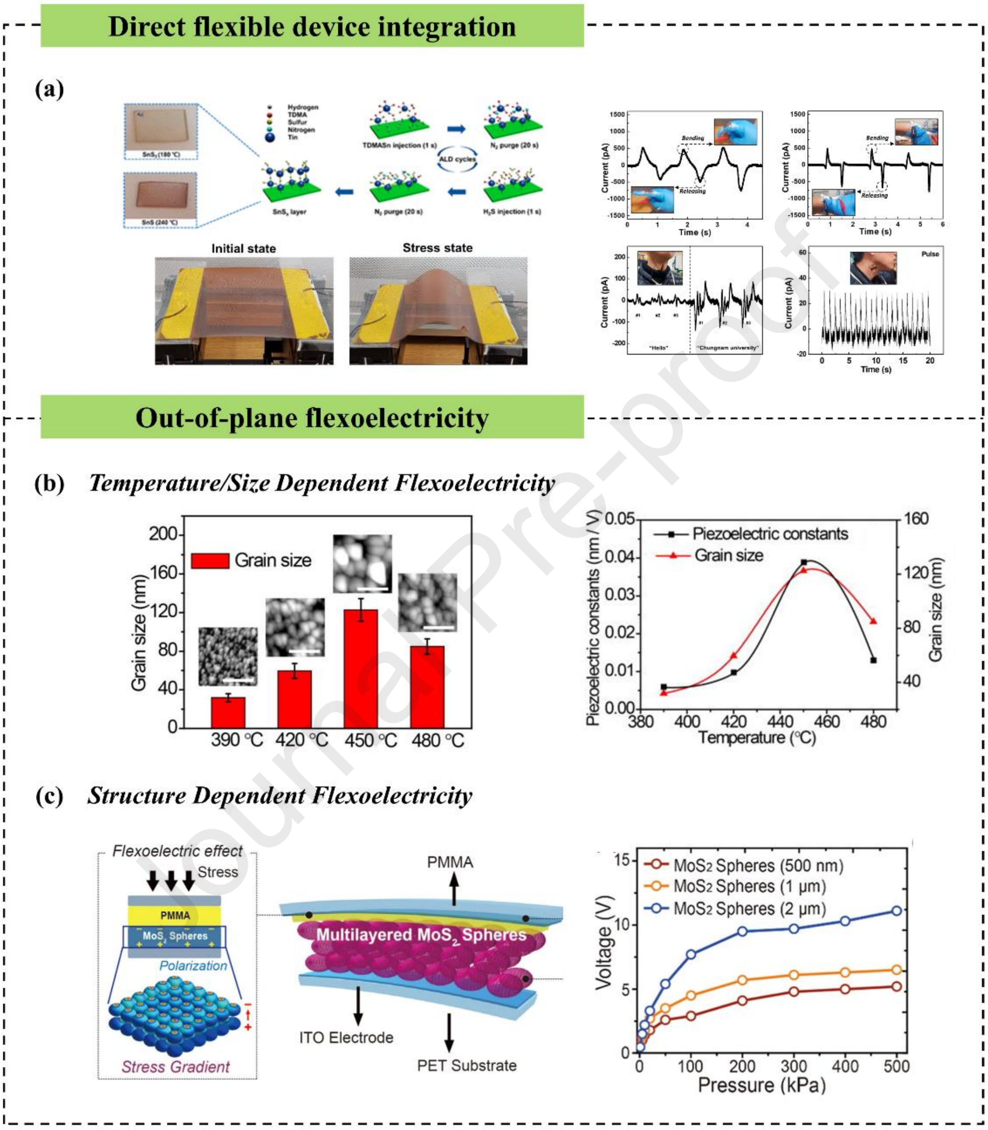

Figure 7. Soft Integration and performance-parameter relationship of TMDs piezoelectric devices deposited by ALD. (a) In-situ integration of SnS_2/SnS piezoelectric device via ALD. Reproduced with permission from the study by Cao et al. [74]. Copyright 2021, American Chemical Society. (b) Temperature/size dependent flexoelectricity of MoS_2. Reproduced with permission from the study by Huang et al. [137]. Copyright 2017, AIP Publishing. (c) Structure dependent flexoelectricity of MoS_2. Reproduced with permission from the study by Kim et al. [130]. Copyright 2021, American Chemical Society.

Effect of temperature/size and structure on out-of-plane piezoelectricity/flexoelectricity

Although there is almost no out-of-plane piezoelectricity in 2D TMDs, the out-of-plane flexoelectric response can be detected in such materials due to some specific surface morphologies or device structures [138-140]. The flexoelectric effect points to an electric polarization response under a gradient mechanical strain condition. In 1964, Kogan introduced the concept of flexoelectricity by investigating the inhomogeneous deformation induced electric polarization in a symmetric crystal [140]. With this effect, mechanical-electricity transformation can be introduced into materials with crystal center of symmetry. Furthermore, flexoelectric effect has been proved exist in MoS_2, $MoTe_2$, $MoSe_2$, WS_2, WSe_2 [141-143].

For the as ALD-deposited MoS_2 thin film, surface roughness or morphologies would introduce an out-of-plane flexoelectricity. Huang et al. reported the size-dependent piezoelectricity of the MoS_2 films prepared by ALD technique [137]. The self-limiting nature of ALD lead to the controllable thickness and morphologies of the films via the adjustment of the cycle number and temperature. When the reaction temperature increased from 390 °C to 480 °C, the grain size of MoS_2 gradually grew and reached a summit at 450 °C with the grain size of ~122.6 nm. Above 450 °C, the grain size decreased with further increment of the temperature. Under the PFM (piezoresponse force microscopy) test, the piezoelectric response for these samples showed a similar linear trend to their grain size (Fig. 7b) [137]. This phenomenon can be explained by the principle of flexoelectricity: larger diameter of the grain size, stronger the flexoelectric effect, which can be tailored by ALD growth temperature. Jin et al. fabricated 2D MoS_2 sphere piezoelectric nanogenerators (PNGs) with an output voltage of about 1.2 V under the pressure of around 4.2 kPa [144], which was a successful trial for the TMDs based flexoelectricity enhanced energy harvesting devices. Although the technology they used was thermal decomposition deposition instead of ALD, it should be noted that this process can be easily replicated using ALD technique. The output of the flexoelectric device varied with the diameter of the MoS_2 sphere [130]. The trend is: larger sphere, higher output (Fig. 7c). These successful examples show the potential to increase the piezoelectricity of TMDs by taking advantages of conformal deposition by ALD. However, there should be a balance between the output and stability of the integrated devices: larger curvature, higher output, lower stability.

In summary, ALD technique has shown high potential in the applications of piezoelectric TMD-based materials with the advantages of self-limiting, precise thickness controlling and conformal membrane on 3D structures. Precise thickness control is the key to induce the intrinsic piezoelectricity of TMDs, which is the most attractive feature of ALD technique in the thin film deposition. To significantly enhance the piezoelectricity of TMDs, introducing dopants, heterostructures and vacancies via ALD are effective ways of breaking inversion symmetry further. By taking advantage of conformal deposition on 3D structure of ALD, out-of-plane piezoelectricity can be easily introduced in TMDs by flexoelectric phenomenon. On the other hand, these 3D structures can further increase the piezoelectric output by reducing the contact area or inducing higher strain. Moreover, ALD is expected to remove the need of thin film transfer process to satisfy the demanding in flexible electronics industry due to its lower deposition temperature. With these benefits, giant in-plane piezoelectricity and/or out-of-plane piezoelectricity can be induced into the TMDs, making it as novel piezoelectric materials.

4. Metal Oxide Piezoelectric Materials

4.1. Intrinsic piezoelectric properties of metal oxides

Due to their low cost and high environmental stability, metal oxides (MOs) have been extensively studied for piezoelectric applications [145, 146]. In recent theoretical simulation, monolayer MOs have been found to exhibit relatively high piezoelectric coefficients (Fig. 8a, b) [147], including cadmium oxide (CdO) [148, 149], hafnium oxide (HfO_2) [150-152], zinc oxide (ZnO) [24, 148, 153], and zirconium oxide (ZrO_2) [150, 154, 155], etc.

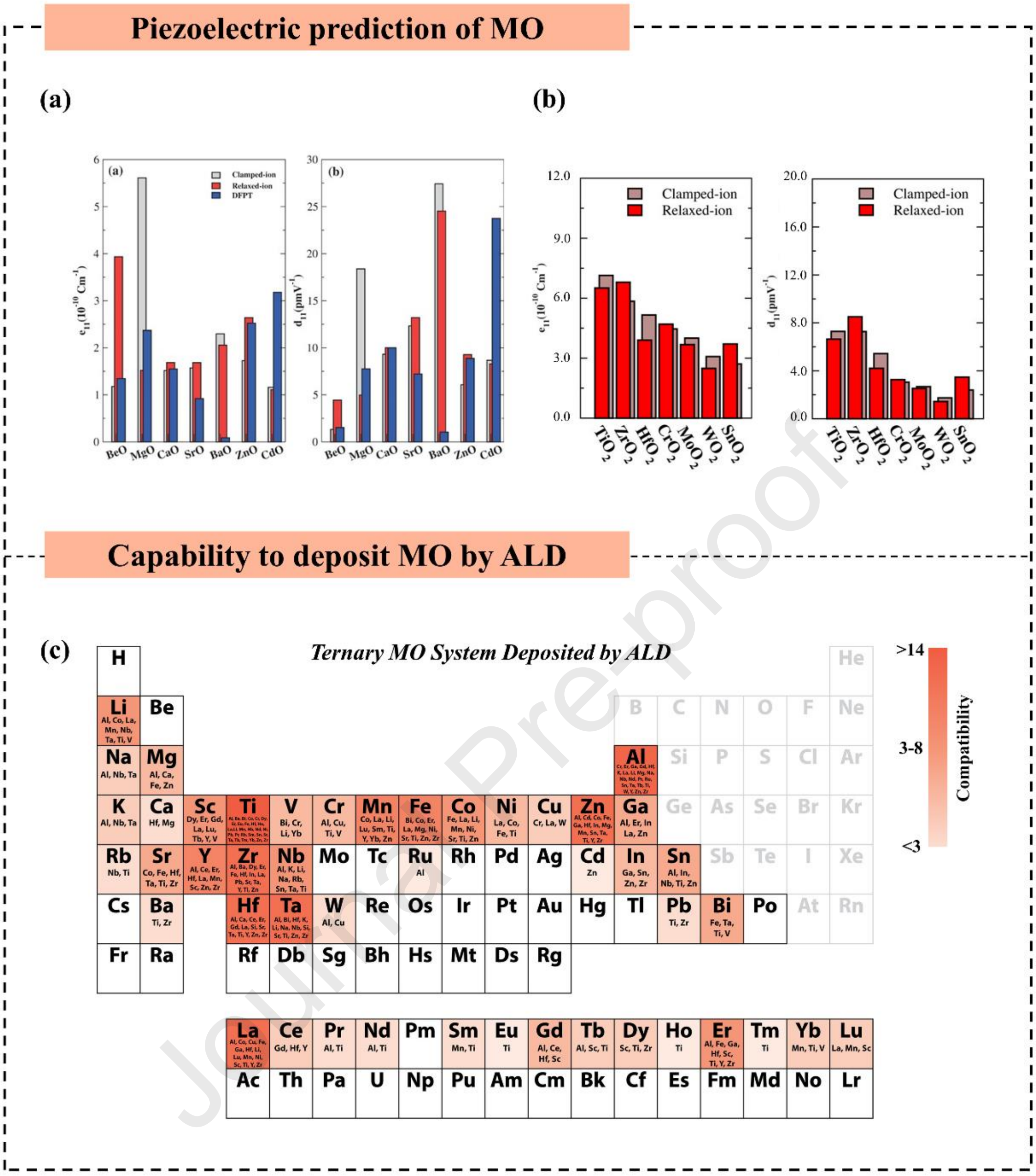

Figure 8. Piezoelectric prediction of MO and capability of ALD to deposit MO. Calculated e_{11} and d_{11} of valency II MO (a) [147] and valency IV MO (b) [95]. Reproduced with permission from the study by Alyörük et al. Copyright 2016, WILEY-VCH; Alyörük et al. Copyright 2015, American Chemical Society. (c) Ternary MO systems deposited by ALD. Reproduced with permission from the study by Mackus et al. [156]. Copyright 2018, American Chemical Society.

Some of the common methods to fabricate such MOs include hydrothermal [157-159], atomic layer deposition (ALD) [160-162], chemical vapour deposition (CVD) [163-165], chemical solution deposition [166-168], and electrochemical deposition [169-171]. ALD technique in particular is recognised for its high reproducibility, high conformity and excellent thickness control in thin film fabrication. In addition to single phase MO deposition, ALD also shows the capability of preparing binary or ternary MO systems (Fig. 8c) [156]. Due to the bandgap adjustability of MOs, binary or ternary MO systems tend to increase their piezoelectric properties. Moreover, ALD can be used to fabricate MO films into different nanostructures: one-, two- and three-dimensional nanostructures. It is important to note that nanostructure of MOs affect their piezoelectric coefficient since there is a change in the surface area-to-volume ratio, hence, affecting the extent of deformation of the structure. Table. S3 lists the precursors and reaction temperature that are available for MOs deposition via ALD.

4.2. Common strategies to improve the piezoelectricity of MOs

Although varieties of MOs were predicted to have strong piezoelectricity (Fig. 8a, b), only a portion of them were studied. Among these piezoelectric MOs, Zinc Oxide (ZnO) is the most widely investigated one due to its wide bandgap (~3.7 eV), large piezoelectric coefficient in d_{33} direction (as high as 50 pm/V), feasible preparation and versatile nanostructures (such as nanowire, nanorod, nanoflower and nanowall) [19]. However, the present fabrication route of piezoelectric MOs is mainly solution-based process, e.g., hydrothermal, lacks the abilities of controlling the thickness of MOs and dopant's concentration. These are not beneficial to their piezoelectric properties and meet the emerging demand for piezotronics, which requires compact and thin piezoelectric MOs layer. ALD can be applied to MO piezoelectric devices in the following aspects (Fig. 9): a) Deposit MO seed layer by ALD. MO nanorod is one of the most attractive structures for piezoelectric application due to its aligned orientation. A compact and thin seed layer is necessary for the following nanorod growth. The size and density of the nanorods, determining the piezoelectric performance, are highly dependent on the coverage and quality of the seed layer. With decent quality and thickness customization of the deposited seed layer, ALD can be used to control the dimension and the quality of nanorods, therefore tuning the piezoelectric output; b) Down to 2D MOs by taking advantage of ALD technique. Although MOs show stable piezoelectric properties in the bulk state, their piezoelectric properties may significantly vary when thickness are reduced to few atomic layers. For example, ZnO shows the layer-dependent piezoelectricity, where its d_{33} varies from ~9.9 pm/V in bulk state to ~25 pm/V in nanobelt with the thickness of ~60 nm [172]. The robust thin MOs with outstanding piezoelectric properties show the potential in piezotronics (including the applications of piezo-controlled field-effect transistor and logic circuit). For example, 2D ZnO was reported to act as gate by piezoelectric polarization charges [30] to realize ultrafast reaction in transistors. The performance of such transistors is determined by the thickness and quality of the MO films. Therefore, ALD can be used to tailor the device performance by tuning the thickness of the deposited MO films; c) Doping/composite. Doping and composite are the effective ways to further enhance the piezoelectric properties of MOs by tuning their crystal structures and band structures. ALD is one of the most attractive methods to achieve doping and composite fabrication. “Super cycle” ALD has been developed to introduce dopants by incorporating x cycles AO deposition and y cycles dopant BO deposition in one “super cycle”. The dopant concentration can be easily controlled by changing the ratio of x/y. The doped films can be homogeneous due to the low growth rate of each material and the layer-by-layer deposition characteristics. In addition, *in-situ* or *ex-situ* composite/heterostructure can also be prepared by ALD process due to the low thermal budget, which would not destroy the as-deposited first layer (denoted as AO layer in Fig. 9). ALD *in-situ* composite can be fabricated by depositing AO with specific thickness followed by deposition of BO. This process may require the adjustment of deposition temperatures, while same temperatures are applied during “super cycle”. The difference between “super cycle” and *in-situ* composite is the clear interface can be only observed from in-situ composite. Due to the low growth rate and the small injection dose for dopant in one “super cycle”, the doped MOs are more homogeneous than *in-situ* composites. Moreover, ex-situ deposition of MOs on top of another prepared films can be easily achieved by ALD, which has been widely applied in electronics. d) 3D devices integration. In addition to the intrinsic properties of the active materials, piezoelectric output is highly dependent on the device structure. The structure engineering is a feasible way to adjust the piezoelectric performance at device scale by tuning its deforming mechanism. ALD is probably the best technique to achieve conformal 2D films on 3D structures due to its self-limit characteristic. With the 3D integrated device, the performance can be further enhanced.

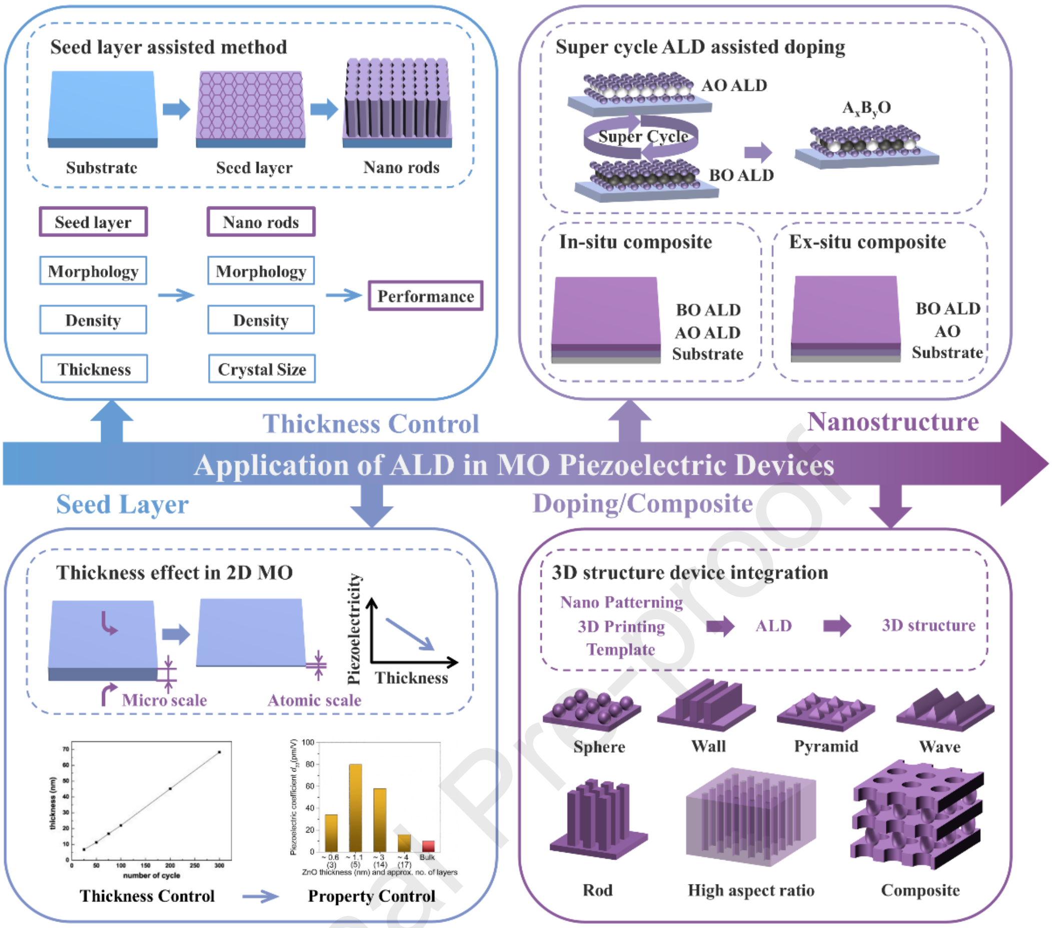

Figure 9. Application of ALD in MO piezoelectric devices. Reproduced with permission from the study by Park et al. [173]. Copyright 2006, IOP Publishing; Reproduced with permission from the study by Mahmood et al. [172]. Copyright 2021, Elsevier.

Manipulate MO nanorod by ALD-deposited seed

Due to the anisotropic piezoelectricity of MOs, nanorod/nanowire structures with aligned orientation are preferred during the fabrication process. For example, the largest piezoelectric coefficient of ZnO is along the c-axis direction (002 direction) [174]. Therefore, controlling the MO nanorod with preferred orientation and good alignment is the key to increase their piezoelectricity. Although tailoring the orientation by adjusting the parameters during hydrothermal process attracts lots of attentions, the quality of pre-deposited MOs seed layer also plays an important role in determining the orientation, density and diameter of nanorods [175-177]. However, most of the present routes utilize spin-coating, PVD, magnetron sputtering to prepare the seed layers without a precise control in their qualities [178, 179]. ALD is a better alternative in seed layer deposition with decent characteristics. The orientation of MO nanorod can be guided by altering the ALD cycle number of seed layer, i.e., the thickness of the seed layer (Fig. 10a) [176]. The direction of ZnO nanorod distributed from acute angle, right angle and bimodal angle with the increasing of ALD cycle number of seed layer, where the thickness of seed layer gradually increased from ~2.88 nm to ~115 nm. In addition to orientation, the density and diameter of ZnO nanorod also can be determined by the ALD cycle number of seed layer (Fig. 10b) [177]. These models show the effective adjustment of MO nanorod growth by taking the advantage of ALD controllability on materials at nanoscale. Moreover, ALD can be applied to grow MO nanorod or other structures with the combination of templates, e.g., porous anodic aluminium oxide (AAO) is commonly used as a nano-template to fabricate MO 1D nanostructures in ALD [180].

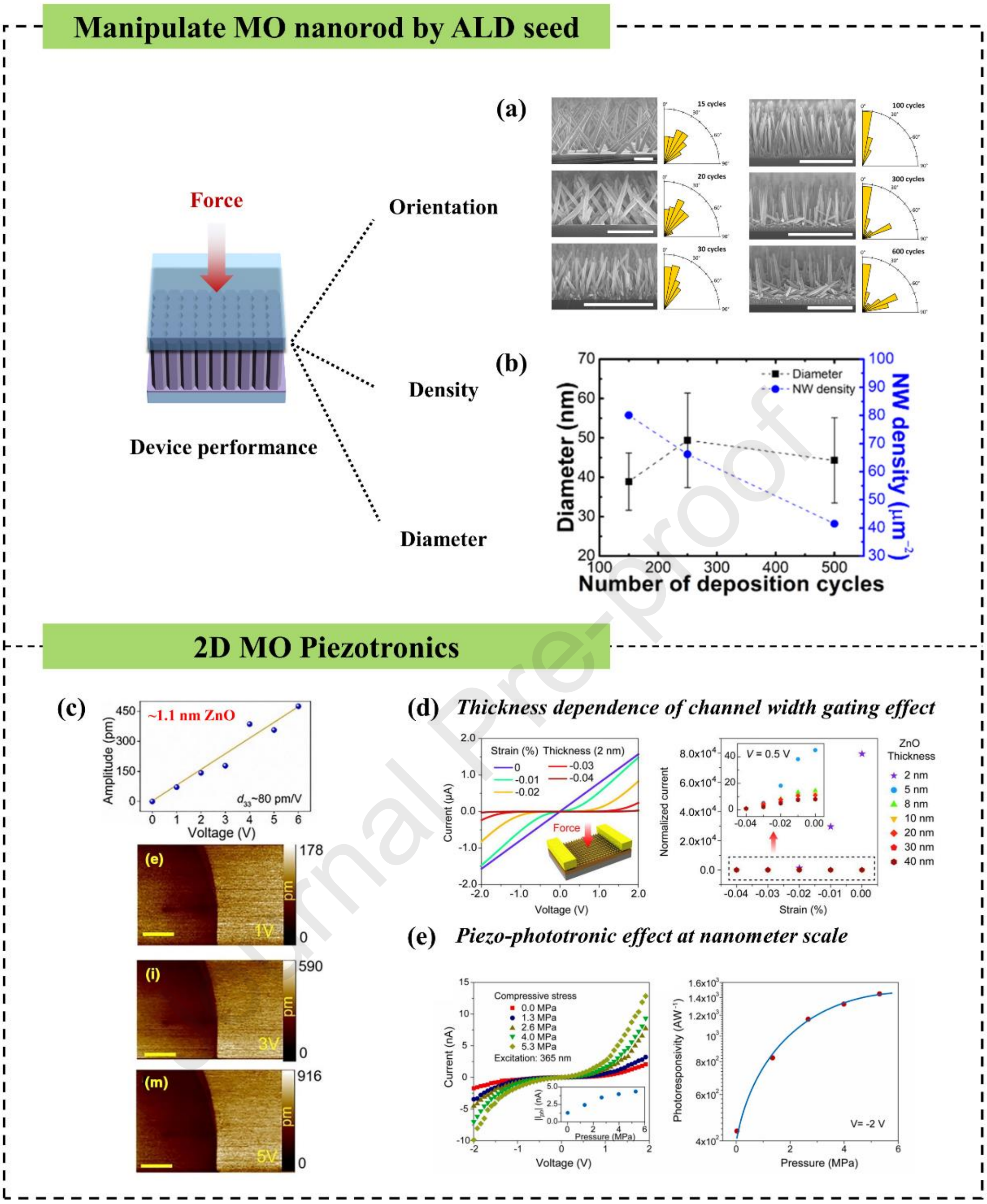

Figure 10. Use ALD to manipulate MO nanorod and 2D MO towards piezotronics application. (a) Adjust ZnO nanowire orientation by changing the thickness of seed layer, Scale bar =1 μm. Reproduced with permission from the study by Bielinski et al. [176]. Copyright 2015, American Chemical Society. (b) The effect ALD cycle number on the diameter and nanowire density of ZnO. Reproduced with permission from the study by Galan-Gonzalez et al. [177]. Copyright 2019, IOP Publishing. (c) Layer dependent piezoelectricity of 2D ZnO. Reproduced with permission from the study by Mahmood et al. [172]. Copyright 2021, Elsevier. (d) Channel width gating effect. Reproduced with

permission from the study by Wang et al. [30]. Copyright 2019, Elsevier. (e) Nanoscale piezo-phototronic effect. Reproduced with permission from the study by An et al. [181]. Copyright 2021, Elsevier.

2D MO piezotronics

While 1D nanostructures allow high sensitivity, 2D thin films contribute to more stable and robust devices. Down to 2D (monolayer or few layers), materials normally show distinct electrical, optical, catalysis properties in comparison to their bulk counterparts [182-185]. Although most of MO piezoelectric devices are focus on their bulk or 1D conditions, 2D MOs also show decent piezoelectricity [30, 172, 181]. This is because reducing the thickness could lead to an increase in surface area-to-volume ratio and optimised surface energy, which have been shown to introduce or enhance the piezoelectricity in MOs. ZnO has been predicted to show highest piezoelectricity when its thickness is down to 5 layers (~1.1 nm), which has been confirmed by experiments (Fig. 10c) [172]. With the increased piezoelectricity, 2D MOs exhibit the potential in 2D piezotronics, which can be used in human-machine interfaces, robotics and wearable electronics. For instance, 2D ZnO can act as a “gate” to control the conductive channel width, hence control the current path (Fig. 10d) [30]. The strain sensitivity of this field effect transistor (FET) could be enhanced by over three orders of magnitude with the decreasing of ZnO thickness (from tens of nanometre to atomic scale). Moreover, 2D ZnO FET has showed enhanced electronic transport properties under pressure, which can be applied in piezo-phototronic (Fig. 10e) [181]. With this effect, the sensitivity of the as-fabricated UV detector under pressure could be 2.3 times higher than that without pressure and over thousand times higher than commercial detectors. It should be noted that the 2D MOs were synthesized by solution methods or mechanical exfoliation. Due to the dimensional limit of these methods, the size of 2D MOs can only reach tens of microns, confining their further applications. To enlarge their effective areas, ALD can be an alternative to synthesize 2D MO films due to its advantage in thickness control. Although there are several successful trials of 2D MOs deposited by ALD [186, 187], growing single crystal 2D MOs through ALD is still a problem preventing its further improvement. Grain boundary is always regarded as defect due to the interference once carriers pass through it, so single crystal materials play an indispensable role in electronics. Deposition of single crystal MOs can be the next milestone in the development of ALD in this field. In addition to the exhibited interesting traits that are shared with their bulk counterparts, 2D MOs also show unprecedent characteristics due to the quantum confinement effect and structure change. It leads to a higher pressure/strain sensitivity by thinning the MOs.

Doping/composite

As an effective approach to introduce lattice distortion of MOs, doping/composite can significantly enhance the piezoelectricity of MOs. For example, when Zn^{2+} in ZnO is substituted with a dopant ion with a smaller ionic radius, there could be an increase in its piezoelectric coefficient due to the ease in rotation of metal ion-oxygen bonds as compared with the zinc-oxygen bonds. Additionally, when a dopant is more positively charged, such as Cr^{3+} or V^{5+}, there could be a formation of the collinear bonds of Cr–O or V–O bonds which have higher polarization than Zn–O bonds. As a result, in the presence of applied electric field, easier rotation could happen, which results in the increment of the piezoelectric coefficient of doped ZnO [104, 188]. With the capability of "super cycle" deposition, ALD is a practical method to fabricate doped MOs and/or MOs composite. In addition to binary oxide systems, such as ZnO/SnO_2 composite (Fig. 11b) [188] and N/F doped ZnO (Fig. 11c) [32], MOs/metals or MOs/TMDs composites can be also co-deposited by ALD [31, 189]. This provides more possibility in improving the piezoelectricity of MOs by versatile parameters introduction. Moreover, discontinuous nanoparticles deposited on continuous films also show advanced properties in optical, electrical and catalytic fields. Fig. 11a shows the representative ALD fabricated structure with the promising application in many fields [31]. However, most of the published works are related to the simple doping/composite fabrication by ALD with few piezoelectric applications. More focus should be paid to following aspects: a) DFT calculation of piezoelectric properties in binary, ternary MO systems to determine the effective dopant and concentration; b) investigate the compatibility of MOs with other type of materials, especially those easily oxidized materials. This is because the co-reactants of MOs in ALD process are mainly water, O_2 and O_3; c) seek the possibility of doping/composite in MOs at nanoscale, such as 2D doped MO systems.

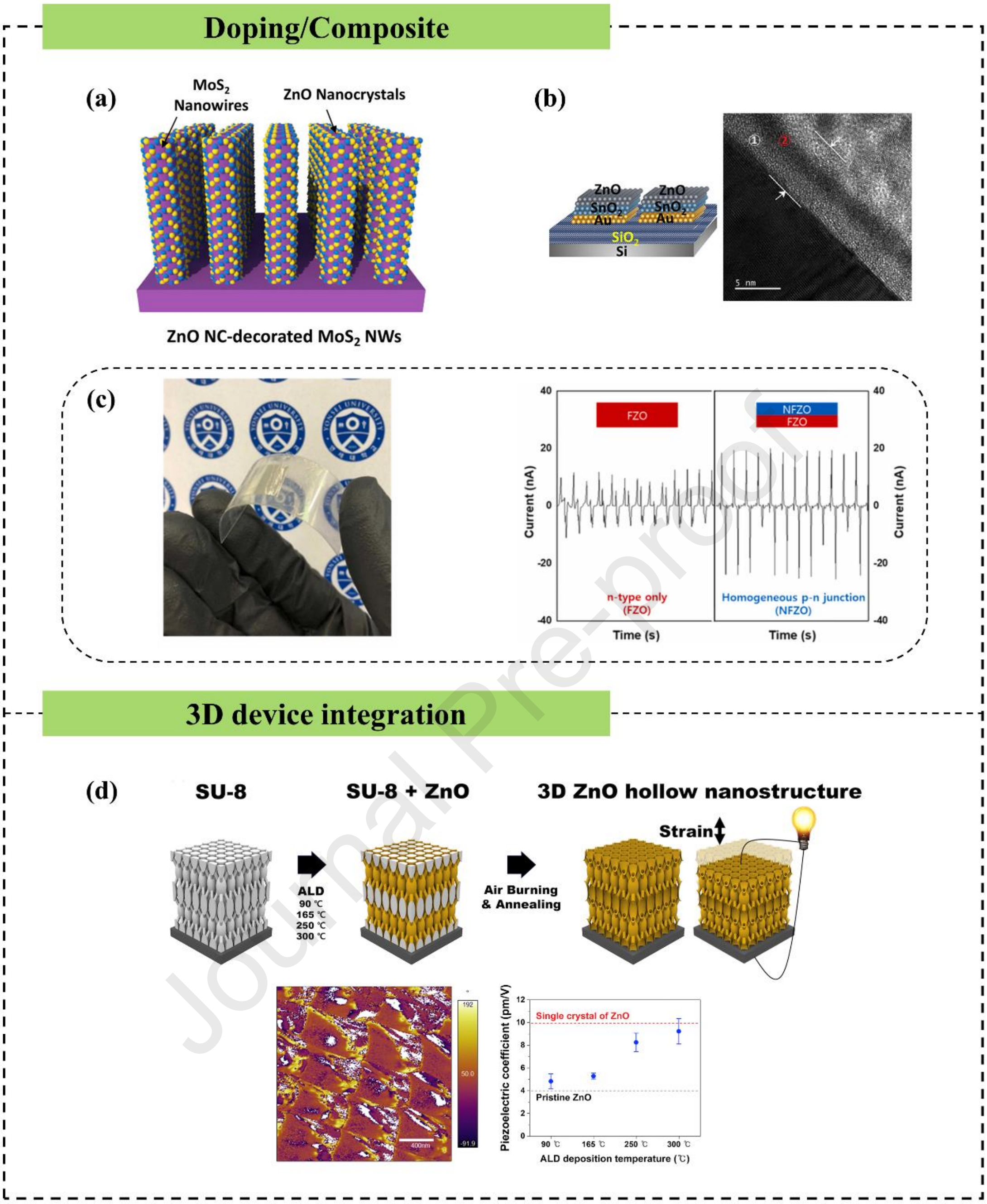

Figure 11. Enhance piezoelectricity of MO by doping/composite and 3D integration via ALD. (a) ZnO decorated MoS_2 nanowalls. Reproduced with permission from the study by Oh et al. [31]. Copyright 2020, American Chemical Society. (b) ZnO/SnO_2 composite. Reproduced with permission from the study by Lopa et al. [187]. Copyright 2023, American Chemical Society. (c) ZnO p-n junction. Reproduced with permission from the study by Kang et al. [32]. Copyright 2022, Elsevier. (d) Enhance piezoelectric performance by introducing 3D structure and increasing deposition temperature. Reproduced with permission from the study by Kim et al. [24]. Copyright 2020, Elsevier.

3D device integration

Similar to TMDs, MOs also show higher piezoelectric performance when they are in 3D nanostructures in comparison to their bulk counterparts without any structures. Combined with nanostructures, the whole devices can generate greater strain, accordingly, produce higher piezoelectric output. 3D ZnO hollow structure has been reported to deposit on epoxy template by ALD. It should be noted that the ALD deposited MOs are polycrystalline. Additionally, the piezoelectricity was positively correlated with deposition temperature. The piezoelectric coefficient of the fabricated 3D ZnO hollow nanostructure was ~9.2 pm/V with the deposition temperature of 300 °C, almost the same performance as single crystal ZnO (Fig. 11d) [24]. Although post-annealing is required to remove the organic skeletons and increase the crystallinity of MOs, lower temperature is still compulsory at the first deposition step due to the poor stability at higher temperature of the organic skeletons. Hence, PVD and CVD are not suitable here due to the drawbacks of depth penetration and high processing temperature respectively. However, the 3D device integration is limited by the resolution of skeleton/template fabrication. To look for structures with better performance, Finite Element Analysis (FEA) is needed in the future.

5. Conclusions and Prospects

As the piezoelectric devices are getting smaller and more flexible, the demand for high-performing sensors and integrated sensing systems has never been higher. Precisely tailoring the active layer of piezoelectric device is the key to address these emerging problems. ALD exhibits the potential as a thin film fabrication technique to satisfy these requirements due to its powerful capabilities in depositing materials with thickness control at the Angstrom level and uniformity at Wafer-scale. In this review, we briefly highlighted the advantages of ALD, including thickness control, conformality and composition customizing, along with the progress in ALD-based piezoelectric materials and devices ranging from 1D-, 2D- and 3D metal oxides to 2D TMDs to even complex 3D structures. The lower thermal budget and the capability of 2D materials deposition are the most attractive points of ALD. Combined with flexible substrates and/or 3D nanostructures, ALD would produce piezoelectric materials with high properties approaching or even exceed their theoretical limits, which is unattainable by other thin film fabrication techniques. With the ability of 2D piezoelectric materials deposition, ALD technique can further pave the way to piezotronics and piezo-phototronics industrials. Despite some progress, several critical issues still deserve more attention.

Expand the "ALD window"

The ALD window defines the linear growth range and consequently the quality of the deposited film. The intrinsic property of piezoelectric material is highly dependent on the film quality, which can be satisfied only when the deposition temperature is within the ALD window (the coverage-thickness balance can be reached within ALD window, as show in Fig. 2). To further enhance the piezoelectric output, doping and/or composite is one of the most effective methods by tunning the crystal structure and the band structure. However, overlapped ALD windows for matrix precursor and dopant precursors are normally required due to the fast-switching injection of different precursors in one "super cycle" (elucidated in strategies for enhancing the piezoelectric performance of MOs). Without overlapped ALD windows, only stacking hetero films (e.g., few nm film A deposited on few nm film B) with distinct in-between interface can be fabricated because few hours stabilization period is always required after adjusting the operation temperature. Therefore, expanding the ALD window can enhance the compatibility of piezoelectric materials with other dopants, hence providing the possibility to further increase the piezoelectric performance.

Challenges of single crystal materials deposition

Compared with single crystal materials, polycrystalline materials show worse piezoelectricity, and no piezoelectricity exists in amorphous materials. Crystallinity is an essential factor affecting the piezoelectricity of materials. Although some materials deposited via ALD are more likely to be amorphous due to the low reaction temperature, plasma-enhanced ALD (PEALD) can be a possible approach to overcome this problem [73]. However, the introduction of several parameters, such as plasma power, plasma duration and bias voltage, may bring more uncertainty to composition and thickness of the resulting films. To further enhance the piezoelectricity, depositing single crystal materials is next bottleneck in this field. Tailored substrate can be a promising method to fabricate single crystal materials by ALD, which has been proved effective in CVD process [190]. In addition, the introduction of extra fields also makes it possible to prepare single crystals, e.g., directional magnetic or electric fields. With the development of next generation piezoelectric devices, polycrystalline materials may not satisfy the demand for higher performance. Therefore, the effective single crystal films deposition by ALD is a research topic deserving investigation in the future.

Explore the scope of nanostructures applications

Another challenge arises from the compatibility between ALD and the structural design. While some researchers have proved structural engineering can be the promising way to further enhance the device performance [66], the specific directions in these area have not been found yet. With the developments of emerging techniques, such as 3D printing, photolithography and Micro-electromechanical systems (MEMS), a variety of nanostructures can be fabricated. Although the performance of the as-produced device is confined by the resolution of these techniques, pairing a nanostructure with specific ALD deposition route is still an issue. To deposit conformal 2D films over 3D structures, enough time for precursors to spread through the nanostructures is needed. For normal ALD process, the precursors will only stay in the chamber for hundreds of milliseconds, which is not enough for those complex nanostructures. Holding of precursors to promote the adsorption process is a possible solution. However, the adsorption groups are tending to be unstable under longer thermal period. Hence, developing effective ALD recipes are essential to enlarge the compatibility of nanostructures, which is also the key to further increase the piezoelectric performance.

It should be noted that these challenges also can be denoted as the motivation and directions for the future research of ALD-based piezoelectric materials and devices. Overall, ALD technique shows a promising route to improve the performance and expand the ranges of piezoelectric materials and devices.

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Highlights

- Atomic Layer Deposition is an attractive method to fabricate piezoelectric materials.

- Strategies to improve the piezoelectric properties of TMDs and MOs by ALD routes are proposed.

- Prospects and challenges for ALD fabricated piezoelectric materials are proposed.

Declaration of interests

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

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