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Perspectives on the mechanical robustness of flexible perovskite solar cells

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Flexible perovskite solar cells (FPSCs) show promise for future applications in wearable electronic devices, biosoft robots, and advanced sensors due to their high power-conversion efficiency, low manufacturing cost, and high flexibility. The inevitable performance degradation during long-term service is the major challenge for the commercialization of perovskite solar cells. Mechanical robustness, which is the stability of PSCs under large deformation and high-number cyclic loading, is of particular concern for FPSCs. Numerous strategies have been proposed to improve the mechanical robustness by enhancing the inherent toughness of the major components, relieving stress by optimizing the structure of the device, and developing high-quality film fabrication methods. FPSCs are not as mechanically robust as other types of flexible cells, and the coupled mechanical–optoelectronic degradation mechanism for FPSCs and the associated experimental and theoretical methods require further investigation. This perspective highlights recent advances in improving the mechanical robustness of FPSCs with respect to internal and external structural optimization and the influence of mechanical deformation on photovoltaic behavior. Current challenges and future perspectives for clarifying the qualitative mechanism between the mechanical behavior and the photovoltaic performance as well as establishing quantitative descriptions of the mechanical robustness are elaborated, aiming to provide more scientific and practical guidance for the design of FPSCs with high efficiency and robustness.

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1. Introduction

Perovskite solar cells (PSCs) are a promising photovoltaic technology due to their high power conversion efficiency (PCE) and low manufacturing cost as well as their great potential for use in flexible applications.¹ Over the past decade, researchers have achieved a substantial leap in PCE for these cells (from 3.8% to 25.7%).² Flexible perovskite solar cells (FPSCs) deposited on polymer substrates are ideal for wearable electronic applications, as they are lightweight and bendable.^{3–5} However, FPSCs have a PCE up to 22.44% lower than that for rigid PSCs.⁶ Performance degradation will inevitably occur as damage accumulates in the functional layers under various service conditions (a recently reported state-of-the-art residual performance of 81% of the initial efficiency after 20 000 bending cycles⁷). As this loss of performance seriously limits the further commercialization of FPSCs, improving the mechanical robustness of FPSCs is a major goal of the research community.

The mechanical robustness of FPSCs relies mainly on the balance between the flexibility and functionality of the functional layers. As an example, for FPSCs with chromium oxide–metal contacts, the robustness depends on the balance between the increased resistance of the indium tin oxide (ITO) electrodes and the newly formed defects in perovskite crystals under large deformation.⁵ Numerous research efforts have been conducted to investigate the inherent mechanism for the mechanical–optoelectronic degradation and to explore better strategies for simultaneously enhancing the flexibility of the FPSCs and the device performance.

When considering flexible substrates, it is known that polymer-based substrates such as polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) cannot tolerate high temperatures. Thus, producing high-quality electron transport layers (ETLs) at low temperatures is a straightforward strategy for relieving interlayer force. However, the intralayer thermal stress resulting from the large difference in thermal expansion between the flexible substrates and the functional layers may induce defects in the functional layers, resulting in stress concentrations during service and leading to more rapid degradation. To date, many studies on low-temperature fabrication methods have been explored^{8,9} including low-temperature hydrothermal processes,¹⁰ ultraviolet ozone treatment,¹¹ and N₂ plasma treatment strategies.¹² Li *et al.*¹³ recently reported

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that FPSCs subjected to methylamine gas treatment exhibit outstanding flexibility and retain 94.5% of the initial PCE after 3000 bending cycles.

As the ITO electrodes used in transparent electrodes are intrinsically brittle, cracks can form and propagate under relatively small stretching or bending deformation.¹⁴ The rapid increase in the density of these cracks during service is one of the main factors contributing to the poor mechanical robustness of FPSCs.¹⁵ To overcome this issue, numerous flexible transparent electrodes have been investigated to find alternatives that have excellent electrical conductivity and outstanding mechanical properties.^{16–19}

The perovskite layer is another major concern for the mechanical stability of FPSCs. The poor crystalline quality of perovskite results in a large number of grain boundaries, and microcracks can easily form at these boundaries under stress. The microcracks in the perovskite functional layers will reduce the PCE due to nonradiative carrier recombination and device shunting.²⁰ To achieve high-quality perovskite films, strategies such as additive engineering and grain boundary passivation have been proposed to improve both the efficiency and flexibility of the films.^{21–23}

The state-of-the-art in terms of PCE and the qualitative measurement of mechanical robustness (the number of bending cycles/bending radius) for rigid PSCs, flexible PSCs, and other commercially available flexible solar cells are summarized and compared in Fig. 1. While the cyclic bending test is a widely used method for evaluating the mechanical robustness of flexible devices, it does not directly represent the internal stress state of the thin film structures of a solar cell, as the stresses are

dependent on the thickness of the substrate and the bending radius. Because of the thickness of the thin film structure and the lack of information regarding the moduli of the materials, the bending stresses for thin-film flexible devices are rarely reported. Thus, we introduce a ratio (the number of bending cycles over the bending radius) as a qualitative signifier of the mechanical robustness of FPSCs. As shown in this figure, a PCE up to 25.7% has been achieved for rigid PSCs. Achieving a simultaneous improvement in the PCE and the mechanical robustness is extremely challenging. For example, the PCE for an FPSC can be increased to 20.75% by using a ZnSnO₂/SnO₂ electronic transport layer, but the mechanical robustness of the FPSC would be quite limited. The mechanical robustness of FPSCs can be significantly improved by using metal meshes as substitutes for ITO (with nearly no degradation after 1000 bending cycles at a 0.5 mm bending radius); however, the resulting FPSCs have a relatively low PCE (13.62%). Bionic structures have opened up a new avenue to simultaneously improve the efficiency and mechanical robustness for FPSCs as compared to commercially available flexible solar cells (FSCs) such as flexible amorphous silicon solar cells, copper indium gallium selenide cells, and organic solar cells.

Overall, to achieve superior flexibility while maintaining the outstanding optoelectronic properties of FPSCs, new manufacturing and design strategies are needed. Moreover, the underlying coupled mechanical–optoelectronic mechanisms of FPSCs and their impact on PCE degradation have not been thoroughly understood and are worthy of further investigation.

In this perspective, the research progress regarding various internal and external structure designs that have been modified

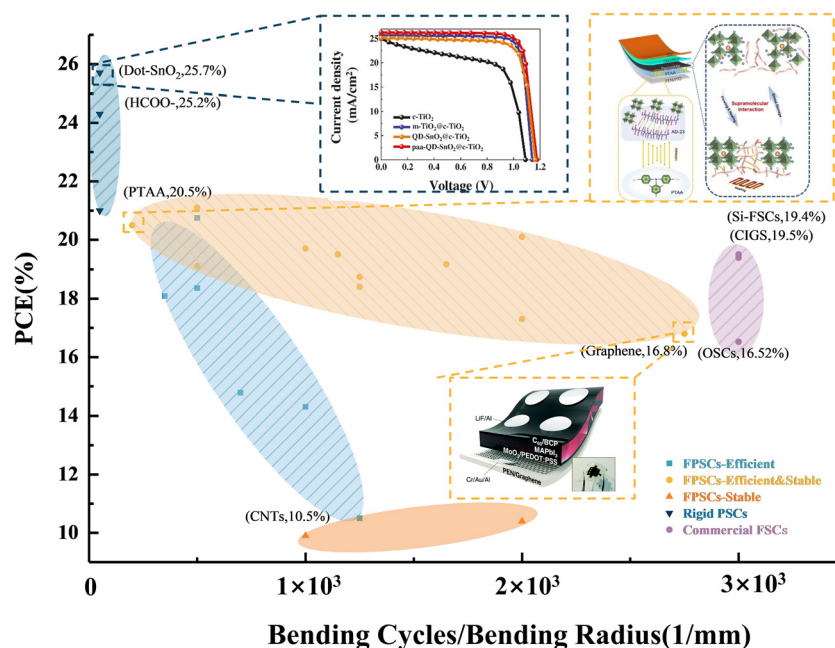


Fig. 1 The power conversion efficiency (PCE) and qualitative measurement of mechanical robustness (the number of bending cycle/bending radius) of rigid PSCs, flexible PSCs and other commercial flexible solar cells (rigid PSCs with high optical performance;^{24–26} blue: FPSCs focus on efficiency enhancing;^{8,9,19,27–29} orange: FPSCs focus on mechanical stability enhancing;^{16,17,30} yellow: FPSCs with both high efficiency and mechanical robustness;^{18,21–23,31–36} purple: commercial flexible solar cells).



to improve the mechanical robustness of FPSCs^{37,38} is summarized and discussed. In addition, the recent progress in electro-mechanical coupled studies on perovskites is reviewed and discussed as well. Areas of future research on the mechanical robustness and degradation mechanisms of FPSCs are proposed to improve their performance and enable wide commercial application.

2. Advances in enhancing the mechanical robustness of FPSCs

The poor mechanical robustness of FPSCs is mainly attributed to the reduction in the electrical properties of transparent electrodes and to delamination between the carrier transport layers and the perovskite layers. Structural optimization methods have been regarded as one of the most promising methods to achieve acceptable photovoltaic performance and mechanical robustness for FPSCs. A variety of structural design options and convenient production processes have resulted in outstanding enhancement in photovoltaic performance and mechanical robustness. In the following subsections, recent advances in improving the mechanical robustness of FPSCs through internal and external structure designs and optimization are presented, along with a discussion of the theoretical analysis and experimental verification.

2.1. Internal structure optimization through the introduction of buffer layers

Due to the typical multilayer structure of a FPSC, the effects of different functional layer materials and mechanical properties on the photovoltaic performance of the FPSC are extremely complicated. The introduction of buffer layers (BLs) with a suitable band structure and outstanding mechanical properties

is an optimal strategy for facilitating carrier transport and releasing mechanical stress.

Researchers have reported that an energy level barrier exists between the perovskite light absorbing/transport layer and the transport layer/electrode (Fig. 2(a)). This barrier, which is particularly evident under mechanical stress, hinders the further improvement in FPSC performance due to the effect of strains on the calculated band structure and the estimated band gap (V_g).³⁷ Introducing BLs with an appropriate band structure will alleviate band mismatch and carrier recombination at the interface under tensile strain. Recent studies have shown that Cu_2O , $\text{Cu}_2\text{O}/\text{Cu}$ ³⁹ and PbS ,⁴⁰ which have higher carrier mobility, can be used between the hole transport layer (HTL) and the electrodes to promote effective transport of the holes. Al-doped ZnO ⁴¹ and polyethyleneimine (PEI) polymer⁴² can act as BLs between the ETL and the cathode. These two materials can accelerate the transport of electrons because the conduction band energy levels of the Al-doped ZnO and the PEI are between those of the electron transport layer and the cathode work function, and this can aid in optimising the energy level gradient. Moreover, PEI is known to have superior ductility, which can facilitate an improvement in mechanical robustness. The interlayer force between PEI and ITO, by acting as a polymer buffer layer, can ease the stress concentration and reduce cracking in a brittle ITO electrode that results from stretching and bending and, thus, can enhance the overall stability of the FPSC. In addition, CsBr ⁴³ and graphene oxide (GO),⁴⁴ by acting as BLs between the carrier transport layer and the perovskite layer, can reduce the non-radiative recombination at the interface, preventing moisture and oxygen from penetrating the light absorbing layer of the perovskite and further improving the stability of the FPSC.

Apart from focusing on the function of BLs in terms of band matching, interface defect passivating, and charge carrier transfer, researchers are encouraged to explore in future studies the optimal thickness, high light transmittance, stable chemical

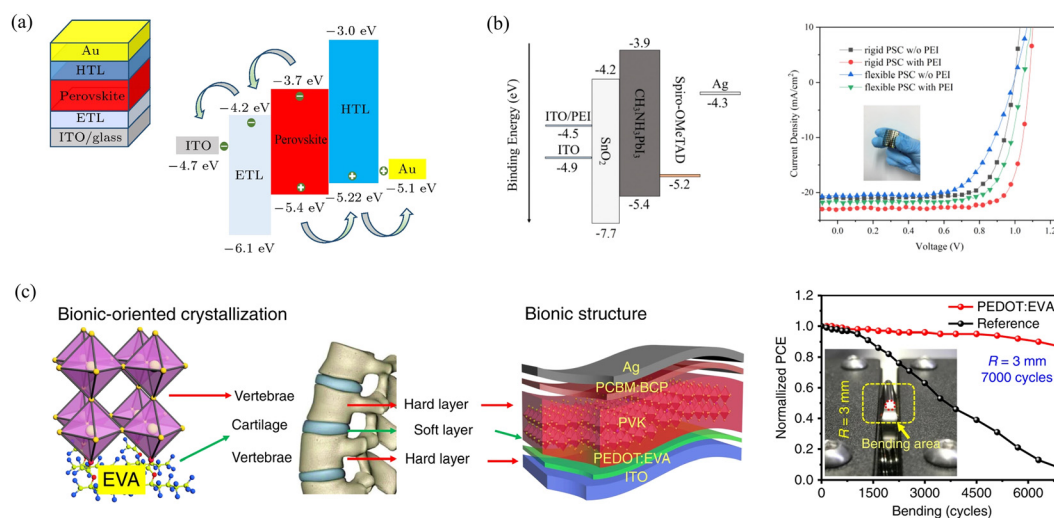
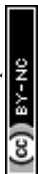


Fig. 2 (a) Structure and energy band diagram of perovskite solar cell;⁴⁴ (b) the energy levels of the materials used in the devices and the J - V curves of the champion devices with and without PEI;⁴³ (c) biomimetic mechanisms of the vertebrae and PSCs and normalized averaged PCE value for the flexible PSCs under bending cycles with radius of 3 mm.⁴⁶



properties, and excellent photoelectric properties of BLs in order to enhance the opto-electronic effect more comprehensively.

It has been generally accepted that the introduction of BLs can not only improve the initial PCE by creating an energy level gradient but can also improve the mechanical robustness of the FPSC through the release of stress because of the relatively high ductility of the BLs. From the perspective of the mechanical mechanism, BLs such as PEDOT:EVA,⁴⁵ s-GO,⁴⁶ and PEDOT:GO have been introduced to optimize the internal structure of the FPSC and release the interlayer and intralayer forces. As shown in Fig. 2(c), PEDOT:EVA was inspired by the way that the bones and soft tissues of the vertebrae work together to execute complex movements. The results of experiments and theoretical simulations demonstrate that this bionic interface layer acts as an adhesive and is able to retain more than 85% of the original PCE after 7000 bending cycles. In addition, based on the toughness and hydrophobicity of GO, sulfonated graphene oxide (s-GO) has been employed to effectively passivate the defects of vacant iodine by interacting with $[\text{PbI}_6]^{4-}$. These cementitious grain-boundary passivation FPSCs retain over 80% of the original PCE after 10 000 bending cycles.

The fracture resistance of perovskite solar cells also has unique implications for mechanical robustness. Watson *et al.*⁴⁷ focused on enhancing the fracture resistance (G_c) of perovskite solar cells by promoting the adhesion of organic materials to inorganic oxide surfaces. They developed a cross-linkable solvent-resistant fullerene adduct, MPMIC₆₀, that exhibits suitable electronic properties and can increase the fracture resistance of perovskite solar cells. PSCs with conventional geometry that utilize cured films of MPMIC₆₀ were reported to exhibit a significant improvement in fracture resistance (205%) over that of a C₆₀ control.

However, to sum up the above studies, we found that the above analysis method is relatively invariable. Based on the theoretical support of the intrinsic properties of semiconductor and mechanical properties, the photoelectric performance and mechanical robustness of FPSCs with or without BLs are normally compared using experiments. Morphological characterization and finite element simulation can also be used to help explain the function of BLs. Furthermore, interlaminar damage or layer separation also has a great influence on the performance of FPSCs, as they are associated with the adhesion properties of the functional layers. Ichwani *et al.*⁴⁸ used force microscopy to quantify the adhesive interactions between the functional layers of FPSCs. They found that the interfacial adhesive forces are correlated to FPSC performance and to the charge carrier transfer resistance between the perovskite layer and the charge carrier transport layers. Further study of the adhesive forces requires an in-depth investigation of the damage and failure modes of the FPSCs during their service as well as a detailed study of the mechanism of delamination failure.⁴⁹

Nonetheless, the quantitative relationship between mechanical stress and photovoltaic performance is still unclear. An innovative analysis tool is needed to further study the relationship between the released stress and the improvement in performance that result from the introduction of BLs, as this

relationship would inform the selection and introduction of BLs when designing FPSCs.

2.2. External structure optimisation to achieve stretchable, bendable, and foldable structure designs

With the rapid introduction of self-powered flexible devices and wearable electronics, the need for stretchable, bendable and foldable FPSCs has surged. However, the transparent electrodes (ITO, *etc.*), electron transport layers (TiO₂, *etc.*) and perovskite light-absorbing layers in these devices are intrinsically brittle and mechanically fragile, which can cause a sharp increase in resistance and a reduction in the optoelectronic performance of the devices.

External structure design has emerged as a highly efficient strategy for the design of flexible solar cells (*e.g.*, for island structures,⁵⁰ serpentine structures⁵¹ and kirigami-origami structures⁵²). Recent studies have shown that a fibre shape can be used for the design of a FPSC structure. In a study by Deng *et al.*,⁵³ a nanostructured fibre and a spring-like modified Ti wire were used as the two electrodes, and other functional layers were coated on a Ti wire. The elastic PSC fibre exhibited a stable photovoltaic performance under both stretching and bending (the PCE was maintained at 90% after 250 stretching cycles at a strain of 30%). Moreover, an elastic fibre-shaped PSC can be woven into various textiles that are used for fabricating self-powered wearable devices. However, the process for preparing the elastic fibre-shaped PSC is complex, and the resulting textile has a limited light absorbing area and a low PCE (5.22%). More research is needed before industrial production and other applications can be realized.

Other researchers have developed FPSCs that were inspired by a prestretching structure. Kaltenbrunner *et al.* fabricated ultrathin and buckling FPSCs with a total thickness of 3 μm ; these FPSCs maintained their initial PCE after 25 compression cycles and exhibited only a 30% reduction after 300 stretching cycles.⁵⁴ Nevertheless, the intrinsic fragility presented a limited initial PCE and stretching strain. To address this bottleneck, Dauskardt *et al.* introduced a new concept in perovskite solar cell design: perovskite compound solar cells with close-packed hexagonal or honeycomb structures. This design addressed the intrinsic fragility of the functional layers through the incorporation of internal scaffolds that provide mechanical reinforcement.⁵⁵ Li *et al.* proposed a structure design for FPSCs that was inspired by kirigami, an ancient paper cutting art.⁵⁶ The kirigami-based PSCs were reported to maintain their performance after 1000 cycles of stretching, bending, and twisting. Fig. 3(c) shows the finite element analysis results for a kirigami device with optimal geometric parameters under 150% stretching strain. As the stress is concentrated near the cut ends of the functional layers can be placed on areas with lower stress to achieve high stretchability and mechanical stability.

Encapsulation of perovskite solar cells is also an effective way to improve their mechanical robustness. The encapsulated FPSCs of the organosilicate barrier layer, in which the barrier precursors were sprayed at the leading edge of a compressed air plasma, was reported to pass 1000 bending cycles without



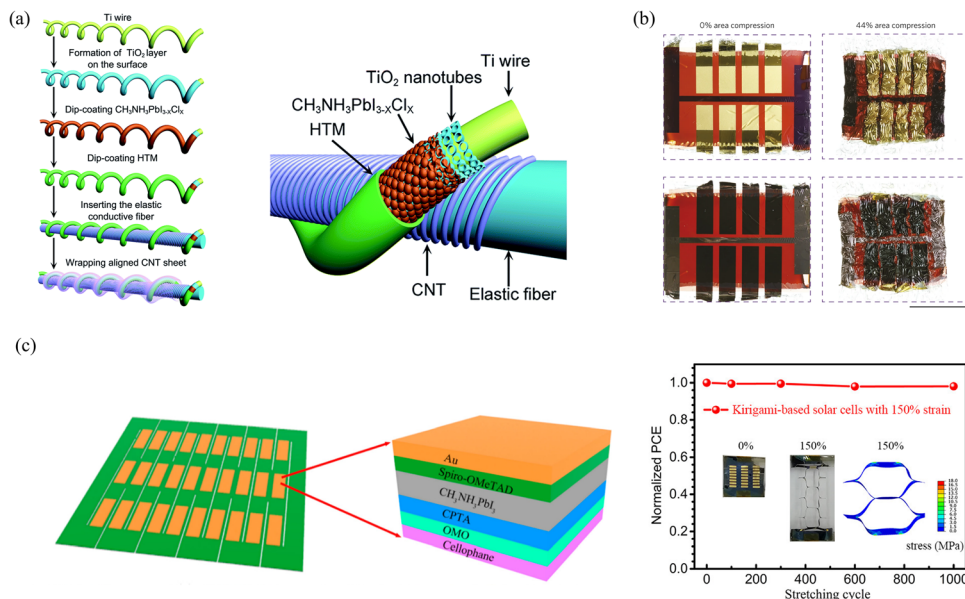


Fig. 3 (a) Schematic illustration of the elastic PSC fibre;⁵³ (b) schematic of the ultrathin and buckling FPSCs;⁵⁴ (c) schematic illustration of the kirigami-based PSC array and structure of PSCs and normalized PCE of kirigami-based PSCs after different stretching cycles.⁵⁵

visible cracks.⁵⁷ However, most experimental studies focus on the effect of encapsulation on the long-term stability, temperature resistance, and humidity resistance of the PSC.^{58–60} Nevertheless, it is expected that the encapsulation layer can release the interlayer stress and delay the propagation of cracks. A more detailed understanding of the effect of encapsulation will require further experimental and theoretical investigation.

Overall, it is obvious that the enhanced stretchability and flexibility of FPSCs result from the focus on the patterned structure as a means for optimising the stress distribution, which can significantly reduce the large strain and stress concentration in the PSC area. Still, it is not clear how the strain and stress distribution can affect the degradation of FPSCs. In other words, in the mechanical failure models of FPSCs, the trend in the deterioration of photovoltaic performance as well as the critical stress and strain needed to cause failure are still unclear. Therefore, there is an urgent need to qualitatively explore the relationship between the mechanical behaviour and photovoltaic performance in order to design an optimal structure and enable a more accurate prediction of the PCE.

2.3. Progress in the study of electromechanical multifield coupled behaviour for PSCs

To date, a number of studies have been conducted to investigate electromechanical multifield coupling in perovskite solar cells. These studies have concentrated on the impact of strain on the electronic band structure and the optoelectronic properties of perovskites.

There is an optimal range in the band gap for measuring the photoelectric performance of solar cells. Zhu *et al.*⁶¹ calculated the band structure of FAPbI₃ through first principles, demonstrating

that the band gap increases under tensile strain; this increase can impact the optical properties to some extent.

The optoelectronic properties of perovskites, such as the carrier dynamics, can also change under tension or compression strain. Chen *et al.* reported that hole mobility increased at a small range of compressive strain and that perovskite crystals under -1.2% strain had the highest hole mobility.⁶² Pei *et al.* showed that FPSCs exhibit better mechanical stability with concave bending than with convex bending.⁶³ In particular, the series resistance (R_s) was found to increase sharply after convex bending. Pei *et al.* also explored in-depth the effect of stress on the optoelectronic properties of FPSCs.⁶⁴ It was reported that a reduction in the band gap and an increase in the lifetime of the carrier can be achieved by a slight stretching of the perovskite functional layer.

The impact of strain on FPSC performance degradation can also be attributed to strain-induced decomposition of MAPbI₃. Zhao's group examined XRD patterns and showed that tension strain accelerates the degradation of perovskite films under illumination.⁶⁵ Rolston *et al.* also carried out experiments to explore the different decomposition rates of perovskite under tension and compression.⁶⁶ As shown in Fig. 4(c), a perovskite layer under compressive strain exhibited a slower decomposition rate from MAPbI₄ to PbI₂, which was attributed to the higher formation energies of defects and the higher activation energies required for ion migration.⁶⁷

Moreover, the influence of ferroelectric polarization on photovoltaic conversion is worth considering. Zhao's group demonstrated that spontaneous polarization caused distortion of the lattice structure and that photoinduced surface charges and potentials were shifted;⁶⁸ these findings prove that interactions occur among the photoinduced charges, polarization, and ions in perovskite.⁶⁹ Based on the results of these studies,



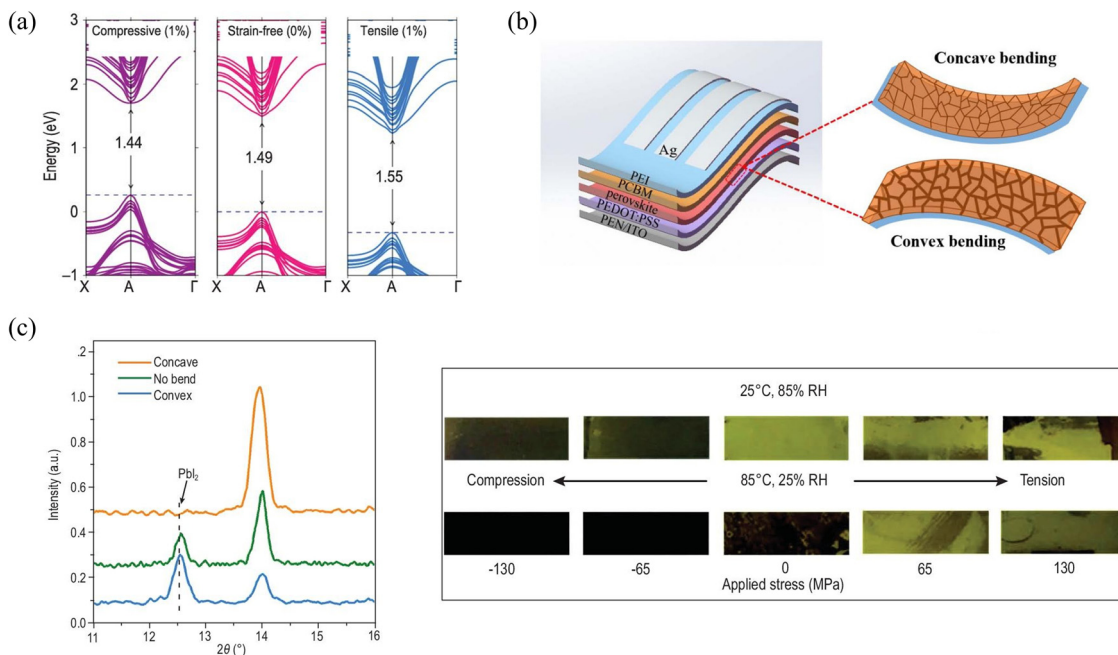


Fig. 4 (a) Calculated band structures under biaxial tensile, zero, and compressive strains from first-principles density functional theory (DFT)-based approaches;⁵⁶ (b) hole mobilities by Hall effect measurements showing that α -FAPbI₃ with a strain of -1.2% has the highest hole mobility;⁶¹ (c) out-of-plane XRD of the concave, no bend and convex films.⁶⁴ Photographs of MAPbI₃ on PET revealing compressive stress-enhanced film stability and tensile stress reduced film stability.⁶⁵

we can reasonably speculate that ferroelectric polarization might influence the photovoltaic conversion, offering strong evidence that reveals the mechanical–electric–optical multifield coupling mechanisms of PSCs.

In general, the previous studies of the electromechanical multifield coupling mechanisms for PSCs were mainly focused on the perovskite light absorbing layer, which was explored at a

microscopic scale. To clarify the relationship between the local structure and global performance, scanning probe microscopy (SPM) techniques were used to detect partial morphological changes as well as to characterize the mechanical, electrical, and chemical performance of the PSC;⁷⁰ these techniques included conductive atomic force microscopy and kelvin probe force microscopy. Zhao's group also proposed some future

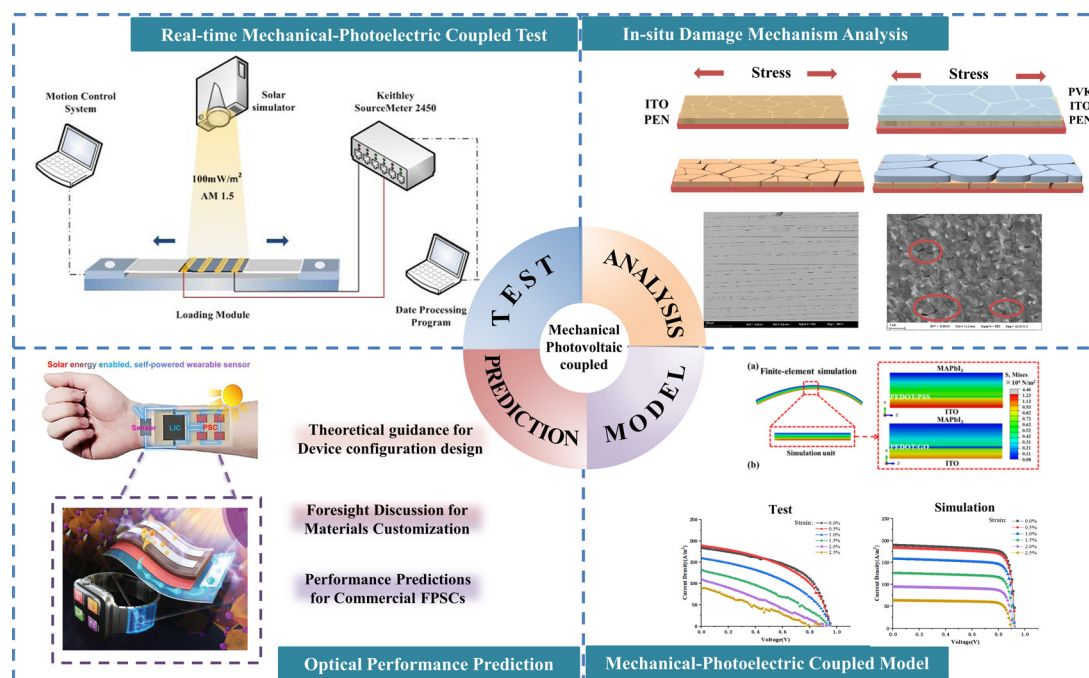


Fig. 5 Some possible directions and perspectives for further studies on the mechanical–photovoltaic coupling mechanism.



directions for using SPM techniques to study perovskite microstructures and multifield responses.⁶⁹

As no critical link between the defects in different functional layers and global performance has yet been established, we propose to quantify the impact of each functional layer by conducting experiments and simulations in addition to studying the electromechanical coupling behaviour of the perovskite layers. Based on preliminary experimental results, we can conclude that ITO electrodes are fractured and delaminated under stretching and bending stress, causing a significant degradation of electrical conductivity.⁷¹ Defects can also be generated easily at the interfaces of functional layers and grain boundaries, contributing to nonradiative recombination and reducing the carrier lifetime.⁷² Because of this, a clear description of coupled electrical and mechanical property measurements is urgently needed. Determining which factor plays a major role in the global performance degradation is undeniably a promising direction for future research on high-efficiency FPSCs.

3. Conclusions

Although much progress has been made in the structural optimisation of PSCs, the research prospects are still wide and broad. Here, we propose some possible directions for further studies.

First, designing a real-time monitoring test to study mechanical–photovoltaic coupling is the first step in revealing the qualitative mechanism between the mechanical behaviour and the photoelectric performance of a FPSC. Current tests for mechanical stability are conducted in a two-step fashion: cycles of stretching or bending testing followed by comparison of the PCEs before and after the test. However, the behaviour of efficiency loss caused by deformation cannot be determined. Real-time tests to monitor mechanical–photovoltaic coupling will ease these gaps;⁷³ however, considerable viability challenges remain, including how to ensure the stability of the test connections and how to collect data during service. Rational design of the FPSC geometry and the use of silver wire and conductive silver glue to reinforce the electrode connections may contribute to harvesting stable data during the process. Moreover, it was reported that the temperature of an FPSC can reach up to 65 °C during its service, and the annealing temperature can reach up to 100 °C during the preparation process. The service temperature will affect the photoelectric and mechanical properties and will generate additional stresses due to the mismatch in the thermal expansion coefficients for different functional layers, subsequently causing a reduction in mechanical robustness. Therefore, investigation of the temperature effect and the development of a suitable *in situ* thermal-mechanical testing platform are necessary to further reveal the mechanism for the drop in mechanical robustness.

Based on the device performance degradation law, the damage and synergy of each functional layer in a FPSC deserve more in-depth study. Here, we present *in situ* tests of ITO, polyvinylcarbazole (PVK), HTL, and ETL using individually

coated specimens based on flexible substrates to obtain valid information on the critical strain for initial crack generation as well as the crack saturation density. Crack density, which can be considered as a coupling parameter between the conductivity and strain, offers a reliable analysis method to determine how damage to each functional layer will affect the overall efficiency of the FPSC. In addition, SPM and synchrotron radiation X-ray tomography techniques also show great potential for dynamically characterizing the evolution of the internal microstructure of FPSCs in real time.

After a basic determination of the coupling parameters has been made, we propose to establish a refined high-precision coupling model for FPSCs. Mechanical–photoelectric coupling experiments, combined with multiphysical field coupling simulations, would be the most effective method for clarifying the qualitative mechanism between the mechanical behaviour and the photoelectric performance of FPSCs. Comsol Multiphysics, with its user-friendly multiphysics coupling solution, is a valid tool for analysing the underlying mechanisms.^{74,75} Recently, we implemented the coupling of a semiconductor module with a solid mechanics module through parameter calls. The *J–V* curve initially obtained and the evolution of the photoelectric properties with increasing strain are shown in Fig. 5. We rationally foresee that effective validation and reasonable prediction of the coupling model through parametric optimisation will be an important direction for future research.

With the rapid growth in self-powered wearable electronic devices, FPSCs are demonstrated to be the most commercially exploitable power source. To accommodate the unprecedented demand for devices with excellent deformability, we need to clarify the qualitative mechanism between the mechanical behaviour and the photoelectric performance, establish quantitative descriptions of the mechanical robustness of the devices, and reimagine some additional structural engineering strategies in the near future.

Author contributions

Meihe Zhang: conceptualisation, methodology, investigation, formal analysis, writing – original draft, visualisation. Zhihao Li: conceptualisation, methodology, writing – original draft. Zheng Gong: writing – original draft, visualisation. Zhen Li: conceptualisation, writing – review and editing. Chao Zhang: conceptualisation, writing – review and editing, supervision, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

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