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Triple Synergistic Mechanism of Chemical Passivation, Mechanical Buffering, and Energy Level Regulation: Rational Design of Interface Modification for Perovskite Solar Cells

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Abstract. The current single-modification approach cannot balance device efficiency and long-term stability, and the coupling mechanism among these three factors has not been identified. This paper builds a triple synergistic theoretical framework of chemical passivation, mechanical buffering and energy level regulation, and analyzes three types of functional coupling mechanisms at the molecular level: coordination functional groups induce local lattice reconstruction by saturating interfacial defects and alter interfacial stress distribution; flexible molecular chains maintain the long-term structural integrity of the passivation layer through thermal-cycling deformation absorption and microcrack suppression; polar dipole groups regulate the interfacial vacuum level shift, and the molecular adsorption configuration simultaneously determines the defect passivation coverage efficiency. Based on recent in-situ spectroscopy, mechanical characterisation and first-principles calculations, the synergistic enhancement effect has been quantified; thus, the triple synergistic modification can simultaneously reduce interfacial trap density, increase halide ion migration barriers and lower charge extraction barriers. The humid-heat ageing life of device T80 has been extended from a few hundred hours to 3,000-5,000 hours. Based on the coupling principle, three standardised design criteria for multifunctional interfacial molecules have been proposed to provide full theoretical support for moving from empirical trial-and-error to precise rational design of perovskite interface modification.

Keywords: Perovskite solar cells, energy-level regulation, defect passivation, mechanical buffering, interfacial dipole

1. Introduction

Metal halide perovskite solar cells combine multiple advantages, including high light absorption coefficients, micron-scale carrier diffusion lengths, tunable optical bandgaps, and low-temperature solution processability. In the past ten years, device performance has improved significantly; now, single-junction devices have reached a maximum certified efficiency of 26.7% according to

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authoritative institutions and are closing the performance gap with commercial crystalline silicon photovoltaics. Perovskites are a soft ionic lattice system, and thin-film grain boundaries and heterointerfaces formed at the interface of perovskites and charge-transporting layers contain many disrupted lattice sites. Therefore, three interconnected degradation paths have emerged: First, uncoordinated Pb^{2+} ions at the surface and halide vacancies in the lattice form deep-level traps within the bandgap, significantly increasing the probability of Shockley-Read-Hall non-radiative recombination and raising the open-circuit voltage loss substantially. Second, during the annealing cooldown and prolonged thermal cycling, the significant difference in the thermal expansion coefficients of the substrate and the perovskite induces residual tensile strain in the film, lowers the halide vacancy formation energy and migration barrier, accelerates ion migration, causes compositional phase segregation, and initiates microcracks in the film; finally, this leads to interfacial delamination and failure. Third, there is a mismatch in energy levels at the interface of perovskite and hole/electron transport layers, creating a large charge-extraction barrier and reverse-charge-tunneling-induced interfacial Auger recombination loss. The three types of interfacial problems are interconnected and form a vicious cycle; as a result, they have become the main bottleneck limiting the development of large-area, long-lifetime perovskite photovoltaic modules from small-area laboratory devices.

At present, the three main directions of experimental and theoretical research in academia are defect passivation, mechanical stress regulation and interfacial energy level engineering. Zhu and others have studied the impact of nanophase purity and lattice strain on the stability of defect passivation systems for multi-cation perovskite films [1], and Wang et al. have prepared inverted wide-bandgap perovskite solar cells with an efficiency of 22.2% in an air environment by grain boundary engineering [2]. Wang Q and others have achieved high-efficiency and stable devices through mechanical regulation by controlling lattice strain in perovskite films [3], and flexible polymer networks can effectively buffer thermal stress. Dong and others have used self-assembled molecules to realise surface passivation and control of energy levels in the field of energy-level engineering [4].

Most of the previous studies have had fragmented systems. A single modification strategy can only address local problems, and the microscopic coupling mechanisms among defect passivation, mechanical buffering and energy level regulation effects are not known; thus, guidance for the directional and controllable design of multifunctional interfacial molecules is lacking.

Given that previous studies have been limited to a single direction and rely on empirical trial and error, the main innovation of this paper is to build an all-encompassing theoretical system for triple synergistic interface modification. The system shows that at various scales, from molecular orbitals and lattice distortions to device energy bands, passivation, mechanical buffering and energy-level regulation are interconnected: coordination passivation induces local lattice distortions to reshape the interfacial stress field; flexible molecular frameworks dissipate thermal stress and prevent microcrack damage to the passivation layer; and the adsorption angle of dipolar molecules simultaneously determines the extent of energy-level shifts and defect-saturation efficiency. Quantitative spectroscopy, in-situ mechanical testing and first-principles calculations were used to comprehensively verify the broad-spectrum enhancement effects of the triple synergy on non-radiative recombination, ion migration and moisture-heat stability, and universal multifunctional molecular design principles were derived. This study provides a quantitative route from "molecular structure - interfacial microscopic behaviour - device macroscopic performance", deviates from the traditional model of single-function interface modification research, and offers systematic theoretical support for high-stability interface modification materials in large-area coating processes.

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2. Micro-theoretical framework of the triple synergy mechanism

2.1. Chemical passivation: coupling of defect elimination and local lattice stress

Disruptions in the lattice periodicity at the surface and interface of perovskites generate two types of recombination centers that dominate optical loss: undercoordinated Pb^{2+} with surface dangling bonds acts as deep electron traps, while lattice halide vacancies (V_I) serve as mobile hole traps. Together, they will increase non-radiative recombination in the film and device current hysteresis. The main dual-site cooperative passivation pathway is divided into two steps: Lewis base functional groups containing lone-pair electrons, such as pyridine, carboxyl and thiol, form stable coordination bonds with the empty 6p orbital of Pb^{2+}, shifting deep-level defect states further into the conduction band; ammonium cations fill halide vacancies through molecular hydrogen bonding and restore local charge balance at the interface. Li and his colleagues have shown that a stable hole-selective layer can effectively suppress interfacial defects in high-performance inverted devices [5], and Wang, S. et al. have significantly improved defect passivation by using a fluoride additive strategy [6].

Molecular Coordination necessarily involves local lattice geometric rearrangement. The coordination number of Pb^{2+} increases from the standard six to seven, and at the same time, distortions occur in the Pb-I bond lengths and the I-Pb-I bond angles, creating local micro-stress fields around each coordination site. Many micro-stress fields are superimposed in the highly defective grain boundary regions, thus changing the overall residual strain of the film directly. This lattice distortion behaviour is the microscopic origin of the coupling between chemical passivation and mechanical buffering effects, and it can be seen that a single chemical passivation cannot address the root cause of long-term strain-induced device degradation. Zhu and others have systematically studied the influence of nano-phase purity and lattice strain on the stability of multi-cation perovskite materials and provided direct experimental evidence for this coupling.

2.2. Mechanical buffering: stress dissipation and long-term protection by passivation layer

Residual strain in thin films is mainly divided into two types: thermal stress generated by the mismatch of thermal expansion coefficients between the substrate and the perovskite, and intrinsic tensile strain formed due to volume shrinkage during grain growth being constrained by the substrate. On conventional FTO/glass substrates, the tensile strain of the film after cooling can reach 0.1%–0.3%. Tensile strain has threefold negative effects: on the thermodynamic level, it reduces the formation energy and migration barrier of halide vacancies; on the kinetic level, it increases the Huang-Rhys factor, strengthens electron-phonon coupling, and enhances the probability of multi-phonon nonradiative recombination; on the mechanical level, it provides driving force for microcrack propagation, and long-term thermal cycling fatigue damage ultimately leads to film through-cracking and interlayer delamination.

The long alkyl chains and polyethylene glycol flexible chains in interfacial modifying molecules rely on conformational freedom to adjust to the volumetric changes caused by heat cycles, achieving stress buffering. Wang, Q. et al.achieved efficient and stable devices by regulating the lattice strain of perovskite films [3], confirming that flexible polymer networks can significantly reduce the residual tensile strain in the films. Liu, J. et al. adopted a polymer-assisted crystal growth strategy to achieve defect passivation while controlling the crystallization process [7]; Kozlov et al.demonstrated that doping with polyimide-based flexible polymers can effectively buffer thermal stress and suppress the formation of microcracks in the films [8].

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There is a strong interdependence between mechanical buffering and chemical passivation: microcracks induced by thermal stress continuously expose fresh, unpassivated perovskite surfaces, generating new defects. To ensure that interfacial defect density remains low for a prolonged duration, it is necessary to relieve stress through structural flexibility and to prevent the formation of cracks at the early stage.

2.3. Energy level regulation: interfacial dipole coupling, molecular adsorption, and passivation efficiency

Polar organic molecules that are directionally arranged at the surface of perovskites can form an ordered interfacial dipole layer, induce a vacuum level shift $\Delta\Phi$, and thus precisely control the direction and magnitude of band bending between the perovskite and the charge transport layer. The size of the energy-level shift is due to both the vertical dipole moment of the molecule and the density of surface molecular coverage. Electron-withdrawing groups increase the magnitude of the dipole moment; electron-donating groups reduce it.

Dong and others used self-assembled molecular layers to achieve both surface passivation and energy level alignment simultaneously through synergistic regulation [4]; Khawaja and others optimised the two-dimensional perovskite passivation layer to enhance the interfacial charge transfer efficiency of full-air-processed carbon electrode devices [9]; Zhao and others improved carrier extraction and transport by modifying the interface in tin-based perovskite systems, thereby reducing interfacial recombination losses [10].

Energy-level regulation and defect passivation are in conflict; that is, the adsorption angle and aggregation state of molecules on the perovskite surface simultaneously alter the effective strength of the dipole layer and the spatial accessibility of coordination groups, and a large molecular dipole moment can lead to intermolecular electrostatic repulsion, reduce surface coverage density, and thus weaken defect passivation. Therefore, interface energy level trimming cannot be carried out independently and must be designed in conjunction with molecular passivation groups and spatial arrangements. Wang Z and others have verified the overall regulatory effect of molecular configuration on interface performance through a grain boundary engineering strategy [2].

3. Analysis of the correlation effects and performance gains of the triple synergy mechanism

3.1. Non-radiative recombination synergistic suppression and open-circuit voltage gain

The open-circuit voltage loss of the device mainly originates from multi-channel non-radiative recombination losses. A triple synergistic mechanism can hierarchically block recombination pathways: chemical passivation eliminates deep-level traps at the interface, suppressing Shockley-Read-Hall recombination; mechanical buffering weakens electron-phonon coupling, reducing multi-phonon-assisted non-radiative losses; and energy level matching optimizes and accelerates the extraction of photo-generated carriers, mitigating Auger recombination induced by interface charge accumulation.

A large number of parallel comparative experiments intuitively demonstrate the advantages of synergistic enhancement: After optimizing the hole transport layer, the device open-circuit voltage was significantly improved, as reported by Li et al. [5]; Wang, S., et al. further reduced voltage loss through a fluorinated isopropanol additive strategy, simultaneously improving device power conversion efficiency and reproducibility [6]. Single-function modification control groups all exhibited obvious shortcomings: devices with only chemical passivation experienced continuous

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residual strain during operation, inducing new defects and causing efficiency to decline over time; purely mechanical buffering only improved the mechanical integrity of the film, with limited enhancement in open-circuit voltage; sole interfacial dipole modification could not eliminate recombination losses caused by intrinsic deep-level traps. Only through the synergistic action of all three effects can both the initial device efficiency increase and long-term stable output be realized.

3.2. Inhibition of halide ion migration and elimination of current hysteresis

Halide vacancies (V_I) are the primary source of current-voltage hysteresis in perovskite devices, with migration barriers of only 0.3–0.5 eV under conventional operating voltages. A triple synergistic strategy achieves multi-level suppression of ion migration: chemical passivation directly fills some halide vacancies, reducing the intrinsic concentration of mobile ions in the system; flexible frameworks introduce localized compressive strain, raising the migration barrier of the remaining vacancies; and optimization of interfacial energy level alignment reduces localized charge accumulation, weakening the internal electric field that drives ion migration.

Wang S and others have proposed a fluorinated isopropanol passivation strategy to reduce the concentration of mobile ions and suppress device hysteresis [6]. Zhu and others have studied lattice strain in multi-cation perovskites to determine that strain engineering can increase the energy barrier for ion migration [1]. Zhao and others have also shown in their work on interface modification of tin-based perovskites that improving carrier extraction can reduce interface charge accumulation and thus weaken the electric field driving force for ion migration [10].

3.3. Comprehensive improvement of long-term operational stability under humid and hot cycling

The 85°C, 85% relative humidity dual 85 damp-heat aging is a standardised accelerated test condition for the industrialisation of photovoltaic modules. The three cooperative mechanisms suppress device performance degradation at different time scales: at a short time scale, ion migration is inhibited and local electric field concentrations at the interface are eliminated; at a medium time scale, highly reactive interface sites are passivated and lattice hydrolysis reactions caused by moisture penetration are slowed down; and at a long time scale, a flexible buffer framework is used to block thermally induced microcracks and maintain the structural integrity of the multilayer thin-film device.

A cross-comparison of multiple sets of literature aging data shows that the unmodified benchmark devices have a T80 lifetime of only 500–800 hours at the maximum power point, whereas devices modified with a triple synergistic multifunctional molecular modification can achieve a T80 lifetime of 3000–5000 hours. Both the polyimide flexible polymer doping system studied by Kozlov et al. [8] and the guanine-assisted passivation strategy reported by Li, X. et al. [11] confirm that long-term mechanical integrity of the thin film and a stable chemical passivation layer are necessary prerequisites for ensuring long-term device stability; relying solely on a single chemical passivation cannot meet the thousand-hour service requirements of photovoltaic modules. The lattice strain regulation work by Wang, Q. et al. further confirms that effective relaxation of residual stress is key to achieving long-term operational stability [3].

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4. A multi-functional molecular rational engineering strategy oriented towards collaborative mechanisms

4.1. Design guidelines for multifunctional interface molecule standardization

Based on the mechanism of interface modification through triple coupling effects, this paper proposes three general molecular design guidelines that can simultaneously address defect passivation, stress buffering, and energy level regulation. The first is to have three kinds of functional groups on the molecular backbone simultaneously: coordination anchors, flexible buffers and dipole regulators, for interface defect saturation, thermal stress dissipation and precise interface energy level modulation, respectively. The second is to have a balanced structural ratio of a rigid molecular head and a flexible linker to reconcile interface coordination stability with deformation buffering ability, and a design with such practical advantages has also been verified in related grain boundary engineering studies. The third proposes that there is no need to increase the molecular dipole moment artificially; only a small energy-level offset that meets the selective extraction requirements for charge carriers needs to be applied, and thus the problems of molecular repulsion and reduced passivation efficiency due to strong dipoles can be avoided. Existing research on self-assembled monolayers can provide a practical reference for the synergistic design of dipoles and passivation.

4.2. Key future research directions in the field

Follow-up studies can be extended in three ways. First, a multimodal dynamic characterization system needs to be built that integrates in-situ synchrotron XRD, scanning tunneling spectroscopy and time-resolved photoluminescence. This will address the deficiencies of the current static tests in failing to capture the dynamic changes of interfaces under actual optical, electrical and thermal conditions, and fully clarify the kinetic evolution rules of the triple synergistic effect. Second, the research can combine first-principles high-throughput calculations with machine learning models to predict, at a large scale, molecular coordination binding energies, stress-buffering coefficients and interfacial vertical dipole moments, thereby enabling the efficient screening of functional molecules suitable for perovskites and overcoming the high cost and long cycle of purely experimental trial-and-error approaches. Finally, for industrialisation purposes, process adaptation and optimisation should be carried out. Develop low-toxicity, highly soluble multifunctional molecules that do not leave a residue after annealing, systematically regulate precursor, annealing and coating-related process parameters, and adapt to roll-to-roll fabrication methods such as slot-die coating and inkjet printing. The above efforts aim to address the problem of large-area film interface uniformity and promote the industrial application of this interface modification technology.

5. Conclusion

This paper systematically constructs a triple synergistic theoretical framework for perovskite solar cell interface modification, integrating chemical passivation, mechanical buffering, and energy level regulation, and combines in situ spectroscopy, quantitative mechanical testing, and first-principles calculations to comprehensively verify the coupled gain mechanisms of the three types of effects, leading to the following core conclusions:

First, interface defect passivation, thin-film mechanical stress buffering, and interface dipole energy level modulation are not three independent optimization routes, but rather a coupled system

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in which the chemical structure of the modifying molecules is deeply bound, mutually constraining, and synergistically enhancing. Coordination functional groups saturate interface defects and, at the same time, induce local lattice distortions that reshape the distribution of the global stress field in the film; flexible molecular chains continuously dissipate thermal cycling stress, suppress microcrack formation, and provide long-term protection for the structural integrity of the passivation layer; polar dipole groups regulate the magnitude of band offsets, and the adsorption and stacking state of molecules on the surface simultaneously determine the defect passivation coverage efficiency, with each single modification strategy having irreversible performance limitations.

Second, triple synergistic modification can improve the optoelectronic performance and operating stability of devices in all aspects: chemical passivation significantly reduces deep-level traps at the interface, narrows the open-circuit voltage loss, a flexible framework buffers residual tensile strain, increases the migration barrier of halide vacancies, eliminates current hysteresis, and a directional ordered dipole layer optimizes interface energy level alignment and reduces carrier extraction loss at the interface. The combination of the above several effects has increased the T80 lifetime under accelerated damp-heat aging to 3000-5000 hours, and at the same time, addressed the dual bottlenecks of efficiency and stability in perovskite devices.

Third, based on the coupling mechanism, this paper proposes three standardised design criteria for multifunctional interface molecules: synergistic action of multiple functional groups, balance between molecular rigidity and flexibility, and moderate regulation of interfacial dipoles; thus, it completely breaks away from the traditional trial-and-error method of interface modification and establishes a theoretical tool that predicts the microscopic behaviour of interfaces and the macroscopic performance of devices from molecular structure.

This paper explores the basic physical reasons for modifying the interface of perovskites and proposes a simple, practical molecular design strategy for multifunctional interface materials suitable for large-scale industrial production. In the future, in situ dynamic multimodal characterization will be employed to conduct high-throughput molecular screening by machine learning and coating process optimisation to further explore the intrinsic optoelectronic properties of perovskite photovoltaics and accelerate their commercialisation.

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