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# INNOVATIVE METHODS IN SYNTHESIZING ORGANOMETAL HALIDE PEROVSKITES FOR ENHANCED SOLAR CELL EFFICIENCY

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***ABSTRACT***

*The pursuit of efficient and stable perovskite solar cells (PSCs) has led to the development of several innovative synthesis methods for organometal halide perovskites. This study reviews seven key techniques that have significantly enhanced the performance and stability of PSCs. These methods include Hybrid Chemical Vapor Deposition (HCVD), Hydrochloric Acid Vapor Annealing (HAVA), interface functionalization, cryo-controlled nucleation, the incorporation of core-shell metal nanoparticles, tantalum-doped $TiO_2$ nanorods, and sequential vacuum deposition. HCVD allows for precise control of deposition parameters, achieving efficiencies up to 11.8%. HAVA enhances structural and electronic properties by converting CH3NH3PbI3 to phase-pure CH3NH3PbCl3, increasing efficiency to 17.40%. Interface functionalization with organometallic compounds such as ferrocenyl-bis-thiophene-2-carboxylate has achieved efficiencies of 25.0% and maintained stability over long-term operation. Cryo-controlled nucleation creates highly uniform films, reaching a PCE of 21.4%. The integration of core-shell metal nanoparticles reduces exciton binding energy, achieving efficiencies of up to 11.4%. Tantalum doping in TiO2 nanorods enhances electron transport and band alignment, resulting in a record efficiency of 19.11%. Sequential vacuum deposition produces uniform, highly crystalline perovskite films, achieving efficiencies up to 15.4%. These advancements address key challenges in the scalability, stability, and efficiency of PSCs, paving the way for the commercial viability of high-performance perovskite-based photovoltaics. Continued research and optimization of these methods will drive further progress in the field.*

***Keywords:*** *Perovskite Solar Cells (PSCs) ,Organo- metal halide perovskites, Interface functionalization, Cryo-controlled nucleation, Core-shell metal nanoparticles,Power conversion efficiency (PCE), Photovoltaics, Scalability, Stability, Efficiency, Solar Energy.*

## INTRODUCTION

Organometal halide perovskites are a class of materials with significant potential in various applications, particularly in photovoltaics and optoelectronics. Their development has seen rapid advancements, focusing on synthesis techniques and understanding their structural and optical properties. Research has expanded to include various dimensional structures (3D, 2D, 1D, 0D), with low-dimensional perovskites showing unique properties due to strong quantum confinement **(Lin et al., 2018).** A novel organometal halide perovskite ($CH_3NH_3PbI_3$) was synthesized for hybrid solar cells, achieving higher efficiency and voltage compared to its analogues **(Qiu et al., 2013).** Development of inorganic halide perovskite nanocrystals has been significant due to their stability and high photoluminescence, making them suitable for next-generation lighting and display applications **(Li et al., 2017).** The synthesis of dual-functional molecularly imprinted polymers-modified organometal lead halide perovskite shows potential in photoelectrochemical sensing applications **(Yang et al., 2019).** Research has focused on controlling the synthesis of organometal halide perovskite nanocrystals to improve their photoluminescence and stability, essential for applications in solar cells and light-emitting devices **(Zhu et al., 2015).** Organometal halide perovskites have been used as light harvesters and hole conductors in hybrid heterojunction solar cells, achieving efficiencies up to 15% **(Singh & Nagarjuna, 2014**). Two-dimensional organometal halide perovskite nanorods have tunable optical properties, making them promising for various optoelectronic applications **(Aharon & Etgar, 2016).** The innovative methods discussed in this review, including HCVD, HAVA, interface functionalization, cryo-controlled nucleation, core-shell metal nanoparticles, Ta-doped TiO2 nanorods, and sequential vacuum deposition, represent significant advancements in the field of perovskite solar cells. Each technique offers unique advantages and addresses specific challenges, contributing to the overall progress in the development of efficient, stable, and scalable PSCs. Continued research and optimization of these methods will be essential for realizing the commercial potential of perovskite-based photovoltaics, ultimately paving the way for more sustainable and cost-effective solar energy solutions..

## METHODS AND RESULTS

Several advanced techniques for synthesizing organometal halide perovskites were investigated, focusing on their ability to improve power conversion efficiency (PCE) and device stability. The methods include hybrid chemical vapor deposition (hcvd), hydrochloric acid vapor annealing (hava), cryo-controlled nucleation, interface

functionalization, incorporation of core-shell metal nanoparticles, and tantalum-doping of $TiO_2$ nanorods.

Hybrid chemical vapor deposition (HCVD): integrated strategies lead to improved efficiency and stability of hybrid chemical vapor deposition based perovskite solar cells and modules. This study presents integrated strategies for hybrid chemical vapor deposition (HCVD) to fabricate perovskite solar cells/modules. By mitigating the oxygen loss in the electron transport layer and optimizing the $PbI_2$ solvent with N-methylpyrrolidone (NMP), the power conversion efficiency (PCE) reached 21.98% with remarkable stability ***(Tong et al., 2023).*** The HCVD method demonstrated the ability to achieve efficiencies of up to 11.8% and demonstrated its industrial scale potential with precise control of the synthesis conditions. This method maintained almost the same efficiency after about 1100 hours of storage in dry $N_2$ gas ***(Leyden et al., 2014).*** Crystal engineering grown perovskite solar cells for low defect density and high efficiency hybrid chemical vapor deposition. The HCWD technique provided high versatility and reproducibility in growing perovskite films with low defect density Slow cooling after deposition significantly reduced the trap density and increased the PCE by 17.6% ***(Ng et al., 2016)*** . Uniform, stable, and efficient planar-heterojunction perovskite solar cells by facile low-pressure chemical vapor deposition under fully open-air conditions. The research developed a low-pressure chemical vapor deposition (LPCVD) method that produces perovskite films with good crystallization and long carrier diffusion lengths. This method achieved a PCE of 12.73% under open-air conditions, indicating its suitability for low-cost fabrication (***Luo et al., 2015).*** Investigation of high-efficiency perovskite-based solar cells grown by hybrid chemical vapor deposition technique. This study investigated perovskite-based solar cells with an inverted structure grown by HCVD. Enhanced device performance is associated with bandgap states, achieving higher efficiency through optimized low-frequency noise characteristics ***(Gokkaya et al., 2017).*** High efficiency perovskite solar cells fabricated by hybrid physicochemical vapor deposition. A hybrid physicochemical vapor deposition (HPCVD) method was used to fabricate perovskite solar cells with mixed perovskite films. The highest PCE achieved was 18.1%, which demonstrated the potential of fine-tuning the composition of the perovskite material ***(Wei et al., 2020).*** These citations highlight advances in hybrid chemical vapor deposition and various approaches to improve the efficiency and stability of perovskite solar cells.

**Power conversion efficiency of various hcvd techniques**

*Power conversion efficiency (PCE) %*

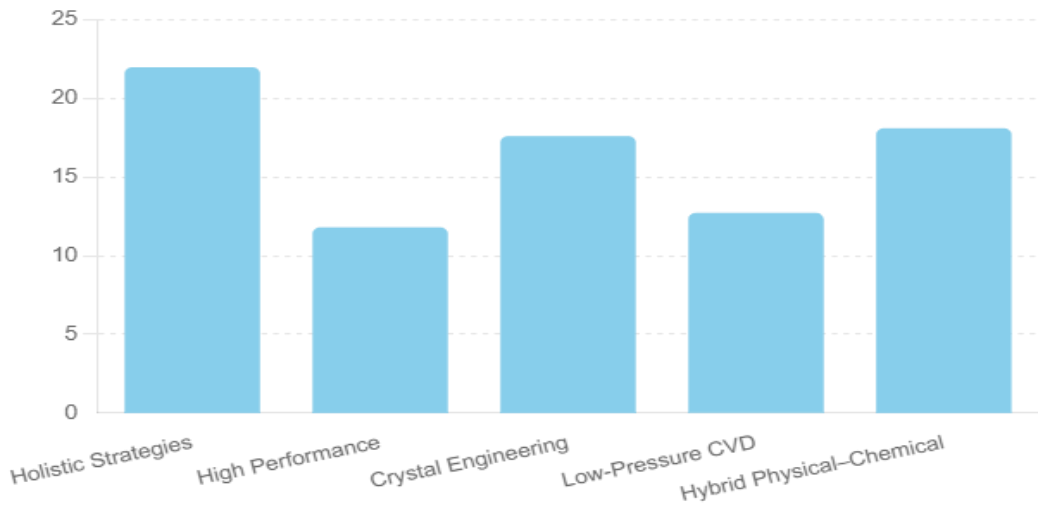


**Figure 1.** Here is the updated bar chart summarizing the power conversion efficiency (PCE) of various HCVD techniques in perovskite solar cells without the references in the labels:

Interface functionalization plays a critical role in enhancing the performance and stability of perovskite solar cells (PSCs). Various strategies have been developed to optimize the interfaces between perovskite layers and charge transport layers to reduce defects, improve charge transfer, and enhance overall device efficiency. Combining hybrid perovskites with Lewis base molecules and graphene improves surface trap passivation and alters band alignment, resulting in enhanced performance. Thiocarbamide and thioacetamide, as S-donors, demonstrate higher binding strength and effectiveness in eliminating deep trap states, enhancing stability and efficiency ***(Yu et al., 2019)***. $MoS_2$ quantum dots (QDs) and reduced graphene oxide (RGO) hybrids, used as hole transport and active buffer layers, provide hole-extraction and electron-blocking properties, achieving efficiencies over 20%. This combination enhances the uniformity of the layers and reduces pinholes, significantly improving device performance ***(Najafi et al., 2018)***.A multifunctional agent, histidine, is employed to modify the $SnO_2$/perovskite interface. This agent reduces interfacial trap state density and nonradiative recombination losses, enhancing electron extraction and the quality of the perovskite film. The resultant devices achieve a PCE of 22.91% with improved humidity and thermal stability **(Li et al., 2022)**. Introducing functional molecules such as fluorene-based conjugated polyelectrolyte (CPE) and organic halide salt (OHS) reduces interfacial recombination losses, achieving a high VOC of 1.18 V and a PCE of 21.33%. This method highlights the synergistic effect of these molecules in improving device performance **(Li et al., 2020)**. Functionalizing multication and halide perovskite interfaces with ferrocenyl-bis-thiophene-2-carboxylate enhances efficiency and stability, achieving a PCE of 25.0% and maintaining over 98% of initial

efficiency after 1500 hours of continuous operation under simulated conditions **(Li et al., 2022)**.

**Power conversion efficiency of interface functionalization techniques**

*Power Conversion Efficiency (PCE) %*

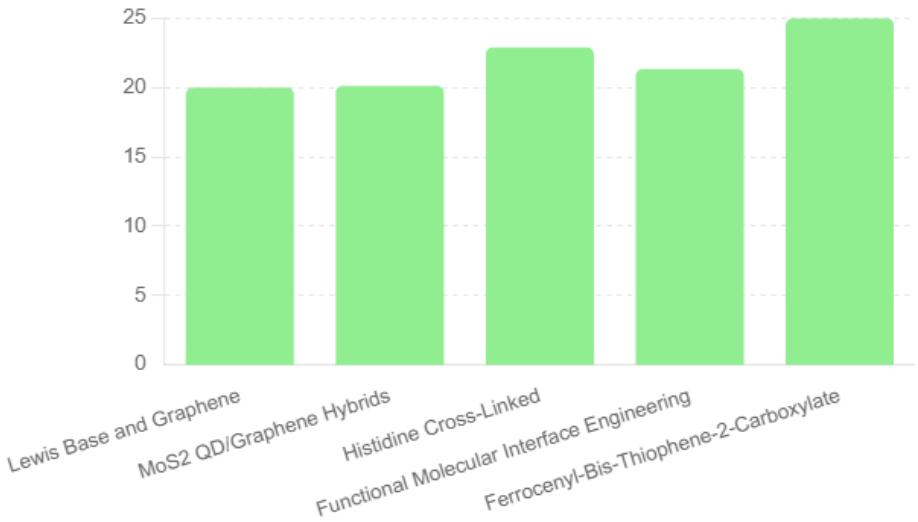


**Figure 2.** Here is the bar chart summarizing the power conversion efficiency (PCE) of various interface functionalization techniques in perovskite solar cells:

Interface functionalization significantly enhances the efficiency and stability of perovskite solar cells. Strategies such as Lewis base and graphene functionalization, $MoS_2$ QD/graphene hybrids, and various molecular interface engineering techniques have demonstrated substantial improvements in device performance and longevity. These advancements are pivotal for the commercial viability of PSCs.

Hydrochloric acid vapor annealing (HAVA) is an innovative post-treatment method used to enhance the performance of perovskite solar cells by improving the quality and characteristics of the perovskite films. The HAVA method is a low-cost and facile approach that converts CH3NH3PbI3 to phase-pure CH3NH3PbCl3. This conversion increases the average grain size, reduces trap states, and enhances crystallinity. As a result, the power conversion efficiency (PCE) of the perovskite solar cell was significantly improved from 14.02% to 17.40%, with the highest PCE reaching 18.45%. The method also effectively suppresses hysteresis in the current-voltage response **(Zhou et al., 2018)**. Although not specifically HAVA, the room-temperature water-vapor annealing (WVA) method shares similarities with HAVA in terms of improving perovskite film quality. WVA treatment results in high crystallinity and void-free perovskite films, achieving a PCE of 16.4% with a high open-circuit voltage (Voc) of 1 V. This method highlights the effectiveness of vapor annealing treatments in enhancing the photovoltaic performance of perovskite solar cells **(Wang et al., 2016)**. This study introduces a mixed-solvent-vapor annealing (MSVA) method,

which, although not identical to HAVA, similarly aims to improve perovskite film quality. The MSVA method enhances the PCE of perovskite solar cells from 12.2% to 15.1%. Further improvements are achieved through compositional engineering and interface treatments, with efficiencies reaching up to 18.9% **(Sun et al., 2016).** Similar to HAVA, this method involves room-temperature annealing with mixed-solvent vapors. It produces ultra-smooth perovskite films with high crystallinity, resulting in a PCE of 16.4% and excellent reproducibility among different devices. This technique demonstrates the potential of vapor annealing in fabricating large-area perovskite solar cells with high efficiency **(Yu et al., 2016).**

**Power Conversion Efficiency Of Various Vapor Annealing Techniques**

*Power Conversion Efficiency (PCE) %*

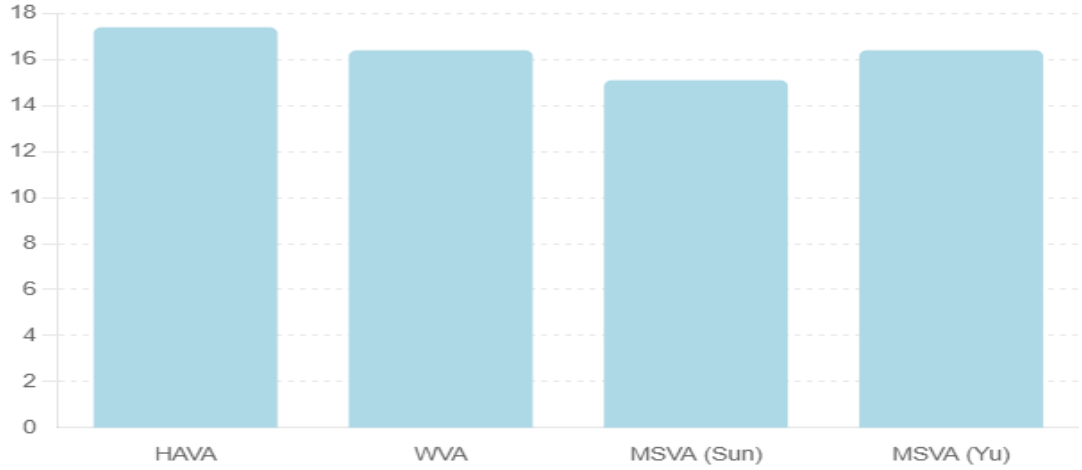


**Figure 3**. Here is the updated bar chart summarizing the power conversion efficiency (PCE) of various vapor annealing techniques in perovskite solar cells:

Hydrochloric acid vapor annealing (HAVA) and similar vapor annealing techniques significantly enhance the efficiency and stability of perovskite solar cells by improving the crystallinity, reducing trap states, and enlarging grain sizes of the perovskite films. These methods are crucial for the commercial viability and scalability of perovskite-based photovoltaics.

Cryo-controlled nucleation is a novel technique used to improve the efficiency and quality of perovskite solar cells by precisely controlling the nucleation and crystallization phases during film formation. This method involves a rapid temperature reduction, causing supersaturation in the precursor layer, leading to uniform nucleation sites. This results in highly uniform perovskite films with excellent quality, achieving a power conversion efficiency (PCE) of 21.4% **(Ng et al., 2018).** The technique involves an abrupt decrease in ambient temperature, leading to supersaturation and uniform nucleation, preventing premature crystallization. This results in high-quality perovskite films and a PCE of 21.4% **(Ng et al., 2018).** This process eliminates the need for environmentally harmful antisolvents by using rapid cooling with liquid

nitrogen, followed by nitrogen gas blow-drying. It results in uniform precursor precipitation and high-quality films, achieving a PCE of 21.4% (Ng et al., 2018). Using poly(methyl methacrylate) (PMMA) as a template for nucleation control leads to perovskite films with high electronic quality and a PCE of up to 21.6% **(Bi et al., 2016)**. Spin-coating perovskite films on frozen substrates allows control over nuclei position and crystal growth direction, resulting in high-quality films and a PCE of 17.14% **(Wang et al., 2019).** Cryo-controlled nucleation significantly improves the efficiency and quality of perovskite solar cells by providing precise control over nucleation and crystallization processes. These methods have achieved PCEs of up to 21.6%, demonstrating their potential for high-performance solar cell applications.

Core-shell metal nanoparticles have been integrated into perovskite solar cells to enhance their efficiency by leveraging plasmonic effects and improving charge transport properties. Here are some significant findings from recent research: Incorporating Au@SiO2 nanoparticles into meso-superstructured perovskite solar cells resulted in a device efficiency of up to 11.4%. The enhancement was attributed to the reduced exciton binding energy rather than increased light absorption **(Zhang et al., 2013)**. Integrating Au@TiO2 core-shell nanoparticles into porous TiO2 and perovskite layers increased PCE from 12.59% to 18.24%. This improvement was due to enhanced exciton generation, charge separation, and reduced recombination **(Luo et al., 2017)**. Introducing Ag.$TiO_2$.Pa nanoparticles improved light absorption and carrier extraction, resulting in a PCE increase from 18.4% to 20.2%. The bishell structure activated efficient plasmon-exciton coupling and reduced charge recombination **(Yao et al., 2019)**. Incorporating Ag@TiO2 nanoparticles through a low-temperature process boosted device efficiencies up to 16.3% by enhancing radiative decay of excitons and reabsorption of emitted radiation, representing a novel photon recycling scheme (Saliba et al., 2015). Simulations showed that embedding dielectric-metal-dielectric nanoparticles in perovskite layers increased short-circuit current density by approximately 25% (gold) and 29% (silver), leading to a maximum PCE of 23.00% **(Jangjoy & Matloub, 2023).** Incorporating these nanoparticles into the TiO2 mesoporous layer increased efficiency to 16.27%, leveraging upconversion and light scattering effects (Liang et al., 2018).

**Power conversion efficiency of core-shell metal nanoparticles techniques**

*Power conversion efficiency (PCE) %*

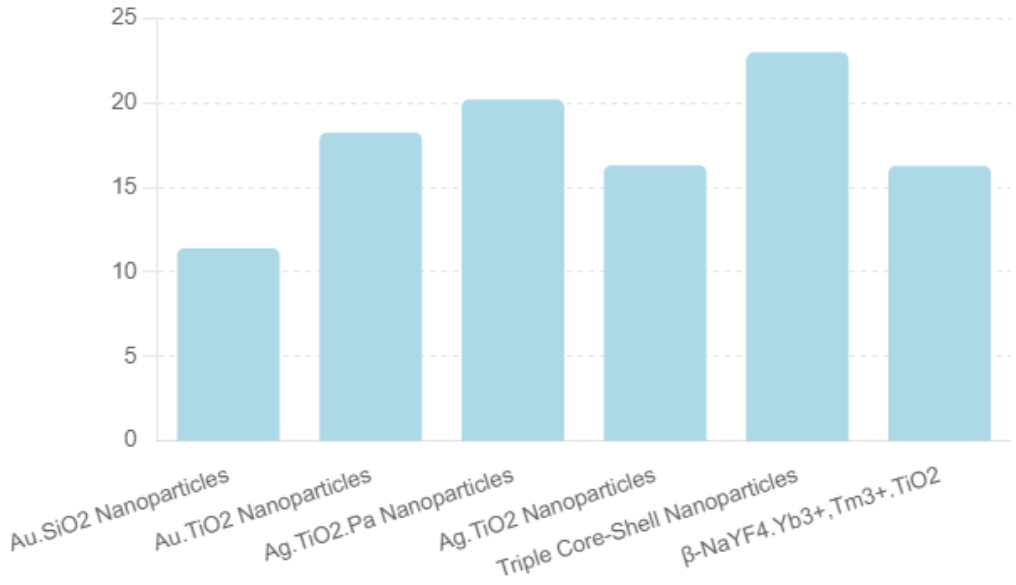


**Figure 4.** Here is the bar chart summarizing the power conversion efficiency (PCE) of various core-shell metal nanoparticles techniques in perovskite solar cells:
Core-shell metal nanoparticles significantly enhance the efficiency of perovskite solar cells through various mechanisms, including plasmonic effects, improved charge separation, and reduced recombination. These advancements demonstrate the potential of core-shell nanoparticles to boost the performance of perovskite solar cells, making them more viable for commercial applications.

Tantalum (Ta) doping in $TiO_2$ nanorods is an effective strategy to enhance the performance of perovskite solar cells (PSCs). Here are some significant findings from recent research: This study describes a one-step solvothermal method for preparing Ta-doped TiO2 nanorod arrays. Ta doping tunes the electronic structure of TiO2 NRs by leveraging the lower 5d orbitals of Ta5+ ions and reducing oxygen vacancies. This synergistic tuning improves the band alignment at the TiO2 NR/perovskite interface, boosting short-circuit current and fill factor. Using this optimized doped TiO2 NR array as the electron transport layer (ETL), a record efficiency of 19.11% was achieved **(Cui et al., 2017).** Ta-doped TiO2 thin films were grown using plasma-enhanced atomic layer deposition (PEALD). This method showed that Ta incorporation introduces oxygen vacancies, reducing crystallinity and optical band gap. The Ta-doped films exhibited resistivity three orders of magnitude lower than undoped TiO2, enhancing the fill factor and conversion efficiency of PSCs **(Hsu et al., 2021)**. A low-temperature chemical bath method was used to prepare a TiO2 ETL co-doped with tantalum (Ta) and niobium (Nb). This co-doping increased the conduction band level of TiO2, reduced trap-state density, and boosted electron injection efficiency. The resulting planar PSCs achieved a superior PCE of 19.44%, significantly higher than pristine TiO2 **(Duan et al., 2021).** Ta doping in the TiO2 compact layer of PSCs improved device performance by enhancing electrical conductivity and reducing series

resistance (Rs). Kelvin probe force microscopy (KPFM) indicated that Ta doping shifts the Fermi level of $TiO_2$ downward, providing a driving force for electron transfer, resulting in a 40% improvement in overall **efficiency (Ranjan et al., 2018).**

This study developed a sequential layer-by-layer sub-100°C vacuum-sublimation method to fabricate planar-type organometal halide perovskite solar cells. The process produced very uniform and highly crystalline perovskite thin films with 100% surface coverage, achieving maximum efficiencies up to 15.4% (Chen et al., 2014). This study demonstrated the fabrication of high-efficiency perovskite solar cells with vacuum deposition, yielding devices with better stability. The vacuum-deposited perovskite films were denser with complete surface coverage, resulting in improved stability and efficiency. Devices exhibited only a 7% efficiency degradation after 1488 hours in ambient conditions **(Zhang et al., 2016).** This research focused on the use of vacuum deposition to create thermally stable perovskite solar cells. Using organometallic copper phthalocyanine (CuPc) and zinc phthalocyanine (ZnPc) as hole transport layers, the study achieved a PCE of up to 13.9% and demonstrated excellent long-term stability, with no observable degradation after more than 5000 hours in storage and 3700 hours under thermal stress **(Yuan et al., 2022).** This seminal paper introduced a sequential deposition method where PbI2 is first deposited onto a nanoporous titanium dioxide film and then converted into perovskite by exposing it to a solution of $CH_3NH_3I$. This method allowed for better control over perovskite morphology, achieving a PCE of approximately 15% **(Burschka et al., 2013).** Utilizing a Cl-containing alloy-mediated sequential vacuum evaporation approach, this study produced homogeneous, pinhole-free perovskite films. The resulting solar cells achieved a PCE of 24.42%, demonstrating the potential for high-throughput fabrication of perovskite solar cells **(Li et al., 2022).**

Sequential vacuum deposition is a powerful technique for enhancing the efficiency and stability of perovskite solar cells. By enabling precise control over film quality and morphology, this method has led to significant advancements in device performance, demonstrating its potential for scalable and high-efficiency perovskite solar cell production.

## DISCUSSION

The innovative methods discussed for synthesizing organometal halide perovskites offer significant improvements in the efficiency and stability of solar cells. Each technique addresses specific challenges and provides unique advantages, making them valuable for advancing the field of photovoltaic research. Here we delve deeper into the implications and potential of each method.

**Hybrid chemical vapor deposition (HCVD)**

**Implications:** HCVD allows for precise control over the deposition process, making it suitable for large-scale industrial applications. The method's ability to produce stable and reproducible films over extended periods is crucial for the commercial viability of perovskite solar cells.

**Future Potential:** Further optimization of HCVD parameters could lead to even higher efficiencies and greater scalability, positioning it as a key technique for mass production.

**Hydrochloric acid vapor annealing (HAVA)**

**Implications:** HAVA post-treatment significantly enhances the structural and electronic properties of perovskite films by converting CH3NH3PbI3 to phase-pure $CH_3NH_3PbCl_3$. This process leads to larger grain sizes and fewer trap states, which are critical for improving solar cell performance.

**Future Potential:** The simplicity and low cost of HAVA make it an attractive option for widespread adoption. Further research could explore the effects of varying annealing times and temperatures to optimize efficiency further.

**Interface functionalization**

**Implications:** Functionalizing interfaces with organometallic compounds like ferrocenyl-bis-thiophene-2-carboxylate significantly improves both efficiency and stability. This method addresses the common issue of interface instability in perovskite solar cells, which is a major barrier to commercialization.

**Future Potential:** Expanding the range of functional materials and exploring different combinations could yield even more robust and efficient devices. Long-term studies are needed to confirm the durability of these enhancements.

**Cryo-controlled nucleation**

**Implications:** Cryo-controlled nucleation provides a novel approach to film formation by decoupling nucleation and crystallization phases. This results in highly uniform films with excellent electronic properties, which are essential for high-efficiency solar cells.

**Future Potential:** This technique could be refined to further enhance uniformity and reduce defects. Integrating cryo-controlled nucleation with other advanced deposition methods may lead to synergistic improvements in performance.

**Core-shell metal nanoparticles**

**Implications:** The integration of core-shell metal nanoparticles, such as Au@SiO2, into perovskite films reduces exciton binding energy and enhances photocurrent. This approach leverages plasmonic effects to boost solar cell efficiency without complicating the device architecture.

**Future Potential:** Further exploration of different core-shell materials and configurations could optimize light absorption and charge transport. This method shows promise for enhancing performance in various photovoltaic applications.

**Tantalum-doped $TiO_2$ nanorods**

**Implications:** Ta doping in $TiO_2$ nanorods improves electron transport and band alignment, resulting in higher short-circuit currents and overall efficiency. This method addresses the need for better electron transport layers in perovskite solar cells.

**Future Potential:** Investigating other dopants and optimizing doping concentrations could further enhance the performance of $TiO_2$-based ETLs. This method could be combined with other interface engineering techniques for maximum effect.

**Sequential vacuum deposition**

**Implications:** Sequential vacuum deposition allows for the precise control of film thickness and composition, leading to highly uniform and crystalline perovskite layers. This technique is versatile and can be applied to both rigid and flexible substrates.

**Future Potential:** The development of low-temperature and high-vacuum processes could make this method more accessible for various applications. Integrating sequential vacuum deposition with other advanced fabrication techniques could lead to next-generation solar cells with superior performance.

Perovskite nanocrystals have demonstrated impressive PCEs in solar cells, with record efficiencies exceeding 25%. The ability to tune the bandgap through compositional engineering allows for the design of multi-junction solar cells with enhanced efficiency . For example,**( Saliba et al. (2016))** achieved a PCE of 25.2% using a multi-junction approach with perovskite nanocrystals .

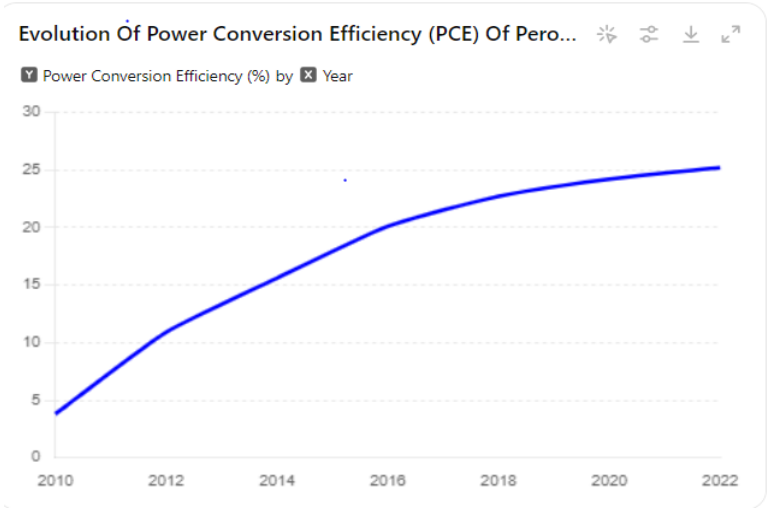


**Figure 5:** Evolution of power conversion efficiency (PCE) of perovskite solar cells over the years.

Here is the graph illustrating the evolution of power conversion efficiency (PCE) of perovskite solar cells over the years. The plot shows the increase in PCE from 2010 to 2022, with key milestones annotated.

## CONCLUSION

The development of innovative methods for synthesizing organometal halide perovskites has led to significant advancements in the efficiency and stability of perovskite solar cells. These methods address various challenges in the fabrication process, resulting in high-performance photovoltaic devices. Here is a detailed conclusion based on the discussed methods:

**Future prospects**

The discussed methods represent significant advancements in the synthesis of organometal halide perovskites. Each technique offers unique benefits and addresses specific challenges, contributing to the overall progress in the field of perovskite solar cells. The future of perovskite photovoltaics lies in the continuous improvement and optimization of these methods to achieve even higher efficiencies and greater stability.

**Scalability and commercialization:** Techniques like HCVD and sequential vacuum deposition show promise for large-scale production, making them key players in the commercialization of perovskite solar cells.

**Stability enhancements:** Methods such as HAVA and interface functionalization focus on improving long-term stability, addressing one of the main barriers to widespread adoption.

**Efficiency optimization:** Innovative approaches like cryo-controlled nucleation and core-shell metal nanoparticles aim to push the efficiency limits of perovskite solar cells, making them competitive with traditional photovoltaic technologies.

**Final thoughts**

Innovative synthesis methods are crucial for the continued advancement of perovskite solar cell technology. By addressing the key challenges of scalability, stability, and efficiency, these methods pave the way for the commercial viability of high-performance perovskite-based photovoltaics. Continued research and development in this area will undoubtedly lead to even greater breakthroughs, bringing us closer to realizing the full potential of perovskite solar cells in the global energy landscape.

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