

Next-Generation Flexible Photovoltaics Interfacial Engineering, Stability Enhancements, And Advanced Nanocomposites In All-Perovskite Solar Cells

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ABSTRACT

Innovations in buried interfaces low temperature processing and solvent orthogonal recombination layers have brought flexible all-perovskite photovoltaics from proof-of-concept devices to mechanically robust modules with increasingly plausible efficiency-stability balance. Device records generally measure only one type of stress, such as tensile stress or only bending in radius, or only measure stress on a rigid substrate without accounting for polymer-substrate constraints. This review critically synthesizes the literature exploring key questions: What is the relationship between interfacial selectivity and adhesion? What is the relationship between strain transfer and fracture? What is the relationship between chemical changes under heat, light, bias, oxygen, and moisture and both interfacial selectivity and adhesion? This article introduces a new framework, Interface–Mechanics–Chemistry Coupling (IMCC), which focuses on explicit linkage among interfacial selectivity and adhesion, strain transfer and fracture and chemical changes under heat, light, bias, oxygen and moisture. We introduce flexible all-perovskite tandems and expand on their device physics and architectures, highlighting the key differences with flexible single-junction, rigid all-perovskite and perovskite/silicon devices. Although the flexibility of all-perovskite tandems is proven, processing-aware recombination layers serve as both electrical contacts and barriers, molecule-bridged electrical contacts can enhance adhesion and reduce recombination, scalable in situ additive/gas-quench coating can offer large-area wide-bandgap films and durable modules, and nanoscale control of PEDOT:PSS phase distribution can transform wettability, interfacial dipoles and extraction in the narrow-bandgap subcells. Flexible single-junction studies provide fracture-mechanics perspective, and multifunctional nanocomposite studies provide insight into dissipation and self-healing, which are not yet demonstrated to be effective in flexible tandems. In the IMCC framework study the interplay of band alignment, ion migration and electronic axis (interfacial dipoles) with neutral axis (placement), fracture energy of interface (IF), and cyclic fatigue/fracture (MF) (mechanical axis) and Sn(II) redox (chemical axis) and halide segregation and moisture/oxygen ingress. We recognize gaps that are testable: lack of harmonized multi-stressor mechanical standards, lack of operando access to buried interfaces under deformation and lack of integrated control of the Sn-rich redox and wide-bandgap phase stability through low-temperature, scalable processing. Finally, we finish by discussing the translation from a coupon cell to roll-to-roll module and sustainability, where all the considerations of lead containment, recycle and choice of solvents are treated as part of the design process, not as a last resort.

Keywords Flexible All-Perovskite Tandem; Interface–Mechanics–Chemistry Coupling; Recombination Junction; Sn–Pb Narrow-Bandgap; Wide-Bandgap Perovskite; Solvent Orthogonality; Nanocomposites; Mechanical Reliability; Multi-Stressor Stability; Sustainable Processing

Introduction

The flexible PV market is no longer constrained by weight and conformability, but also energy and cost savings without sacrificing environmental management or economics over the life of the product—curved surfaces, wearable power, aerospace skins and rollable infrastructure. In this area, all-perovskite tandems are promising since they can current-match two low-temperature thin-film subcells to surpass the Shockley–Queisser limit and be compatible with polymer substrates. For flexible stacks, recombination layers and transport interfaces perform many roles other than energy selectivity, including: they must be able to resist low-temperature deposition, adhere under bending, resist moisture/oxygen, and resist solvents, not necessarily orthogonal [1–2]. Rigid all-perovskite tandems and perovskite/silicon tandems have proven high performance, but their thermal budgets, transparent-electrode options, and encapsulation regimes are all in stark contrast to flexible realities, where mechanical fatigue and polymer-barrier limitations play a major role in failure statistics.

Recent flexible all-perovskite tandem milestones set in the lab clearly outline a path towards reliable modules. Ultrathin conformal nucleation of transparent oxides using atomic-layer deposition (ALD) was shown to be possible on organic electron contacts, while reducing water-vapor transmission and promoting solvent orthogonality for the upper perovskite [1]. The molecule-bridged hole contacts then enhanced adhesion and reduced recombination at the interfaces of flexible tandems with power conversion efficiency (PCE) of up to 25% that showed superb bending endurance [2]. In situ additive techniques combined with continuous gas quenching delivered wide-bandgap films of tens of centimeters on PET, which equaled ambient-processed films in performance, and were certified for repeated bending and thermal cycling, up to ambient-processed modules [15] (ambient-processed film tandems above 27%). At the same time, by controlling the phase segregation at the nanoscale in an ultrathin (subcell) PEDOT:PSS layer, engineered surface chemistry and extraction of (NBG) subcells boosted flexible tandem performance to over 25% efficiency, enabling flexible mini-modules [16]. While these advances exist, there remain significant questions: What contribution does chemistry or mechanics make to these observed layers and what methods can be found to control the chemistry and mechanics of these layers without compromising scalable processing?

Coupling is essential for a critical answer. Electronic selectivity and adhesion are controlled by interfaces. Mechanics is the one that both determines the paths of cracks and the distribution of strains. The evolution of defects and ingress of environment determines both of them; in case of narrow- and wide-bandgap absorbers, the canonical problems are Sn(II) oxidation and halide segregation, respectively [17–20]. Our theory is that flexible performance and durability arises from the interplay of these three axes, and optimizing any one alone will lead to brittle gains that fail when subjected to multiple stresses, such as simultaneous illumination, heat, bias, humidity, oxygen, and cyclic strain. The linkages developed here are explicit and testable in the form of the Interface–Mechanics–Chemistry Coupling (IMCC) framework.

This is a strict evidentiary hierarchy review. We separately discuss flexible all-perovskite tandems, flexible single-junction perovskites, rigid all-perovskite devices,

and perovskite/silicon tandems. Adjacent-field literature is used, but only as an example of another method or precedent for design, not as a real-world example of PV performance [21–28]. Lastly, we place flexible all-perovskite tandems into the context of sustainability and systems that are increasingly determining technology adoption, such as solvents, lead handling and recycling, and life cycle performance [7–13,29–31].

Scope and definitions

Flexible all-perovskite tandems are monolithic devices that include two terminals (2T) or four terminals (4T) that both include halide perovskite absorbers grown on polymeric or ultrathin metal-foil substrates. All perovskite cells on a flexible substrate and with one active junction are considered flexible single-junction cells. “Rigid all-perovskite” devices are those that are mounted on a glass or rigid substrate. Unless otherwise stated, the tandems are rigid “Perovskite/silicon” tandems consist of a pair of top and bottom cells, one perovskite and one silicon. Throughout, class performance is not mixed together.

Mechanics is defined in terms of strains. The bending radius R and the total stack thickness h provide information on the surface strain ϵ under small-deflection bending, but the actual response of the device is influenced by the position of the neutral axis, the distribution of moduli between the layers of the stack, the orientation of the layers (tensile or compressive), the shape of the bending cycle, bending rate, dwell time, and the environment [4-5]. We follow the recent mechanical reports and the stability community as a whole, and use the reporting focus proposed to them: reporting on strain instead of only radius, defining failure metrics (e.g. T80 under given stressors), and having clear test conditions [5,14]. In the case of flexible perovskites, stability descriptors are used following the ISOS nomenclature where applicable, with the addition of the following caveat: "No widely accepted ISOS-equivalent mechanical protocol exists for flexible perovskites [5,14]."

Interfaces are considered coupled electrical–mechanical–chemical components. In 2T tandems, the “recombination junction” is an electrical recombination contact, as well as a processing barrier that must be solvent-orthogonal and conformal to the rough underlying layers [1]. Band alignment, defect passivation, adhesion, fracture energy and chemical reactivity with both absorbers are measured and assessed on the transport layers and interlayers. The term ‘device stack’ was applied to the encapsulation and barrier films, as they are not only packages, but also provide pathways for ingress, increase stiffness and move the neutral axis [3,5].

Review methodology and evidence-transfer protocol

We have three pillars to our methodology. First, we compile device physics and materials advances that are specific for flexible all-perovskite tandems, with a focus on reports that report devices' performance and mechanical/stability parameters explicitly under specific conditions [1-2,4-6,15-16]. Second, we use the IMCC framework to analyze how interface interventions impact on the interaction simultaneously via their electronic, mechanical, and chemical effects. Third, we use cross-domain evidence in the form of analogies and methods when they shed light on optical management, thermal regulation, nanocomposite design, or systems-level

constraints, but without promising PV results.

Standards and reliability. We compare our stability practices to ISOs and acknowledge that there are some areas that we are not yet in the same degree of mechanical standardization [14]. Based on recent attempts to link laboratory stressors with field lifetimes, we suggest that multi-stressor testing be used, which incorporates cyclic bending under maximum-power-point (MPP) operation, heat, humidity, oxygen, and light [32–33]. In our multi-scale, energy-dissipation approach to mechanical reliability, links are established between microstructure, interfaces and device design, such as neutral-axis engineering and interfacial fracture energy [5]. While tandem papers provide evidence of tandem performance, flexible single-junction studies provide a deeper insight into the mechanics, which we cite as mechanistic precedents, but not as evidence to the tandem effect [4,6].

Device-class distinctions. Flexible all-perovskite tandems are separated from flexible single-junction, rigid all-perovskite and perovskite/silicon devices. Thus, scalable rigid all-perovskite tandem modules with 21.7% PCE are relevant to solvent orthogonality and large area coating but do not necessarily demonstrate any flexible performance [34]. However, the flexible single-junction demonstrations of self-healing and dynamic bonding provide information on nanocomposite design, but need to be validated in tandem architectures [6].

Analogies and methods across domains. Light management and spectral coupling within thin, flexible stacks is discussed as analogies to how we may manage light with perovskite tandems – whispering-gallery-mode microspheres and bowtie/split-ring plasmonic geometries suggest methods for concentrating the field and shaping the spectrum where possible without exceeding thickness and roughness budgets [22,24–26]. The composition–property examples are cited in materials studies of tunable $\text{Cd}_{1-x}\text{Zn}_x\text{S}$ nanoparticles and oxide thermoelectrics, which do not constitute perovskite-device evidence [23,27]. We also draw on the results of the phase-change-thermal-management to set design constraints for the thermal design at the module level, as a precedent and not as a demonstration of the perovskite case. The value of flexible PV in a distributed context is emphasized by energy-systems and microgrid modeling, but has not directly informed device physics [35–37]. Sustainability and circularity analyses are used to inform our considerations of the co-design variables of solvent choice and lead management [7-13,29-31].

Novel about this review

The most novel aspect of this review is the IMCC approach to flexible all-perovskite photovoltaics. In contrast to a separation of interfaces, mechanics, and chemistry, we consider all three positions taken by interfaces—electronic (selectivity/recombination), mechanical (strain transfer/fracture/adhesion), and chemical (redox/ingress/ion-migration)—simultaneously. For instance, we demonstrate how a different band alignment can be realized by a recombination layer that also makes the stack stiffer, increases the stress under bending, and changes the moisture transport path, or how a soft, zwitterionic nanocomposite that dissipates strain can also passivate defects and chelate lead, but can also alter ion pathways or change solvent interaction when coating [1–6,12].

The second novelty is a taxonomy of advanced nanocomposites based on functions.

We categorize additives and hybrid layers according to the interfacial problem they address: passivation, adhesion, dissipation, barrier, optical management, self-healing; as well as measurable loading thresholds for percolation, modulus, transparency, roughness, and chemical reactivity. This addresses the missing dose–structure–function rules [3–6] for many flexible single-junction gains and the lack of translation of these flexible gains to flexible tandem.

Third, propose a multi-stressor stability map for flexible tandems that includes cyclic mechanical loading under MPP, temperature/humidity/oxygen exposure, and light soaking. This is in keeping with the demands for the lab to link to field, and for harmonization of reporting, and recognizes the lack of a mechanical protocol for ISOS [5,14,32–33].

Fourth, describe a translation ladder from “coupon cells” to “roll-to-roll modules”, including the principles of solvent orthogonality, large-area uniformity, the robustness of the recombination layer, interconnection resistance, encapsulation fatigue and yield, from recent large-area flexible results to manufacturability constraints [1–2,15–16,34].

Lastly follows a circular-design approach in which the efficiency and lifetime of sequestration of the lead, the sustainability of the solvent, and the recycling/remanufacturing aspects are defined as part of the design space as opposed to after-the-fact adjustments, in line with recent sustainability studies [7–13,29–31].

Device physics and flexible all-perovskite architectures

Figure 1 (original synthesis) outlines the architecture of flexible all-perovskite tandems, highlighting the dual-function of the buried interfaces as electronic contacts and, at the same time, mechanically and chemically strong. A recombination junction between a wide-bandgap (WBG) top absorber (~ 1.7 eV) and a narrow-bandgap (NBG) bottom absorber (~ 1.2 eV) is used in 2T monolithic tandems, which requires current matching and low loss interconnections. In 4T tandems, the subcells are optically and electrically decoupled but integration challenges shift to optics and interconnects. Flexible substrates add extra constraints: low thermal budgets, increased gas permeability, rougher surfaces, and residual stress, adhesion, and neutral axis control [3-5]. While numerical design of all-perovskite multijunctions offers a valuable upper bound and architecture-screening perspective, simulated stacks must be translated into potentially achievable performance through a combination of measured interfacial recombination, optical parasitics, processing tolerances and mechanical constraints [38]. [1–5,15–20,38–40]

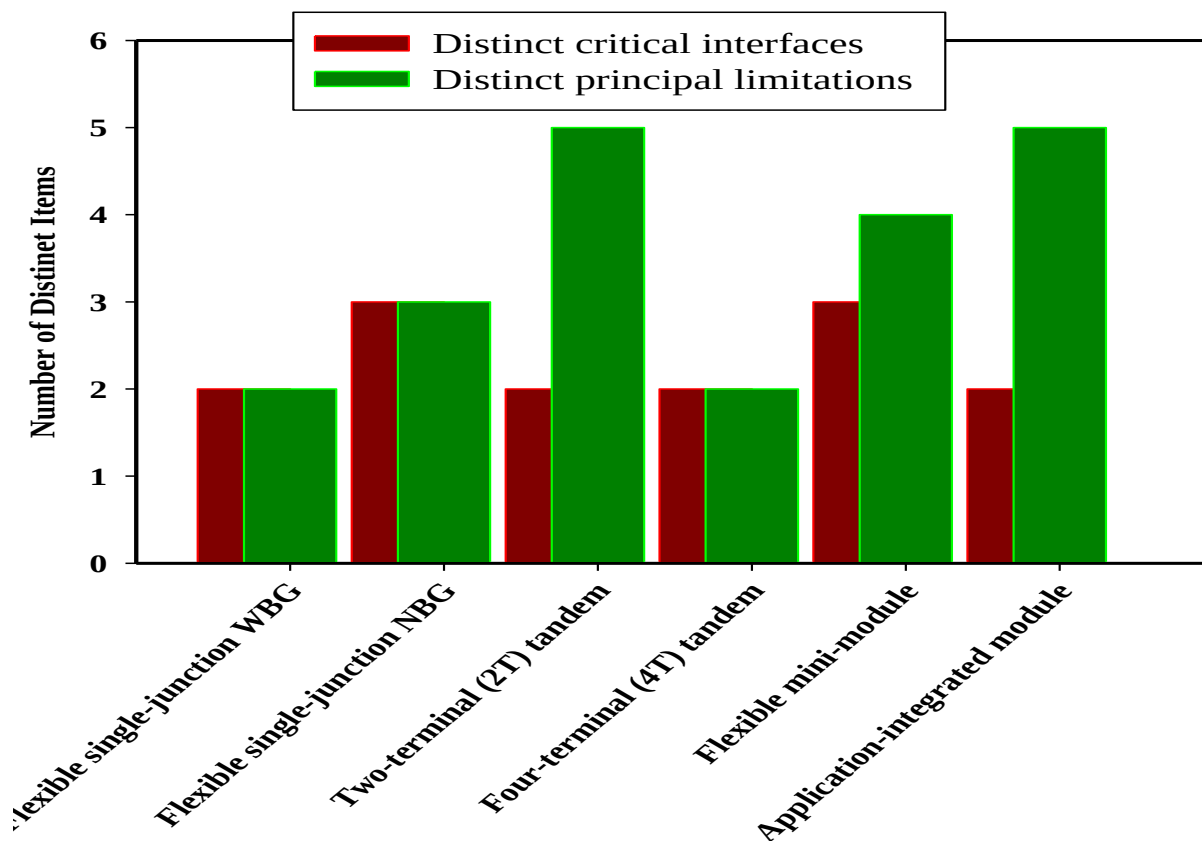


Figure 1: Complexity in interface coupling of flexible all-perovskite architectures. Bars are assigned for the number of distinct critical interfaces and principal limitations for flexible single-junction WBG and NBG cells [2–3,41], two-terminals and four terminals [1,38], flexible mini-modules [15–16,34,41] and application-integrated modules [9,28,35–36,41].

Bandgap pairing and current matching

Ideal 2T tandem combination requires a top cell with a high open-circuit voltage (VOC) and minimal risk of halide segregation (WBG) along with a bottom cell with long minority carrier lifetimes and a stable tin chemistry (NBG). Palmstrom et al. further developed the WBG alloying, finding a ~ 1.7 eV reduction with light by changing the light chemistry to cation size compensation, which reduces the occurrence of a crucial chemical failure mode, phase segregation, that is coupled with local strain and the formation of traps [1]. The major challenge in the NBG subcell is the control of Sn(II) oxidation resulting in p-type self-doping and recombination active defects. They include comproportionation chemistry in the precursor ink to buffer Sn(II)/Sn(IV) ratios [17], high quality bulk with microsecond scale carrier lifetimes [18], surface attached zwitterionic antioxidants to minimize Sn/SnO₂ interfacial redox [19], and passivation of the grain surface to reduce nonradiative losses [20]. Most of these strategies have been developed on hard substrates and they

need to be flexible: low temperature, solvent orthogonality and maintenance of mechanical compliance.

Recent research on the chemistry of the NBG interface for flexible devices has focused on the development of nucleophilic additives and interfacial treatments that can minimize shallow defects and promote adhesion, such as the passivation of NBG using carbonyldiimidazole and use of specific fluoride chemistry, which has shown to boost the NBG stability and performance, including flexible cell configurations, although these are not yet fully validated in flexible tandems [39-40]. However, when adjusting the band gaps and interfaces, current matching becomes sensitive to angular and spectral conditions, which in flexible applications can range from one to the other. In principle, the spectral response and field localization can be customized in ultrathin stacks using photonic/microwave/plasmonic resonators; earlier research on whispering-gallery mode microspheres and bowtie/split-ring photonic/microwave/plasmonics provides methodological, but not photovoltaic, examples of controlling the spectral response and field localization [22,24–26].

Two-terminal versus four-terminal designs

The advantages of monolithic 2T tandems are small thickness and the number of parts, and the ability to continuously coat them at low temperatures in the process makes them a preferred choice for flexible demonstrations. The recombination junction is a central challenge they have to face, due to its requirement to exhibit low optical/electrical loss properties and be chemically orthogonal not only to the neighbor absorbers, but also to the underlayer to which the polymer would be conformal. [1-2] Theoretically flexible 4T tandems will be able to relax optical coupling and the requirement of transparent electrodes, which should facilitate current-matching and possibly NBG redox management, but will add optical coupling and transparent electrode constraints to the stack which could increase the stiffness. The selection therefore combines all three IMCC axes: electrically, the addition of 2T junctions introduces a series element; mechanically, the addition of 4T stacks introduces the possibility of moving the neutral axis; chemically, the addition of 2T stack materials changes ingress pathways, and the materials of 2T and 4T together change the interfacial reactivity.

Recombination junctions and solvent orthogonality

The recombination junction in flexible 2T tandems has emerged as a processing gatekeeper. Palmstrom et al. implemented ALD AZO grown from a ~1 nm polyethyleneimine ethoxylated (PEIE) nucleation layer to enable conformal low temperature growth of AZO, which subsequently reduced water-vapor transmission through the stack, and then used non-orthogonal solvents for the WBG deposition without damaging the NBG [1]. This is a typical IMCC approach: electronically, the junction allows efficient recombination; mechanically, the oxide layer brings stiffness and adhesion pathways at the interface; chemically, the ALD process and resulting barrier layer lead to a reduction of the ingress and increase in the number of solvents. Li et al. modified the hole-selective contact by molecule-bridging the carbazole/phosphonic-acid units with low-temperature NiO nanocrystals, to minimize the interfacial recombination and enhance the adhesion with flexible substrates,

resulting in all-perovskite batteries reaching 25% efficiency and stabler bending through over 50 cycles [2]. In the case of ultrathin PSS films, Lai et al. demonstrated that the wettability and carrier extraction of NBG is critical to the film's nanoscale phase distribution, with lower surface PSS enrichment leading to better wettability and carrier extraction, and resulting in both NBG and flexible tandem performance improvements [16]. The combined studies put forward the idea that junctions and transport contacts can be viewed as electronic/processing co-designs, meaning a layer can have a direct impact on solvent orthogonality, energy-level alignment, and fracture pathways, which affects defect densities.

Scalable processing accentuates these couplings. High efficiency was sustained in an in situ additive coating under continuous gas quenching, which resulted in complete coverage of PET with void-free buried interfaces and low trap density, and translated to large area flexible modules with strong thermal cycling and bending endurance [15]. A series of rigid demonstrations of scalable monolithic all-perovskite tandem modules with PCE of 21.7% highlight the need for solvent orthogonality and integrity of interconnects, while moving to flexible stacks will encounter polymer-barrier fatigue and neutral-axis shifts [34].

Flexible substrate constraints and demonstrated benchmarks

The flexible substrates have thermal, mechanical and barrier requirements, which are key concerns in device design. Low-temperature processing is required to prevent polymer deformation, which makes solution processes a preferred method, as well as the low-temperature ALD and nanoparticle-based contacts [1-2]. In comparison to glass, polymeric substrates have a greater gas permeability and reduced modulus, making the role of encapsulation/barrier layers and adhesion more significant [3,5]. Roughness can act as a source of interfacial defects and local stress raisers, and residual stress due to solvent drying and thermal cycling can induce a bias in crack initiation [3–5].

Nevertheless, the performance of the flexible all-perovskite tandem has improved steadily, despite these limitations. Such representative values include 21.3% for early flexible 2T devices which used an ALD AZO junction with PEIE [1], 24.7% (24.4% certified) using molecule-bridged NiO hole contacts with durable adhesion [2] and 27.5% at 0.049 cm², flexible module with bending 10,000 cycles at 10 mm radius (~1% strain) and thermal cycling from –40 to 85 °C [15] and 25.4% in PEDOT:PSS phase-tailoring NBG, flexible mini-module at 19.7% [16]. These numbers are based on flexible all-perovskite tandems only tested under the same conditions; they are not combined with rigid or perovskite/silicon numbers.

The goal of flexible single-junction perovskite studies is to elucidate the mechanics and multifunctional materials, complementing this picture. It capped the surface of a 3D perovskite with an in situ low-dimensional capping layer that improved the 3D perovskite efficiency, operational stability and endurance to tensile-bending, resulted in a fracture-mechanics perspective in which bending stress scales with $E \cdot h / (2R)$, and urged the use of tension-only tests for interpretability because alternation of concave/convex cycles can confuse crack/peel behaviors [4]. A cross-linkable zwitterionic elastomeric interlayer provided self-healing properties through dynamic electrostatic and hydrogen bonding, and had a high flexible single-junction efficiency

and bending retention, indicating a potential for dissipation and damage tolerance that has yet to be demonstrated for flexible tandems [6]. In broader flexible perovskite reviews, the same key points of interfacial passivation, transport-layer design, and barrier strategies are highlighted, and there is a lack of the ability to harmonize mechanical testing [41–43]. Encapsulation as an engineered interface is also important for flexible devices for barrier-integrated transparent electrodes and mechanically compliant encapsulation to improve resistance to fatigue and ingress [44]. Simultaneously, it is noted that there are reports of interfaces with interpenetrating morphologies and chemomechanical multifunctionality, but the majority of these are shown on rigid substrates [45–46].

Table 1: Comparison of principal flexible all-perovskite architectures and potential locations of electronic selectivity, solvent protection, adhesion and strain transfer. As mentioned, the recombination junction in a monolithic tandem acts as an electrical contact and as a process barrier, while a mechanically stacked tandem has extra losses due to optical and interconnection between the cells and then trades recombination with current-matching requirements.

Table 1. Flexible all-perovskite architectures and interface-critical layers

Architecture	Typical absorber pairing	Critical internal interface	Flexible-specific opportunity	Principal limitation	Representative evidence
Flexible single-junction WBG	Mixed I/Br, approximately 1.7–1.8 eV	Substrate/TCO/HTL and HTL/perovskite	High voltage, semitransparency, mechanically thin stack	Light-driven halide segregation and voltage loss	[2–3,41]
Flexible single-junction NBG	Sn–Pb iodide, approximately 1.2–1.3 eV	PEDOT:PSS or SAM/perovskite; perovskite/C60	High current and direct relevance to tandem bottom cells	Sn(II) oxidation, self-doping, non-radiative recombination	[16–17,19,39–40]
Two-terminal flexible all-perovs	WBG top + Sn–Pb NBG bottom	Recombination junction and solvent-protection barrier	Highest voltage and specific-power potential	Current matching, parasitic absorption, shunts, solvent	[1,38]–[16,20,34]

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kite tandem			with fewer external contacts	damage, mechanical mismatch	
Four- terminal flexible tandem	Independently contacted WBG and NBG subcells	Optical coupling and laminated interface	Relaxed current matching and separate process optimization	Extra substrate/ electrode mass and optical loss	[1,38– 39,41]
Flexible all- perovskite mini- module	Series- connected monolithic tandem cells	Scribed interconnects , recombination junction, edge seal	Manufacturable voltage and larger active area	Dead area, lateral resistance , non- uniform coating, defect- driven yield loss	[15– 16,34,4 1]
Application- integrated flexible module	Tandem laminated integrated with wearable, aerospace, portable, or BIPV surface	Module/host adhesion and barrier edge	High power- to-weight ratio and conformal harvesting	Application- specific fatigue, abrasion, impact, thermal cycling, repair and safety	[9,28,35 –37,41]

Source: Authors' synthesis of flexible tandem architecture and scale-up studies [1–2,15–16,34].

The Interface–Mechanics–Chemistry Coupling framework

The IMCC structure (original synthesis) is shown in Figure 2, where three types of axes—electronic, mechanical, and chemical—intersect at buried interfaces, whose structure and composition dictate the performance and durability. Loop connections between the axes represent causal couplings, including electronic defects creating

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localizations of strain and thereby enhancing chemical reactions, mechanical defects causing new reactive surfaces and reconfiguration of the electric fields, and chemical species doping interfaces, changing adhesion and modifying band alignment. We use this construct to understand why, despite their positive intent, certain interventions failed when exposed to multi-stressor operation and to design multi-layered interventions that are called selective, compliant, and chemically stable. [1–6,14–20,32–33,39–40].

Figure 2. Interface-Mechanics-Chemistry Coupling (IMCC): axis engagement across the four coupling loops described in Section 3.4

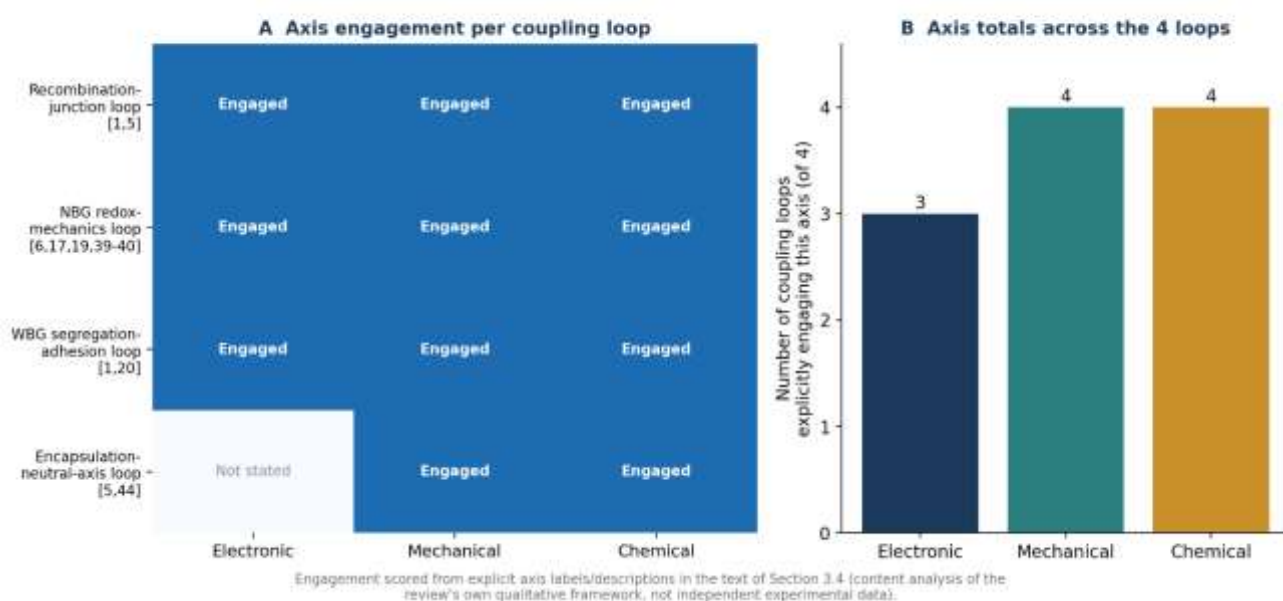


Figure 2. Interface–Mechanics–Chemistry Coupling (IMCC): axis engagement across the four coupling loops of Section 3.4. Panel A shows axis engagement for the recombination-junction loop [1,5], the NBG redox–mechanics loop [6,17,19,39–40], the WBG segregation–adhesion loop [1,20], and the encapsulation–neutral-axis loop [5,44]; panel B totals axis engagement across the four loops.

Electronic axis

Electronically, interfaces should offer a low resistance and selective transport loss, with minimal nonradiative recombination loss. The recombination junction and the adjacent transport layers are the major electronic bottlenecks in flexible all-perovskite tandems. The following is an example of a triple-function layer – PEIE-mediated ALD AZO junction – which allows for an ohmic-like recombination contact, minimizes moisture ingress, and allows solvent-orthogonal top-cell deposition [1]. Molecule-bridged NiO contacts reduce the recombination at the interfaces and enhance the electronic/mechanical contact adhesion without the need of high-temperature processing [2]. By tuning the surface dipole and wetting at the ultrathin limit, PEDOT:PSS phase engineering minimizes energetic barriers and enhances contact quality to NBG absorber [16]. In the NBG case, low recombination pressure on contacts is obtained by using low activation energy contact material (Sn–Pb) and

passivation [18–20]. Sn(II) oxidation at contact interfaces is suppressed by antioxidant and fluoride-based interfacial chemistries, which are important to control interfacial doping and trap formation [19,39-40].

Electronic improvements have to be driven by processing and mechanical limitations. For example, a thicker or denser oxide junction can lead to a decrease in optical/electronic losses, while at the same time causing an increase in stiffness, which shifts the neutral plane, and thus increases the strain at weak interfaces under bending [5]. Similarly, when polyelectrolytes are used to facilitate alignment, the dipole moments between the polymer and the interface can also change the distributions of ions under bias, which can have effects on recombination and chemistry [3]. These compromises lead to the development of function-first nanocomposites that are either passivating and aligning with little stiffness, or incorporate reversible bonds without sacrificed electronic functionality for the ability to dissipate strain.

Mechanical axis

Functionally, flexible devices need to withstand cyclic bending, handling, and retain a continuous conduction path and interfacial integrity. Stack thickness, the distribution of moduli and the location of the neutral axis of the stack determine the device-level strain under bending. Peak tensile/compressive stress is approximately proportional to $E \cdot h / (2R)$, which means that decreasing the overall thickness, decreasing the modulus of each layer and having the layers of low strength closer to the neutral axis all help to decrease peak tensile/compressive stress [4-5]. Delamination under cyclic stress is controlled by the interfacial fracture energy, which can be increased by the use of adhesion promoters and interpenetrating morphologies [3,46]. Raising the adhesion by introducing rigid oxides or dense cross-linkers, however, can make the stack stiffer and swap the peel resistance for a higher force driving the crack somewhere else.

Interpretation of mechanical tests is a matter of importance. Concave/Convex bending can temporarily “close” cracks, hiding damage, while only tension cycling produces clear failure signatures [4]. When reporting strain, it is necessary to report the tensile/compressive orientation, the cycle profile/rate and the electrical operating point under test for comparison with other reports and to correlate test response with the stressors in the field [5,14]. The recombination junction in flexible tandems is important because: it is generally the thickest and stiffest buried layer, and, if not conformal, can cause stress concentration [1]. Other transport materials like ultrathin PEDOT:PSS or NiO nanoparticle films affect mechanics by changing the local modulus and adhesion [2,16]. Reducing ingress can be achieved with barrier-integrated transparent electrodes and by designing them to be at or near the neutral axis; or by incorporating compliant sublayers will help to offset this trade-off [44].

Multifunctional nanocomposites are an exciting way to dissipate energy and stop cracks progressing. Zwitterionic elastomeric layers with dynamic bonds have shown self-healing and strain release in flexibly mounted single-junction devices [6] and interpenetrating organic–inorganic interfaces have shown higher fracture energy and operational stability on rigid substrates [46]. When such concepts are translated to flexible tandems, maintaining solvent orthogonality and low optical/electrical loss, while maintaining compliant mechanics, needs to be preserved.

Chemical axis

From a chemical perspective, the three challenges that flexible all-perovskite tandems must overcome are halide segregation in the WBG absorbers, the oxidation of Sn(II) and self-doping in the NBG absorbers, and the penetration of oxygen and moisture through the polymer substrates and edges. The suppression of the segregation of defects in the process of optimization of the crystallization kinetics was demonstrated to suppress the light induced segregation without compromising the target bandgaps [1,20]; the reduction of the bromide content by cation-size compensation in the WBG phase was demonstrated to improve the phase stability [1]. Comproportionation in the precursor is a useful tool to control the redox state of tin, zwitterionic antioxidants at surfaces, and defect density reduction and stabilization of Sn(II) in NBG chemistry [17,19,39-40]. The chemical controls are susceptible to electrical local fields and mechanical strains at cracks and delaminations, which expose new surfaces for reaction and provide field hot spots for ion migration and redox reaction, which can increase [3-5].

It is essential to have two important chemical levers — ingress control and solvent orthogonality. The ALD AZO recombination/barrier layer minimized water-vapor permeability of the stack and enabled the use of otherwise incompatible solvents in the top-cell deposition [1]. The results of the ambient-processed WBG films on PET with void-free buried interfaces indicate that the reactivity of the trapped species and defects can be decoupled from the ingress by controlling the crystallization process. The chemistry of supramolecular complexes and polymeric chelators now comes into play with lead management; if interface damage occurs, both species can bind lead and, without excessive penalty to their performance, additives or barrier layers could be developed to toughen interfaces and bind lead [8,10,12]. The studies on recycling and circularity show that high-quality material recovery is feasible and environmentally friendly, which will depend on encapsulant/barrier selection that can be separated and is compatible with aqueous or low-toxicity processes [7,13,29-31]. The choice of solvent for scalable coating should also take into account worker exposure and recovery as well as film quality, especially in the case of flexible roll-to-roll coating processes [31].

Thermal management is coupled with chemistry and mechanics: nonradiative recombination causes localized heating which can enhance diffusion and redox, and temperature gradients during bending can introduce a bias in stress. Although integration of PCM has been studied at module level to regulate thermal excursions, they provide a precedent that thermal design is also important for flexible perovskite tandems, which is not yet studied [28].

Coupling loops and design implications

The IMCC framework identifies recurring coupling loops that help account for successful and unsuccessful scenarios:

Recombination-junction loop. A conformal, low-temperature ALD oxide on a polymer compatible interlayer minimizes the ingress, allows for solvent orthogonality (chemical), offers an efficient recombination path (electronic) and provides an increase in interfacial stiffness (mechanical) of the polymer. The net effect is positive

when conformality and adhesion do not create stress concentrations, otherwise, concentrated stress under bending may start cracking stressing both the electrical continuity and barrier function [1,5].

NBG redox–mechanics loop. Hole density increases by Sn(II) oxidation, increasing the nonradiative recombination (electronic/chemical) that causes local heating and ion migration. Microcracks may occur as a result of cyclic strain and can expose new surface area, which can further accelerate oxidation and segregation. A decrease of chemical driving forces and strain localization can be achieved using either antioxidant interlayers or dynamic-bond nanocomposites, provided that the band alignment and low interfacial resistance is maintained when incorporating them [6,17,19,39–40].

WBG segregation–adhesion loop. The light-induced halide segregation in WBG absorbers creates potential fluctuation and defect clusters (electronic/chemical) that reduce the strength of GBs. These weak areas, which are more susceptible to bending, also break apart more readily under bending, resulting in more segregation and ingress of the material. Segregation can be counteracted by cation-size compensation and optimized crystallization, and adhesion-promoting transport layers help to distribute strain more evenly [1,20].

Encapsulation–neutral-axis loop. Barrier-integrated transparent electrodes and encapsulants lower oxygen/moisture ingress (chemical), but they can introduce stiffness which can alter the neutral axis (mechanical). The resulting shift of fragile perovskite/transport interfaces from the neutral axis may be detrimental for bending, as peak strains then increase, which could offset the barrier gain. It is therefore necessary to co-design the barrier modulus and thickness and achieve the symmetry of the stack [5,44].

These loops give rise to design implications and measurement priorities. Simultaneous stressors such as cyclic bending at a defined strain, MPP illumination under humid/oxygenated conditions should be evaluated and operando monitoring of adhesion, defect density, and ion accumulation at buried interfaces should be used to assess the engineering of interfaces [5,14,32–33]. Thickness and modulus distribution (to determine the position of neutral axis), not just bending radius, should be included in minimum reporting. Nanocomposite loading should be correlated with percolation thresholds (electrical/thermal), modulus targets (mechanical) and reactivity/chelation capacity (chemical) with explicit trade-offs reported. In scalable processing, interface conformality and solvent orthogonality should be maintained while avoiding the introduction of residual stresses which lead to delayed cracking. Last but not least, circularity needs to be considered as a co-design limitation: lead immobilization capacity and sequestration lifetime after mechanical fatigue, separable encapsulants compatible with aqueous recycling, and choices of solvent that consider film quality and environmental impact [7–13,29–31].

The IMCC framework does not supplant device physics, but clearly outlines the cross-effects that play a supporting role in the transition of flexible all-perovskite tandems to modules that are both long-lasting and manufactured. It also explains how the concepts that can be applied to the adjacent fields can be used as design hypotheses, not as substitutes for photovoltaic evidence [21–28, 37]. Moving forward, harmonized mechanical-stability protocols and operando metrologies at buried

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interfaces will be required to validate designs based on IMCC and to help bring lab data closer to the field-relevant reliability [5,14,32–33].

Some background contextual notes on the relevance of the broader system are warranted. The value of form factor and deployability is growing increasingly important in the distributed and hybrid energy systems sectors, and flexible PV could be used in conjunction with storage and dispatchable sources in microgrids and mobile infrastructure, but these are system level opportunities, and not necessarily device level evidence [35–37]. The long-term effects of the technology will depend largely on ongoing improvements in specific power and durability of the technology combined with solvent and lead management that meets new sustainability requirements [7–13,29–31].

Interfacial engineering across the flexible tandem stack

Interfaces not only determine the electronic selectivity and recombination but also have a decisive impact on stress transfer, adhesion, gas/vapor ingress and chemical reactivity in flexible all-perovskite tandems. The IMCC framework considers each interface's contribution to the coupled metrics they impose on carrier extraction, neutral-axis placement and fracture energy, and susceptibility to ion accumulation and redox chemistry. These interfaces and failure paths are mapped in figure 3, while modifier classes, mechanisms and flexible devices constraints are systematized in tables 2–3. [1–3,15–20,39–40,44–46].

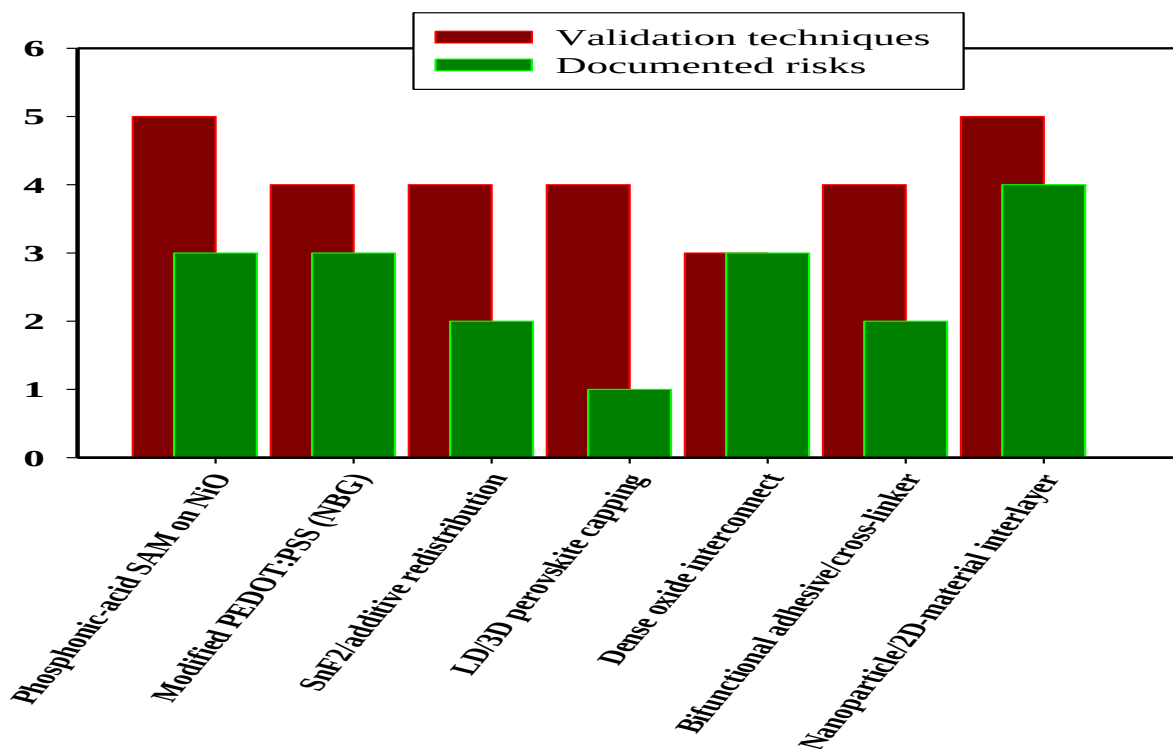


Figure 3. Validation burden versus documented risk across interfacial engineering strategies in the flexible tandem stack. Bars compare validation techniques against

documented risks for the phosphonic-acid SAM on NiO [2], modified PEDOT:PSS for the NBG subcell [16], SnF₂/additive redistribution [39], LD/3D perovskite capping [4], the dense oxide interconnect [1], the bifunctional adhesive/cross-linker [3,5–6], and the nanoparticle/2D-material interlayer [22,24–25,47,56,79].

Substrate/transparent-electrode and contact interfaces

The flexible substrates require the processing at low temperatures, higher surface roughness and higher thermal-expansion mismatch than glass, which increases the residual stress and delamination risk in the whole stack [3-4]. These transparent electrodes for polymers should therefore have a low sheet resistance, low permeability to water and oxygen, good adhesion and durability. There are three levels of evidence demonstrated for each device class:

Water-vapor transmission can be reduced by conformal low-temperature ALD Al-doped ZnO (AZO) over plastic using a 1 nm PEIE nucleation layer to enable conformal ALD to the top subcell, as done by Palmstrom et al. [1]. A follow-up study by Li et al. then showed that molecule-bridged contacts could be used with low-temperature NiO nanocrystals on flexible substrates, to enhance adhesion and minimize recombination in 10,000-bending-cycle-stable tandems with a 15 mm bending radius. With in situ additive coating and continuous gas quenching on PET, the following tests showed high TE/process compatibility: 10,000 bends at 10 mm radius and thermal cycling from –40 to 85 °C [15].

Flexible single-junction perovskites: Dong et al. correlated interfacial flaw populations and fracture mechanics with mechanics-limited reliability, and suggested that not only should strain be reported but also radius, taking into account the $E \cdot h / (2R)$ scaling and neutral-axis effects [4].

All-perovskite or rigid-module precedents: Scalable monolithic tandem modules on glass were highlighted for uniform TE deposition and recombination-contact integrity during roll-to-roll-relevant drying, which were not tested with flexible mechanics [34]. These studies are still references for architecture, not flexible stack validations.

Barrier-film thinking, as a result of membrane work, is also relevant to the design of the interfaces between substrate and TE. While not quite in the form of photovoltaic demonstrations, layered or laminar architectures that include couplings between structure, permeation, and mechanics, such as MXene-based membranes in water purification, illustrate structure–permeation–mechanics couplings that can be applied to flexible TEs and barriers. Likewise, silica–polymer nanocomposite membranes and ceramic gas-separation membranes express analogous trade-offs between modulus, free volume, permeation selectivity, and thermal stability as do flexible PV encapsulation and transparent electrode stacks between water/oxygen barrier properties and adhesion/fatigue. Similarly, the modulus, free volume, permeation selectivity, and thermal stability trade-offs of silica–polymer nanocomposite membranes and ceramic gas-separation membranes are similar to those made between the water/oxygen barrier properties and adhesion properties/fatigue of transparent electrode stacks and flexible PV encapsulation. The tuning of surface charge for the control of adsorption and wetting phenomena in separations implies a similar control of the interfacial dipoles and contact resistance in perovskite stacks [52]. The above cross domain examples serve to further underscore the potential for ALD-oxide to

provide both barrier and work-function control in flexible tandems [1].

The small amounts of oxide nanoparticles can effectively increase the modulus and wear resistance of PNs in dental composites, but can also modify the surface chemistry and surface roughness, which are seen again when loading flexible TEs or adhesives with conductive or barrier nanofillers [53]. In PV, similar ionic modifiers can influence the hydrophilicity and biocompatibility of the polymer matrices, can act as plasticizers or stiffeners, and can enhance the adhesion at interfaces; however, they can also cause shifts in the work function and the formation of traps in the contact layers. Generally, dispersion and aggregation laws from the reviews of metal-oxide nanomaterials [56] and changes in mechanical reliability and barrier function of hybrid particles according to synthesis route, ligand selection and blend morphology [57] should be considered, which means that the same filler might have a positive or negative effect.

Substrate/TE interfaces are also thermal interfaces. Gradients can be generated during lamination or by local heating in ALD or laser interconnection processes in polymer electrodes and adhesives. Local heating during lamination, ALD or laser interconnection can cause gradients in polymer electrodes and adhesives. Although not a photovoltaic study, computational fluid dynamics and radiative/convective transport studies of heat-exchanger optimisation and nanofluid flow [58–61] suggest that there is a need to quantify the thermal diffusivity, heat spreading and cure profiles in multilayer flexible stacks. Recent flexible perovskite demonstrations in which the thermal diffusion of the thermally diffusive layers and barrier-electrode laminates are improved have confirmed that hot-spot induced delamination and device leakage under mechanical loading can be reduced by improving in-plane thermal transport, but these were single-junction cells, not all-perovskite tandems [44,62].

Hole-selective interfaces in WBG subcells

The large band-gap (LBG, ~1.7-1.8 eV) top absorbers control the series current and may also have a large influence on tandem reproducibility because of their sensitivity to interfacial recombination and bromide rich phase separation under illumination and bias. Low-temperature hole-selective interfaces need to satisfy electronic selectivity, mechanical adhesion, and solvent orthogonality requirements on flexible tandems. There are two ways that come to mind:

Molecule-bridged NiO: Li et al. used molecule of carbazole/phosphonic-acid molecules to bridge low-temperature NiO nanocrystals to the WBG perovskite, which reduced the interfacial recombination and enhanced adhesion, achieving 24.7% PCE in flexible all-perovskite tandems, with durability to 10,000 bends [2]. The phosphonic anchor rationalizes persistence of bond with oxides, the conjugated backbone facilitates energy-level alignment and can passivate perovskite surface states. The same interface also decreased the likelihood of interfacial fracture under bending situations, which is in line with the IMCC principle that both chemical bonding and mechanical robustness are optimized [2,4].

Lai et al. discovered a surface effect of surface chemical segregation of the PSS phase in ultrathin PEDOT:PSS films, which caused surface hydrophobicity and hindered wettability and extraction. Wetting, carrier extraction, and reproducibility were improved by reducing the surface PSS/PEDOT ratio of Triton X-100, which resulted

in flexible all-perovskite tandems with 25.4% and a flexible mini-module with 19.7% [16]. It is a study of nanoscale composition gradient controlling both processing compatibility and interfacial dipole of tandem stacking, which is mechanistically clear.

Recent, all-perovskite studies in a more rigid structure confirm that passivation and compositional control at the WBG interface can reduce nonradiative losses and light-induced halide segregation [20]. Palmstrom et al. employed cation-size compensation to lower the Br content to around 1.7 eV with more stable halide distribution and enhance the orthogonality of the processing sequence at the recombination contact [1].

Adjacent, interpenetrating interfaces with better mechanical interlocking and percolation have been demonstrated to increase stability and robustness of rigid perovskites [46]. The transferable concepts of quantum-dot solar cells suggest that oxide morphology and aspect ratio at the hole contact influence the interfacial area, trap densities and pathways of movement of the carriers, hence the nanostructured NiOx or organic hole conductors on flexible substrates should be tested with the view of studying the electronic and mechanical consequences. By analogy, the use of chemical adducts in organic synthesis (e.g., TCT:DMF) suggests that the same should be achievable in WBG inks by exploiting perovskite–solvent–additive adducts to achieve uniform nucleation on rough flexible contacts without the need of high temperatures. In the case of WBG film formation on modified hole contacts, it is possible to make use of a transferable framework for surfactant–polysaccharide interactions which affect conformation and charge [65] in a manner similar to the PEDOT:PSS surfactant strategy [16].

NBG Sn–Pb buried interfaces and redox control

The unique redox and defect-chemistry challenges of narrow-bandgap (NBG 1.2–1.3 eV) Sn–Pb perovskites are crucial for all-perovskite tandems. Rigid all-perovskite tandems have proven that the oxidation of Sn(II) could be suppressed by comproportionation of precursor inks and that the use of surface-anchored zwitterionic antioxidants or better surface passivation of the grains can minimize self-doping and recombination [17–18]. The translation of these chemistries to flexible stacks imposes additional constraints such as low-temperature processing, polymer substrate roughness, increased oxygen permeability, and stress concentration factors at buried interfaces due to bending [3–5]. The materials and interfaces—such as cross-linkable zwitterionic perovskite elastomers with control over crystallization, release of residual strain, and healing capabilities [6]—bear out the potential of multifunctional nanocomposites, but are not yet general-proof for flexible all-perovskite tandems.

The recent NBG-specific buried-interface techniques are applicable to molecule-assisted nucleation and post-deposition treatments. Sn vacancies and iodide interstitials can be reduced by the addition of Tin fluoride in inks and nucleation control; mechanistic studies revealed F[−] complexation pathways that prevent the formation of undesirable iodide-rich phases in NBG films and flexible tandem data sets are starting to incorporate such chemistries [39]. It was reported that surfaces of NBG could be passivated and flexible single-junction performance could be improved

by post-treatments, like N,N'-carbonyldiimidazole. In IMCC, these modifiers should be investigated for: (i) redox buffering of Sn(II)/Sn(IV) under light and bias, (ii) ion-blocking at NBG/contact interface, and (iii) fracture-energy enhancement and flaw blunting under cyclic bending.

The mechanism of cross domain redox science, here applied in the form of a transferable mechanism, explains the failure of the NBG buried interfaces under oxygen/light/bias. In water treatment, advanced oxidation processes are those involving the generation of radicals, metal-redox cycling and ligand-assisted electron transfer that enhance the oxidation of the substrate [66–71]. Similarly, if there are no scavengers or antioxidants at the interfaces, the photoexcited carriers, superoxide formed at the electron-transport surfaces, and minute amounts of moisture can induce Sn(II) oxidation and the loss of halides in perovskites. While the above are conceptual ideas and not proof of photovoltaic demonstration, the design principles of the shuttle species, intermediate complexes, and interphase chemistry in lithium–sulfur batteries, which determine lifetime [72] of the cells, are applicable to the design of NBG interlayers that immobilize mobile Sn/I-containing species, or intercept reactive oxygen at contacts. Another aspect of the chemical stability and reactivity of the organotin carboxylates [73] should also caution for choosing the passivator, such as an organotin complex or its oxide, which will be low in volatility and inactive towards photooxidation in the perovskite environment.

The NBG interface chemistry and flexible processing can be further coupled with green solvent systems and adducts. Although not employed in high-efficiency perovskites, the Lewis basicity, viscosity and hydrogen bonding networks of deep eutectic solvents (DES) would be able to tune Sn–I complexation and nucleation at low temperatures if they were found not to lead to residue or energy-level effects. Likewise, the formation of adducts (e.g., with DMF-like donors) is a kinetic lever that can be controlled [64]. Redox buffering and ion blocking are possible by the use of Nanocomposites, e.g., as doped carbon nitrides as redox reservoirs [75] or sulfide/oxide nanoparticles of controlled ize prepared by dynamic light scattering [76] but must be coordinated with transparency and mechanical-stiffness budgets in NBG stacks.

Recombination junctions and chemical orthogonality

Monolithic tandem recombination junction is required to simultaneously satisfy four properties: (i) low optical and electrical losses, (ii) solvent and chemical orthogonality of subcells, (iii) barrier to oxygen/moisture and (iv) mechanical fatigue resistance. Both recombination contact and processing barrier, Palmstrom et al. demonstrated conformal low-temperature ALD AZO on fullerene, which reduced water-vapor transmission and allowed non-orthogonal solvents for the top cell in a rigid tandem and flexible tandem of 23.1% and 21.3% efficiency, respectively [1]. High efficiency was also achieved for large-area flexible tandems fabricated by in situ additive coating and gas quenching, even after bending and thermal cycling, coupled with orthogonal wetting and nucleation at buried interfaces [15]. These are straightforward flexible all-perovskite tandem demonstrations.

Rigid scalable tandems demonstrated the resistance to interconnects and robustness of the recombination layer during upscaling [34]. Applying the same concept of

interpenetrating interfaces in a rigid cell [46] may provide a pathway to enhance the fracture energy at the interface in a flexible tandem, which is yet to be proven. The multiscale energy-dissipation perspective—which links the energy dissipation per unit of interfacial fracture with the number of reversible bonds and the location of the neutral axis—suggests that recombination contacts can dissipate strain and capture mobile ions [5]. Again barrier-membrane analogies highlight the routes toward chemical orthogonality that can be achieved using multilayers and graded interfaces without brittle thickness [48-49,51].

Encapsulation as an active interface

In flexible tandems, the encapsulation is often externalized as packaging, but in fact it is an active interface which defines the water/oxygen fluxes, neutral-axis position, interfacial adhesion and failure mode under cyclic bending. Flexible tandems already used thin ALD oxides in the stack (e.g. AZO on fullerene) as internal barriers [1]. Transparent-electrode-integrated barrier films illustrate simplified lamination and reduced interfacial counts in flexible cells with single junctions [44] while flexible perovskites reveal that increasing the thermal diffusivity of the encapsulant can reduce the occurrence of thermal/mechanical runaway and limit lead migration in cases of damage [62] also in single-junction applications. All-perovskite flexible tandems still have potential, but they need to demonstrate the integrity of the recombination layer and/or optical penalties.

Lead management takes the moisture barrier to the level of a chemical sequestration interface. Studies of rigid modules revealed that high lead recovery can be achieved and transparent conductor can be recovered from weakly acidic resins, and even reused in the aqueous process [7,13]. However, for flexible modules, the requirement for low-temperature delamination and separation of both the WBG and the NBG layers necessitates barrier/adhesive chemistries that are compatible with the aqueous process. Risk-mitigation frameworks advocate for physical encapsulation in combination with chemical capture and recycling, and for considering the efficiency of sequestration and its lifetime in addition to the costs and penalties related to performance. [8,10] Chemically integrated sequestration of lead using supramolecular complexes that immobilize the lead, conceptually similar to multifunctional nanocomposites that also toughen and passivate interfaces [12]. Sustainability analyses recommend reliability and recovery yield be treated as variables that are co-designed [9,11].

Materials selection is informed by cross-domain capture and barrier precedents, such as chitosan-based nanocomposites immobilizing metals and the mechanical compliance, as well as the surface-functionalized oxides that adjust adsorption selectivity, layered MXene laminates that exhibit high tortuosity and surface reactivity, and synthesis routes that alter particle interfaces drastically, which promotes the change of adsorption and stability. Impregnation with AOPs highlights the role of radical scavengers or redox buffers within encapsulants to prevent the presence of reactive oxygen species (ROS) at Sn-rich buried interfaces, without adding optical and electronic losses [66–71,75]. Rapid oxidation of dyes have been achieved using ozone and peroxide based processes [67-68,78] and this could be a point that requires encapsulation to be resistant or quench these species when exposed

to UV rich outdoor environments, but this is more akin to a mechanical analogy than as a photovoltaic device test.

Critical comparison and design rules

Data differences by device class help clarify the difference between evidence and plausible:

In the mechanical constraint, concrete solutions are offered by flexible all-perovskite tandems that feature seeded ALD-oxide recombination contacts, both as recombination contacts and as barriers, molecule-bridged NiO hole contacts, and surfactant-tuned PEDOT:PSS with controlled vertical segregation, and in situ additive/gas-quench processing on PET which has been shown to be flexible and bendable and thermally stable up to 180 °C and 150 °C respectively [1, 2, 15, 16]. The bending radius, cycle counts and in some cases, even translation in the module scale, is explicitly reported in these studies.

Flexible single-junction perovskites: low-dimensional capping layers to seal surface flaws and raise fracture energy [4]; self-healing zwitterionic elastomers releasing residual strain [6]; integrated barrier-electrode laminates [44] support the notion of using IMCC-driven strategies and need to be tested in tandems.

Rigid all-perovskite devices rely on comproportionation inks, surface-anchoring antioxidants, and advanced passivation [17-20]; these need to be re-optimized for polymer-substrate roughness, oxygen permeability, and bending.

There are no photovoltaic validations for the following mechanistic templates: Cross domain analogies, Membrane barriers [47–51], Ionic/polymer additives [54–55,64–65], Nanoparticle dispersion [53,56–57], Redox/oxidation chemistry [66–71,75], and Thermal-transport modeling [58–61].

In IMCC, we summarize the following design rules for flexible all-perovskite tandems:

Interfaces need to be dual or triple functional, namely, electronic selectivity, strain dissipation and chemical buffering, without adding brittle thickness. ALD AZO, for instance, at the recombination part of the junction should be as thin as possible to prevent gas ingress, but sufficiently compliant to be anchored by appropriate molecular seed [1].

Generally, buried interface adhesion and fracture energy should be evaluated under the appropriate strain state; tension-only tests should be replaced by convex/concave tests to avoid the interpretation of the crack initiation or closure [4]. If possible, position the neutral axis in the most brittle functional stack, via graded-modulus encapsulation [5].

Level alignment and transparency of Sn–Pb interfaces should be achieved by the use of redox buffers and ion blockers, which should immobilize mobile species and scavenge reactive oxygen as well [17-20,39-40]. Potential are multifunctional zwitterions and cross-linkable elastomers, but dose–structure–function relationships need to be developed to prevent stiffening or optical loss [6].

Orthogonality of wetting and solvents should be built into the recombination junction – the phase composition of PEDOT:PSS or molecule-bridged NiO can be used to tune the surface energy for defect-minimized overcoating by WBG inks in tandems [2,16]. Flexible processing windows [64–65] should be optimized by the chemistry of the

adducts and selection of the surfactant.

The use of encapsulation should be viewed as an active interface designed for barrier tortuosity, adhesion, neutral axis control and chemical sequestration; the latter should be quantified after mechanical fatigue, not just on pristine devices [8-12,44].

Table 2 compares and contrasts the various mechanisms, benefits, risks and flexible-device constraints of interfacial engineering. The key message is that improving band alignment is not enough for an interface treatment to be successful; it should also maintain adhesion, chemical orthogonality, and cyclic mechanical integrity [1–2,15–20,39–40,44–46].

Table 2. Interfacial engineering strategies, mechanisms, benefits, and risks

Interface or strategy	Primary mechanism	Electronic benefit	Mechanical/chemical benefit	Key risk or trade-off	Minimum validation
Phosphonic-acid SAM on low-temperature NiO	Molecular anchoring and energy-level tuning	Reduced interfacial recombination and faster hole extraction	Improved adhesion and bending durability	Incomplete coverage, aggregation, sensitivity to TCO surface chemistry	Contact angle, UPS/KP FM, PLQY, peel/fracture test, cyclic MPP-bending [2]
Modified PEDOT: PSS for NBG subcell	Suppression of vertical PSS segregation and interfacial dipole	Higher hole extraction, fill factor, and reproducibility	Better wettability; lower hygroscopic surface fraction	Excess surfactant can over-wet, disrupt film, or migrate	XPS/ToF-SIMS depth profile, sheet resistance, aging, bend test [16]
SnF ₂ /additive redistribution	Defect compensation and control of buried fluorinated	Lower trap density and higher NBG voltage	Partial suppression of Sn oxidation and ion migration	Non-uniform accumulation and dose-dependent insulation	Depth-resolved chemistry, Sn(IV)/Sn(II), PLQY, bias aging

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LD/3D perovskite capping	phases Surface -flaw sealing, passivation, environmental barrier	Reduced non-radiative recombination	Higher crack resistance and reduced ingress	g phases Transport barrier if too thick or poorly oriented	[39] GIWAXS, TRPL, tension-only fatigue, fracture microscopy [4] WVTR/solvent challenge, lateral/out-of-plane conductivity, bend fatigue [1] Fracture energy, modulus mapping, extraction kinetics, repair/recycling trial [3,5–6] Dispersion, loading series, AFM, spectroscopy, dark/light bias aging [22,24–25,47,56]
Dense oxide interconnect on nucleation layer	Conformal ALD growth and solvent/sputter protection	Low-loss recombination contact and fewer shunts	Chemically orthogonal sequential processing	Vacuum throughput, brittle oxide fatigue, parasitic absorption	
Bifunctional adhesive or cross-linker	Multi-point bonding across buried interface	Preserved contact area and lower interfacial recombination	Higher adhesion and stress dissipation	Cross-link density may hinder charge transport or recyclability	
Nanoparticle/2D-material interlayer	Surface-energy tuning, defect coordination, local field control	Passivation, selectivity, possible optical enhancement	Barrier or toughening functionality	Agglomeration, roughness, shunts, catalytic side reactions	

,79]

Source: Authors' synthesis of interfacial engineering and buried-interface studies [1–2,15–20,39–40,44–46].

Stability enhancement under coupled stressors

The electronic selectivity of the IMCCs that underlie high efficiency must also be able to withstand cyclic strain without the formation of crack tips or pathways for oxygen and water, and must chemically buffer halide redistribution, Sn redox, and volatile loss under illumination and bias to ensure device stability in flexible all-perovskite tandems. The evidence base is still inconsistent in terms of stressor and device class; in this section, we synthesize intrinsic and extrinsic pathways, the effect of mechanics on chemistry and the ability to harness encapsulation and interface chemistry.

Intrinsic pathways: ion migration, phase segregation, Sn oxidation, volatile loss

In WBG and NBG absorbers, high defect generation and/or poor surface passivation of the grain boundaries increase the rate of ion migration and field-driven redistribution. Rigid all-perovskite studies showed that lowering the Br content of WBG films, through cation-size compensation [1] and grain-surface passivation [20] respectively, reduced the mobility of I[−]/Br[−] in these films. Flexible tandem reports with molecule-bridged NiO and phase-managed PEDOT:PSS enhanced the wetting and contact selectivity of the WBG, which resulted in the indirect reduction of ionic accumulation at the hole interface during operation [2,16]. But, the kinetics of halide segregation under cyclic bending and bias are poorly known. The oxidation of Sn(II) to Sn(IV) creates p-type self-doping and deep defects in the NBG [17–20]. These pathways can be diminished in rigid and flexible single-junction cells by various means including the use of comproportionation in precursor inks, surface anchored antioxidants, fluoride additives, and post-treatments [17-20,39-40]. However, the redox reactions at the buried NBG interfaces during deformation, which can create new surfaces, affect the integrity of the barrier and focus electric fields, are rarely measured [3-5].

In addition to ion migration and redox chemistry, volatile loss, especially of organic cations or solvent residues, will interact. Since the HV reports reduce volatile components, flexible processing at low temperature can result in higher residual amounts or more soft interphases that change when processed at higher temperature and light exposure [3]. Thus, the efficacy of passivation should be assessed in conjunction with volatility and photochemical stability of additive strategies. Examples of this trade-off include organotin complexes as interfacial passivators [73] where strong bonding passivates Sn sites, but labile organotin species could lead to volatility or undesirable reactions under illumination, requiring careful residue testing. These intrinsic pathways are supported by cross domain mechanisms. Reactive oxygen species (ROS) in AOPs react with electron-rich sites, resulting in the destruction of organics [66–71]; the surface terminations of perovskites and organic cations are susceptible to similar reactions when ROS are formed at the contacts under illumination and trace oxygen. Built-in fields at nanoscale junctions, which have been shown to increase interfacial redox rates in catalysis [76,80–84] but are a drawback when they are not intended in perovskite buried interfaces, support the idea

that fields are beneficial for improving the rates of interfacial redox. These are mechanistic similarities, not photovoltaic proof, and inspire the use of “redox buffers” on the interfaces, that block the ROS sites at the perovskite structure and that capture mobile ions.

Extrinsic pathways: oxygen, water, heat, light and bias

In flexible tandems, the fields of permeation, thermal transport and stress concentration are strongly coupled in and through the polymer substrates combined with thin inorganic barrier layers and soft interphases. Standardized reporting of light, heat, humidity and electrical bias are highlighted in the ISOS consensus [14]. Jiang et al. suggested that translation needs to be performed in the laboratory under the right activation spectra, humidity cycling and electrical operating conditions [32]; standards-oriented analyses require failure definitions and minimum reporting for stability claims [33]. The use of such norms is still very rare for flexible all-perovskite tandems.

Barriers of AZO recombination barriers showed a decrease in water-vapor transmission in Palmstrom et al. [1]; molecule-bridged contacts showed no loss of initial performance after 10,000 bends in flexible tandem operation [2]; and in situ processed PET-based tandems and modules showed that they could complete thermal cycles and bright-light operation without losing performance after mechanical fatigue [15]. However, cyclic mechanical loading-induced ingress of oxygen and simultaneous light/bias-induced chemistry changes at the interfaces are under-reported.

There are other, class-specific, pieces of evidence, such as the capping layers that sealed surface flaws, increasing the T90 and bending endurance [4]; the cross-linkable zwitterionic elastomers that provided self-healing after low-temperature anneals [6]; and the electrode-integrated barriers that simplified encapsulation [44]. The following strategies might also be applicable to tandems, but need to be validated for the compatibility of recombination junctions and auditing of the optical loss. A series of flexible perovskite designs that focused on environmental considerations have been developed, which have resulted in higher thermal diffusivity to limit hot spots and reduced leakage of lead, as measured in single-junction devices [62].

Water and oxygen control is crucial in the encapsulation design. The literature on barrier-membrane emphasizes that multilayer tortuosity slows permeation without thick, brittle films [48-49,51] and barrier-membrane MXene laminates enable tunable gas/liquid permeation by restacking 2D layers [47]. From the transferability of adsorption science, it is known that water uptake and pollutant binding is controlled by the barrier top layer and thus, the chemical nature of the top layers of barriers can be tuned to bind and immobilize corrosive species at their edges. These synthesis routes for changing particle interfaces and nanocomposite integration (56,77) set dispersion and mechanical integrity that influence cracking susceptibility during thermal cycling.

Light and electrical bias modifies degradation paths by photocarrier generation and field-assisted ion transport. In the AOPs and photocatalysis, the mechanisms involve the formation of different radicals depending on the wavelength, and the use of co-catalysts and substrates which are clearly present in transport layers and at the

interfaces of electrodes, and need to be validated for perovskite materials. The relevance of bias conditions is highlighted by rigid scalable tandems [34] and consensus protocols [14,32–33] that find the maximum power point during stress to show more realistic failure onset compared to open circuit illumination.

Thermal management has an impact on chemical and mechanical pathways. The modelling of convective–radiative flows, the modelling of inserts to increase heat transfer, and the modelling of radiative nanofluids [58–61] can be used for quantifying the temperature distribution during the lamination process, during ALD and outdoors. The general rule is that flexible stacks with soft, anharmonic layers will carry inbuilt thermal gradients unless in-plane thermal diffusivity is controlled, although this is not a perovskite study, the lesson to be learned is that thermal gradients can be localized by strong anharmonicity in some materials [62, 85].

Mechanical acceleration of chemical degradation

The stress coupling of flexible tandems is very high due to the concerted effect of polymer substrates, thin inorganic barrier layers and soft interphases on permeation, thermal transport and stress concentration. Standardized reporting of light, heat, humidity and electrical bias is emphasized by the ISOS consensus [14]. Jiang et al. suggested the importance of reproducing the right activation spectra, dehumidification–humidification cycles, and electrical operating parameters for lab-to-field translation [32] and the necessity for failure definitions and minimum reporting in standards-oriented analyses regarding stability claims [33]. The norms are not often followed for flexible all-perovskite tandems.

Progress is demonstrated by the flexible tandem that was successfully shown to maintain water-vapor transmission when recombination barriers were introduced in Palmstrom et al. [1]; the molecule-bridged contacts that maintained performance after 10,000 bends [2]; and the in situ processed PET-based tandems and modules that were tested for thermal cycling or bright-light operation to prove that performance was not lost after mechanical fatigue. However, the ingress of oxygen during cyclic mechanical loading and the change of interfacial chemistry during simultaneous light/bias is underreported.

Additional evidence of the flexibility of single-junction cells included capping layers to seal surface defects to improve T90 and bending endurance [4] cross-linkable zwitterionic elastomers for self-healing following a low temperature anneal [6] and electrode-integrated barriers that simplified encapsulation [44]. These strategies might also be applied to tandems, but would need to be recombinable and loss audited. Flexible designs oriented to the environment raised the thermal diffusivity to reduce hot spots and hot spots were also reported to be lower in single-junction designs [62]. The design of the encapsulation is key to water and oxygen management. Barrier-membrane literature emphasizes that the multilayer tortuosity is capable of decreasing the permeation, even for thick, brittle films [48-49,51] and MXene laminates demonstrate tunable gas/liquid permeation by restacked 2D layers [47]. The ability for water uptake and pollutant binding as identified by transferable adsorption science suggests that chemically tuning the top-layer of a device can help bind and immobilize corrosive species at the edges of materials. The dispersion and mechanical integrity from synthesis pathways that change particle interfaces [57] and

nanocomposite integration [56,77] dictate cracking propensity during thermal cycling. Degradation routes change with light and electrical bias, due to photocarrier generation and field-accelerated ion motion. However, the mechanisms of degradation in AOPs, such as photocatalysis, which involve the formation of radicals depending on the wavelength of the light and the presence of co-catalyst and substrate [66-70,75,84] have clear parallels to that of transport layers and electrode interfaces but must be validated for perovskite materials. The role of bias conditions is illustrated in the rigid scalable tandems [34] and consensus protocols [14,32-33] which show that during stress MPP-tracking is likely to give a more realistic failure onset than open circuit illumination.

Thermal management has an influence on both chemical and mechanical pathways. Models have been developed to describe convective–radiative flows, inserts that improve heat transfer and radiative nanofluids [58–61] aid in the quantification of the transient temperature distribution for lamination, ALD, and outside the laboratory. Low lattice thermal conductivity in some materials is due to high anharmonicity which can focus heat [85] and, although the material is not a perovskite, the overall message is that the in-plane thermal diffusivity needs to be tuned in flexible stacks to avoid high lattice thermal conductivity gradients.

Encapsulation, redox buffers, and ion-blocking strategies

An IMCC-consistent stability strategy is the combination of encapsulation and interfacial redox and ion management:

Barrier plus sequestration: Physical encapsulation, which is less susceptible to flex-induced cracks, but provides reduced ingress [8]. Incorporating chemical capture into encapsulants is an example; the supramolecular chelators that capture Pb²⁺ [12] provide a fail-safe if the barrier should fail. The efficiency of sequestration and lifetime needs to be quantified as proposed by commercial safe devices [10] and tested after mechanical fatigue, and not only in pristine devices. End-of-life recovery protocols that function in water [13] set constraints on adhesive and barrier chemistries to allow low-temperature delamination of flexible tandems with mixed Pb/Sn chemistries [7,13].

Perovskite sites may be outcompeted by surface-anchored antioxidants and zwitterions at NBG interfaces [19–20] that can act as internal redox buffers to stabilize Sn(II) and prevent its oxidation. The addition of redox-reservoir nanocomposites containing controlled amounts of dopants, such as doped carbon nitrides [75], could further scavenge ROS and block the movement of halide vacancies, if there are no optical losses and stiffening effects. Scavengers used in electrochemical and photooxidative systems [66–71] are used to inform the selection of scavengers that are inert under illumination and bias.

Thin thick layers (hole/electron contacts) with large band offsets reduce the ion accumulation at interfaces (interlayers called “ion-blocking”). Flexible tandems have been proven with PEIE-seeded AZO layers [1] and molecule-bridged NiO [2]. Self-healing elastomeric interphases can relieve the residual strain and heal the micro-cracks, thus reducing the rate of ion migration along stress concentrators [6]. The graded-modulus encapsulation scheme, which places the neutral axis in the middle of the brittle recombination/contact stack reduces tensile strain and delays crack

initiation [5,44].

Wetting and Orthogonality: Less susceptible to moisture pooling and oxygen adsorption from wetting during fabrication or use of adduct/surfactant chemistries, give void-free buried interfaces in PEDOT:PSS [16] and other adduct/surfactant chemistries [64–65]. This benefit is both mechanical and chemical: Void-free contacts eliminate stress-enhancing defects.

Other materials that provide cross domain mechanical compliance and metal immobilization are considered as design inspirations rather than device claims: chitosan-based composites for combined mechanical compliance and metal immobilization [77]; oxide surface-charge tuning to adjust adsorption selectivity [52]; layered MXene barriers [47]; and synthesis-controlled hybrid particle interfaces that tune stability [57].

Why single-stressor success is insufficient

Across classes, evidence indicates that tactics that work well when used with one stressor may not be as effective when they are used with more than one stressor:

Cation engineering and surface passivation are means to reduce WBG halide segregation, but this may be restored by bending-induced strains, particularly where wetting has been marginal [4,16,20].

The stabilization of NBG Sn–Pb interfaces from oxidation provided by the ink chemistry and antioxidants [17–20] can be compromised by an increase of oxygen ingress into the interface during cyclic bending or by stiffening of the stack on the sharp modulus transitions [3–5].

For encapsulation that can be passed in static damp-heat testing, built-in sequestration and barrier redundancy are required for flex-fatigue failure by microcracks [8–10,44]. Multi-stressor testing combining cyclic mechanical loading, controlled humidity/oxygen, temperature cycling, light at MPP and clear failure criteria in consensus stability protocols [14,32–33] and multiscale mechanical frameworks [5] all call for multi-stressor testing. Most flexible tandem studies reported direct, mechanical and thermal cycling [2,15] but no harmonized ISOS like standard for mechanics has yet appeared. Furthermore, sustainability constraints demand that lead-sequestration efficiency and post-mechanical damage lifetime be measured [8,10–12] and reliability, remanufacturability and solvents be taken into account during design decisions [9,11,31]. Surrogate-based control strategies for complex treatment systems [89] and data-driven modeling from photocatalysis [84] point the way towards identifying combinations of stressors and provide a means to monitor proxies for buried-interface degradation during treatment.

Flexible all-perovskite tandems, the recombination junction should be a recombination contact, a solvent/process barrier, an ion blocker, and a mechanical dissipater; both proven examples of ALD oxides seeded by PEs and molecule-bridged NiO exhibit these properties when bent [1–2,15].

The controlled surface composition of the WBG hole interfaces (e.g., PEDOT:PSS segregation vertically) as well as molecule-anchored oxides that cooptimize selectivity, wetting and adhesion [2,16] are benefits of using WBG hole interfaces.

Flexible-specific oxygen ingress and deformation need re-optimization, rigid-device chemistries (comproportionation, antioxidants) translate partially, NBG Sn–Pb buried

interfaces require integrated redox control, ion blocking, and flaw blunting [17–20,39–40].

The performance upon flex-fatigue is the relevant metric for encapsulations, which can be considered an active interface that positions the neutral axis, provides multilayer tortuosity and embeds chemical sequestration [7-12,44].

Under coupled stressors, single-stressor gains are unreliable, and it remains an open need to have standardized multi-stressor protocols and operando measurements at buried interfaces [3–5,32–33,34].

There is no uniformity in mechanical figures of merit reported in different studies, bending radius without strain, thickness, mode and operating condition is not adequate [5,43]. Fracture analysis can be made more complex by reports of mixing of concave/convex cycles [4].

Sometimes the property of mechanical enhancement is claimed, but the fracture-energy or adhesion properties aren't measured, stiffness increases can help to reduce the onset of cracks, but they can increase delamination at interfaces with low adhesion [3–5,43].

Cross Domain Analogies: membranes, photocatalysis, batteries and heat transfer provide similar mechanisms but are not necessarily interchangeable for the perovskite case. Optical loss, interfacial dipoles, and chemical stability over time need to be evaluated for the efficiency of DES/tandems, chitosan-based scavengers, or MXene barriers [47,74,77].

Several other studies provided are used to explain evidence-transfer limits. Ligand identity and the pore environment, surface coordination, and nucleation route control uptake, charge transfer and chemical selectivity, with examples of smart adsorbent frameworks, metal–organic-framework hybrids and bio-assisted encapsulation of nanoparticles [90–92]. Although these principles encourage screening modifiers of under-coordinated Pb/Sn and mobile halides, they do not define the PV compatibility. Substituted ceramics and nonlinear optical polymer composites also provide characterization axes—permittivity, dielectric loss, field concentration, and thermal response—but do not provide certain solar-cell additives [93-94]. Apparent removal or oxidation performance may change with oxidant dose, pH, iron chemistry, aeration, mass transfer and secondary product formation [95-99] and so should be tested after fatigue for capacity, selectivity, regeneration and by-product formation in redox buffers and capture layers of flexible modules. The selective behavior can also be compromised due to swelling, fouling or transport resistance, as shown in membrane studies, nanofluidic studies, and desalination studies [79,100–102]. They are thus listed here as examples of experimental design, rather than direct evidence of photovoltaics.

In general, the stability of flexible all-perovskite tandem is understood to emerge from co-optimizing electronic selectivity, strain dissipation, and chemical buffering at each buried interface, in the IMCC framework. Meanwhile, the data to date are representative of feasible pathways, particularly at the recombination junction and hole-selective contacts, but also highlight the need for simultaneous stressor testing, operando buried-interface diagnostics, and dose–structure–function rules for multifunctional nanocomposites to transition promising laboratory strategies to manufacturable, circular, and reliable flexible modules.

Table 3 presents a mapping of stability interventions to the stressors and signatures to be controlled. The comparison shows that the effect of the four external influences such as heat, light, redox reaction and bending cannot be considered individually since each can drive the migration of ions and cause failure of barriers or crack growth [1-10,14-20,29-33,39-40].

Table 3. Stability strategies mapped to stressors and diagnostic signatures

Stressor	Dominant coupled pathway in flexible tandems	Observable signatures	Promising countermeasure	Unresolved issue
Light	WBG halide segregation; trap-assisted redox; photothermal strain	PL red-shift/splitting, voltage loss, local EL darkening	Composition control, defect passivation, redox buffers	Stability under simultaneous strain and MPP bias
Heat	Volatile loss, diffusion, interfacial reaction, thermal-expansion mismatch	Phase change, rising resistance, delamination, accelerated Sn(IV)	Thermally stable contacts, cross-linking, heat spreaders, compliant barriers	Additive migration and adhesion after repeated thermal cycles
Humidity	Hydrate formation and rapid decomposition at cracks/edges	PbI ₂ formation, yellowing, leakage-current change	Multilayer barriers, robust edge seal, hydrophobic interface	Barrier fatigue and edge pumping during flexing
Oxygen	Superoxide chemistry and Sn(II) oxidation	Increased p-type self-doping, trap density, Voc loss	Antioxidants, oxygen barriers, redox-stable contacts	Quantifying oxygen flux through deformed laminates
Electrical bias	Ion drift, interfacial accumulation, electrode reaction	Hysteresis, transient Voc/FF loss, impedance changes	Ion-blocking layers, low-defect absorbers, stable electrodes	Reverse-bias and partial-shading behavior in flexible

Cyclic bending	Cracking, delamination, TCO failure, crack-assisted ingress	Sheet-resistance rise, EL discontinuities, PCE decay	Neutral-axis design, ductile electrodes, adhesive/passivating interfaces	modules Comparable strain-based protocols and failure statistics
Coupled operation	Positive feedback among crack growth, ingress, ion migration, heat, and redox	Nonlinear degradation faster than sum of isolated tests	IMCC co-design plus operando diagnostics	No accepted flexible-PSC multi-stressor standard [4–5,15,32]

Source: Authors' synthesis of flexible tandem, stability, safety, and standardization literature [1–10,14–20,29–33,39–40].

Advanced nanocomposites: a function-first taxonomy

A function-first taxonomy for nanocomposites in flexible all-perovskite photovoltaics is summarized in Fig. 4. These materials are not categorized according to chemistry, but rather by the type of interfacial failure they are designed to prevent, and the performance limitations they are intended to address and overcome: (i) materials that create conductive percolation networks for providing sheet conductance and optical transmission without brittle failure; (ii) defect-passivating nano-objects and hybrid interphases, which reduce recombination, suppress ion migration, and anchor adjoining layers; (iii) stress-dissipating and self-healing polymer-perovskite systems that mitigate crack initiation and propagation while preserving carrier transport; and (iv) barrier, lead-capture, optical, and thermal nanocomposites, which provide slow ingress/egress of reactive species, immobilization of Pb/Sn upon damage, and stress management of light and heat. This project is explicitly designed through the lens of the Interface–Mechanics–Chemistry Coupling (IMCC) perspective, which considers the impact of any additive or nanostructure not only as an electronic function, but also as a strain distribution, adhesion and residual stress modifier, and chemical equilibrium and transport path modifier under operational stressors. [3–6,12,28,42–47,58–62,85,90,103–115]

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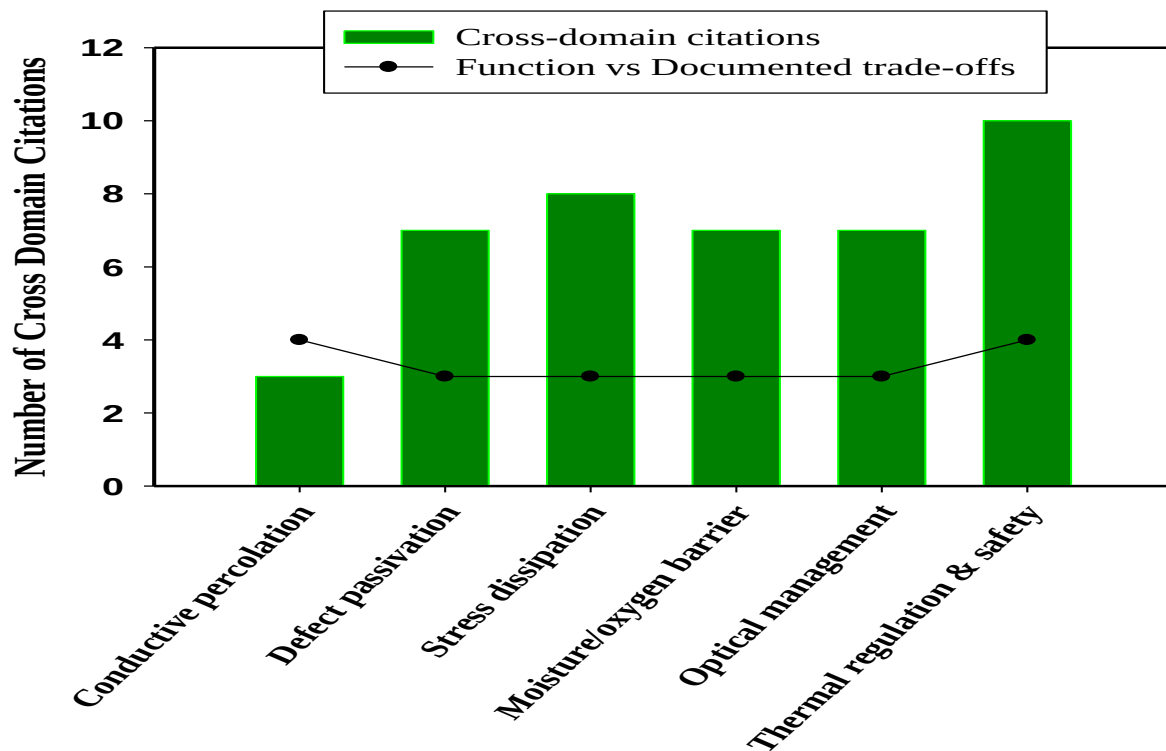
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Figure 4. Function-first nanocomposite taxonomy: breadth of cross-domain evidence versus documented trade-offs. Bars show cross-domain citations for conductive percolation [22,24–25], defect passivation [27,53–56,76,83], stress dissipation [50,106–107,109,113,116,122–123], moisture/oxygen barrier [47–49,51–52,103,115], optical management [22–23,25–27,63,94], and thermal regulation and safety [12,28,58,60–61,85,90,92,104–105]; the connected markers show documented trade-offs for each function.

Important to have device classes differentiated within this map. Functional nanocomposite and/or interfacial concepts have been directly demonstrated in flexible all-perovskite tandems in a number of key studies [1–2,15–16]. Flexible single-junction perovskite cells are also used to develop complementary evidence [4,6,44–45,62,86] and the corresponding rigid all-perovskite evidence yields constraints on the defect passivation and redox stabilization on mechanically more forgiving substrates [17–20]. Carefully restricted analogies are also made with polymer composite mechanics, membranes, sorbents and data-driven optimization to give rules for loadingdispersionfunction relationships that are parameterizable [103–134]. Analogies are not considered as a direct demonstration of the photovoltaic properties of a device, but as a methodological precedent and/or a design heuristic that can be transferred.

Conductive percolation networks

The primary challenge for flexible all-perovskite stacks is the ability to achieve transparent, conductive and fatigue-resistant electrodes. Under bending, Ultra-thin metal, oxide and hybrid networks should also not produce shunts or optical penalties

due to cracks. There are two opposing points of view that define the design space. The first one is flexible all-perovskite tandems, where Palmstrom et al. used a 1 nm layer of PEIE as a nucleation layer for conformal low-temperature ALD AZO to form a recombination junction, which also acted as a barrier and maintained lateral conductivity while preventing solvent damage during top-cell coating [1]. Second, a packaging-oriented approach is to combine transparent electrode and barrier structures into a single flexible film, which eliminates the mechanical mismatch between the transparent electrode and the barrier and simplifies the process of laminating the barrier. Han et al. have reported such a combination of transparent electrode and barrier for flexible perovskite single-junction devices [44]. Although it is not evidence of all-perovskite tandem, it does highlight the importance of co-designing the conduction and environmental isolation in one mechanically coherent layer.

From a function-first perspective, the problem of percolation can be considered as a tunable microstructure problem and mechanics. It has been well established in classic polymer composite studies that filler volume fraction, aspect ratio, and interfacial adhesion all co-determine the modulus, thermal expansion and viscoelasticity, hence the neutral axis and fracture behavior of the composite electrode/encapsulant stack [106–107,109–110,112–113]. For instance, the storage modulus of syndiotactic polypropylene increases with iron loading or clay content, and the thermal response is also changed, but the melt viscosity is increased and the crystallization kinetics is altered with increasing iron loading or clay content, thus reducing the processing window [106–107,109–113]. In the case of nanowire or flake-electrode inks applied onto perovskites that are flexible, these inks should be loaded close to, but not above, the electrical percolation threshold, to prevent excessive stiffening leading to interfacial stresses under bending. Cellulosic fibres decorated with TiO₂ showed enhancements in toughness and biological properties of dental adhesives without sacrificing the polymerization or water uptake [108] and cellulosic or polymer assisted metal nanowire networks can be designed for fracture energy, albeit at the expense of increased moisture sorption (which causes dielectric loss or corrosion in PV stacks analogy) [108]. The literature on membrane engineering provides a formalization of the trade-off between transport and structure through permeability–selectivity curves [115] and replacing them by conductivity–transparency curves gives a directly transferable objective space for designing flexible percolation networks (analogy) [115]. However, as summarized in Table 4, percolation composite is not a “mere conductor”: its modulus, thickness and adhesion redistribute strain across the active device during cyclic bending, which in turn impacts interfacial fracture and electrochemical stability [5,43].

Defect-passivating nano-objects and hybrid interfaces

For thin stacks, aimed at flexibility, and processed at low temperature, defect passivation and interfacial selectivity become more important. In this case, a rigid and a flexible evidence of perovskite is complimentary and needs to be differentiated.

Antioxidants, surface passivants and compositional control have been helpful for mixed-cation wide-bandgap (WBG) and Sn–Pb narrow-bandgap (NBG) subcells. The use of antioxidants, surface passivants, and compositional control for mixed-cation

wide-bandgap (WBG) and Sn–Pb narrow-bandgap (NBG) subcells has been beneficial. A better NBG films and efficient tandems on rigid substrates were achieved by using oxidation control of Sn(II) by comproportionation chemistry. [17] Bulk and interface control of Sn–Pb perovskites led to very long carrier lifetimes [18]. The nonradiative losses were reduced by a surface anchoring zwitterionic antioxidant [19] and by grain surface passivation [20]. These works indicate the presence of chemical motifs, namely zwitterions, Lewis bases, and redox-ready ligands, that are excellent candidates for flexible stacks, but they did not assess their mechanical strength.

Flexible single-junction evidence: The in situ formation of a low-dimensional capping phase closed surface flaws in a 3D perovskite and measurably enhanced not only the PCE, but also fracture resistance under tension-only bending ($T_{80} > 20,000$ cycles) and clarified how applied and residual stresses couple to fracture resistance [4]. Zwitterionic and hydrogen-bonding elastomeric matrices provided self-healing and residual-strain relief, with an encapsulation-compatible healing temperature of 60 °C, and PCEs were >90% following 8,000 bends. Conjugated small molecules have also been introduced as molecular additives to enhance efficiency and mechanical strength in both flexible single-junction cells and minimodules, with recent examples including the use of entinostat to achieve both of these effects in the perovskite single-junction devices [45]. Other chemical treatments have been applied to stabilize NBG films on flexible substrates such as motifs derived from SnF₂ and N,N'-carbonyldiimidazole, which suggests that the judicious choice of small-molecule modifiers can impart passive and cohesive properties. Other chemical treatments have been employed to anchor NBG films to flexible substrates, such as SnF₂-derived motifs and N,N'-carbonyldiimidazole, which supports the premise that the judicious use of small molecule modifiers can establish passive and cohesive properties. These are single-junction demonstrations; do not use as a measure of tandem performance.

Flexible all-perovskite tandem evidence: A molecule-bridged hole-selective contact, combined with carbazole/phosphonic-acid interlayers and low-temperature NiO nanocrystals to minimize recombination, adhesion and improve PCE, holds record of 24.7% PCE (24.4% certified) in flexible tandems with durability to 10,000 bends at 15 mm radius [2]. Using Triton X-100 tuned vertical phase segregation in combination with reducing surface PSS content, increasing wettability and interfacial dipoles, and ultimately increasing NBG single-junction PCE to 21.5%, flexible tandems based on this engineering reached 25.4%, and a flexible mini-module achieved 19.7% [16]. The results confirm that the nanoscale phase distribution in polymeric contacts is an interfacial–mechanical lever; compositional gradients control the charge extraction, the wetting of the solvent and provide greater adhesion and processing compatibility for flexible tandems.

In the framework of IMCC, it is important to note that interfacial chemistry that passivates defects can also cause the interface to be stiffer or to create residual stress. By creating interpenetrating interfaces, the partial diffusion or chemical bonding across the interface led to improved fracture resistance and operational stability of the perovskite cell (rigid study) [46]. Contacts of this type, which are able to address electronic selectivity, chemical stability and mechanical reliability, are classified as hybrid interfaces in the taxonomy of Figure 4, because the fracture energy is

important, not only adhesion strength [5].

Stress-dissipating and self-healing polymer–perovskite systems

Mechanically dissipative, optically compatible, and, importantly, ionically benign nanocomposite polymers provide a direct path to changing the strain–damage landscape in flexible stacks. There are three kinds of evidence out there that can define credible strategies.

Self-healing and release of residual strain have been achieved in flexible single-junction demonstrations that demonstrate cross-linkable zwitterionic elastomers that regulate crystallization and contain dynamic electrostatic/hydrogen-bond networks that facilitate charge transport [6]. Built-in reversible bonding is activated by mechanical retention (>90% after thousands of cycles) and thermal self-repair at 60 °C, which are typical lamination or mild-heating conditions employed in lamination or encapsulation. The use of entinostat-type molecules for enhancing PCE in flexible cells and minimodules, combined with the increase of fracture resistance, suggests that some small molecules are also used as interfacial tie-coats as well as mechanical tougheners, when co-cross-linked or physically interpenetrated with the adjoining layers [45]. In polymer-perovskite composite membranes for generic optoelectronics, fatigue was suppressed by a combination of mechanical gradient stratification (analogy) [86] that may be extended to photovoltaics, but which did not investigate specific PV lifetimes.

Beyond their passivating effect, there are two additional stress-dissipating interfaces: the molecule-bridged NiO contact [2] and phase engineering of PEDOT:PSS [16] that reduce interfacial defects, typically acting as crack initiation sites and enhance adhesion and solubility with solvents to minimize residual stress during low-temperature deposition. The high PCE retention of the modules after 10,000 bends at 10 mm radius (97.2%) was consistent with low trap density and void-free buried interfaces in the large-area gas-quenched WBG films on PET, showing that defect-mediated crack nucleation was reduced in the WBG top cell [15].

The dental and biomedical polymer nanocomposites serve as warning examples: The filler material for the composites can have antimicrobial or mechanical properties that affect water sorption and glass-transition temperature in a way that is coupled with changes in fracture energy [108,116–117]. For instance, nystatin coated CuO nanoparticles had enhanced antifungal activity and modulated the mechanical/thermal properties of PMMA [116] and chlorhexidine coated cotton fillers had enhanced antibacterial activity but altered water uptake and mechanical properties of resin-based sealants [117]. Not photovoltaic studies, but a rule of IMCC: chemical functionalities introduced in order to achieve one target (e.g., antimicrobial, ionic capture) will inevitably affect the viscoelastic response and moisture interactions across the interface, which must be quantified for photovoltaic use (analogy) [108,116–117]. Likewise, the effects of shear-induced crystallization on the viscoelastic spectra of bio-based or inorganic fillers are transferred to blade/slot-die processing of flexible perovskites (analogy) [110–112] are shown in a series of composite studies based on sPP.

Barrier, lead-capture, optical and thermal nanocomposites

Primary differences between deployable flexible tandems and non-deployable flexible tandems are environmental isolation and damage tolerance. The recombination layer can also be used as a barrier: In flexible all-perovskite tandems, AZO grown by low-temperature ALD on top of C60 created a barrier for processing, which decreased the water vapour transmission by approximately one order of magnitude and allowed the use of non-orthogonal solvents during top-cell deposition [1]. Subsequently, Li et al. treated the buried interface with both molecular adhesion promotion and low-temperature NiO, which was implied to further tighten the interface and reduce the electronic and chemical degradation [2]. At the module level, ambient-processed WBG layers and flexible encapsulation was achieved with high efficiencies and mechanical endurance of 10,000 bends and was able to handle thermal cycling and continuous one sun illumination [15]. These studies did not report on damaged module leakage of lead, but exemplify a consistent integration of barrier functions into electronically functional layers, which is also consistent with the IMCC.

The single-junction evidence shows that encapsulation can be reconsidered as a barrier nanocomposite material for managing heat and ions. The evidence shows that for the single junction, it is possible to re-conceptualize encapsulation as a barrier nanocomposite material for heat and ion management. Increasing the thermal diffusivity and reducing the leakage of lead was achieved by designing environmentally friendly materials, showing that thermal transport modifiers can be combined with lead-immobilizing chemistries, respectively, by Yang et al. [62]. The use of transparent electrode integrated barrier films (EEB) allowed easy encapsulation of flexible single junction cells and decreased the risk of failure in the barrier at the vulnerable electrode edges [44]. More fundamentally, reviews categorize lead-risk mitigation as encapsulation, chemical capture, alternative absorbers, and recycling, and highlight the importance of co-designing the chemical capture with mechanical barrier integrity because flexing can cause cracks that null the effect of mechanical barrier alone [8]. The concept of using built-in supramolecular cross-linkers to immobilize lead through chelation has been demonstrated to be a mechanistically appealing concept for flexible tandem encapsulators, in which reversible cross-links can also act as mechanical dissipators [12]. The recycling advances show high yield of Pb recovery and reuse of transparent conductor for rigid modules [7] and the development of aqueous-based recovery with device performance like fresh materials [13]. These are rigid and/or generic perovskite precedents, not flexible tandem demonstrations, and to make perovskite a viable flexible tandem stack barrier/adhesive chemistries must survive mechanical delamination and be compatible with both WBG and NBG streams [7,13].

The sorbent and membrane analogies are also useful in designing barrier and capture nanocomposites. Clay and biomass-derived sorbents have different selectivity and capacity for metal ions in water, and emulsion liquid membranes can be used to selectively extract specific ions like chromium from water with high efficiency under controlled interfacial conditions [103-104]. Based on the above studies, polymer-ligand-filler composites can be engineered to have a specific affinity and selectivity for Pb²⁺ and to be regenerated (analogy) [103-105]. Process-intensified separators like lamella settlers used for water re-use are examples of units used to manufacture

wastewater management for roll-to-roll lines in analogy to the use of capillary driven modules for the capture of lead [118]. On the mass-transport side, the trade-offs between selectivity and permeability in direct-air-capture membranes and reactive membranes are similar: as selectivity is increased, the permeability is decreased, leading to high levels of humidity/oxygen back pressure in the membrane during thermal cycling (analogy) [114–115]. With the exception of the thermal conductivity, which increases the rate of heat flow to reactive interfaces with insufficient chemical stabilization, the barrier metrics and IMCC couplings are important and must be considered in the screening of optical and thermal nanocomposites; too high a thermal conductivity can lead to increased heat flow to reactive interfaces without proper chemical stabilization, while too low a thermal conductivity may worsen local hot spots under partial shading [62]. These functions and couplings are listed in order in Table 4.

Loading–dispersion–function trade-offs

Filler can be beneficial or detrimental, depending on the loading, dispersion, and interfacial chemistry. As a general guideline, loading curves should be plotted at least on four axes: electronic benefit, mechanical change, chemical reactivity, and processability. Polymer composite rheology gives usable quantitative logic: Add nonlinearly and adjust the shear-thinning behavior of the polymer with clay; shear flow during coating alters crystallization pathways, and final microstructure (analogy) [111–112]. Bio-based fillers are starch, which lower density and stiffness and increase damping, with clear viscoelastic fingerprints (analogy) [110]. The presence of iron-containing fillers increases the thermal conductivity and affects the crystallization of sPP; maps of properties are sensitive to the dispersion and the number of nucleation sites (analogy) [106–107,109]. In dental materials, nanoparticulate additives co-determine antibacterial activity, toughness, and water sorption, and that makes us aware that polar functionalities can augment water uptake at the interfaces (analogy) [108,116–117] and, for PV, the risk of hydrolytic degradation.

In flexible tandems, the number of variables involved in this design space is high, and data is sparse, so uncertainty-aware, physics-informed models are required to prevent overfitting. For thermal and separation processes, there has been an improvement in process set-point selection with the use of grey-box models that incorporate both mechanistic drying/crystallization descriptors and data-driven correction (analogy) [126–127]. Reduced-input strategies retain predictive power at the cost of reduced sensor burden or reduced data-collection cost (analogy) [121]. Optimization of exergy and emissions in complex plants has been achieved by surrogate modeling under uncertainty, and the selection of descriptors for pyrolysis (analogy) [128–131]. The same toolchains can be customized for optimizing nanocomposite loading and processing for flexible tandems that explicitly quantify uncertainty [120,128–132]. As a complementary materials design analogy, machine learning on TiO₂ band gaps demonstrates the mapping of structural descriptors to electronic properties [134], paving the way for composition–defect–bandgap mapping in perovskites where NBG subcells are meant to inhibit Sn(II) oxidation and WBG subcells to prevent phase segregation from light exposure [1,17–20]. Table 5 represents a minimal dataset and meta-data schema to report with composition and loading, so that it can be re-used in

such models.

Table 4 is an organization of advanced nanocomposites that categorize them based on their function, not composition. It exposes recurring loading trade-offs, for instance, higher conductivity, capture capacity, or toughness can come at the expense of optical loss, aggregation, excess stiffness, moisture uptake, and new chemical reactivity, as demonstrated by dose–structure–function relationships, which need to be measured explicitly [3–6,12,28,42–47,58–62,85,90,103–115].

Table 4. Function-first taxonomy of advanced nanocomposites

Function	Candidate nanocomposite class	Desired microstructure	Benefit	Dominant trade-off	Evidence-transfer note
Conductive percolation	Ag nanowire/polymer, CNT/polymer, graphene/PE DOT:PSS	Continuous conducting network below roughness/shunt threshold	Flexible transparent electrode or conductive interlayer	Haze, junction resistance, metal migration, roughness	Percolation must be validated in the actual photovoltaic stack; optical/plasmonic precedents include [22,24–25]
Defect passivation	Metal-oxide nanoparticles, 2D sheets, functional polymers, ionic liquids	Uniform low loading at grain boundaries or buried interfaces	Lower traps and non-radiative recombination	Insulating excess, aggregation, altered crystallization	Cross-domain nanoparticle and ionic-liquid studies [27,53–56,76,83] motivate controlled loading but do not prove PV efficacy
Stress dissipation	Elastomer/perovskite, polymer-	Soft continuous or	Toughness, adhesion	Carrier dilution,	Polymer-composite

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	tethered filler, nanocellulose hybrid	interpenetrating phase without breaking carrier pathways	n, fatigue resistance, self-healing	reduce d crystallinity, creep	mechanics and rheology in [106–107,109,116]–[50,113,122–123] are transferable design precedents
Moisture/oxygen barrier	Nanoclay/polymer, silica/polymer, oxide/polymer multilayer, MXene composite	Tortuous diffusion path with low defect density	Lower gas transmission and slower chemical attack	Brittle filler-rich regions, interfacial voids, conductivity-induced reactions	Membrane/barrier analogies [47–49,51–52,103,115] require flexible-PV validation
Optical management	Plasmonic metal structures, dielectric nanoparticles, textured polymer composites	Controlled size, spacing, and refractive-index contrast	Scattering, light trapping, spectral management	Parasitic absorption, local heating, recombination at exposed particles	Optical and plasmonic studies [22–23,94]–[25–27,63] provide design concepts
Thermal regulation and safety	Graphene-oxide polymer, thermally	In-plane heat path plus	Lower hot-spot tempera	Added mass, opacity,	Thermal and adsorption

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conductive	chemica	ture,	phase-	precedent
filler, phase-	lly	reduced	change	s
change	stable	thermal	leakag	[58,90,92
composite,	capture	stress,	e,	,104–
lead-	sites	lead	finite	105]–
chelating		immobi	sorptio	[12,28,60
network		lization	n	–61,85]
			capacit	motivate
			y	coupled
				safety
				testing

Source: Authors' synthesis of nanocomposite, membrane, barrier, transport, and flexible-photovoltaic evidence [3–6,12,28,42–47,58–62,85,90,103–115].

Mechanical reliability and operando characterization

Mechanically coupled stacks: flexible all-perovskite tandems, fracturing and delamination pathways are same as electronic selectivity and chemical potential gradients. Figure 5 presents the last few decades of literature in a directly interpretable statistical benchmark. As seen in Panel A, the reported efficiency of flexible tandem cells has been improved from 21.3% in 2019 to 27.5% in 2025, while the demonstrated efficiencies of flexible modules have remained at around 19.7–23.0%, demonstrating the ongoing cell-to-module translation loss [1–2,15–16]. Similarly, Panel B highlights another important evidence gap: only 2 of the 4 benchmark studies report directly comparable 10,000-cycle bending results, which are that performance was maintained at a 15 mm radius in 2022 (2) and that 97.2% of the initial performance was maintained at a 10 mm radius in 2025 (15). The missing value for any row is indicated as “NR” not zero. The comparison thus reinforces the minimum reporting protocol suggested in Table 5, and shows the shortcomings of efficiency records as a measure of mechanical readiness.

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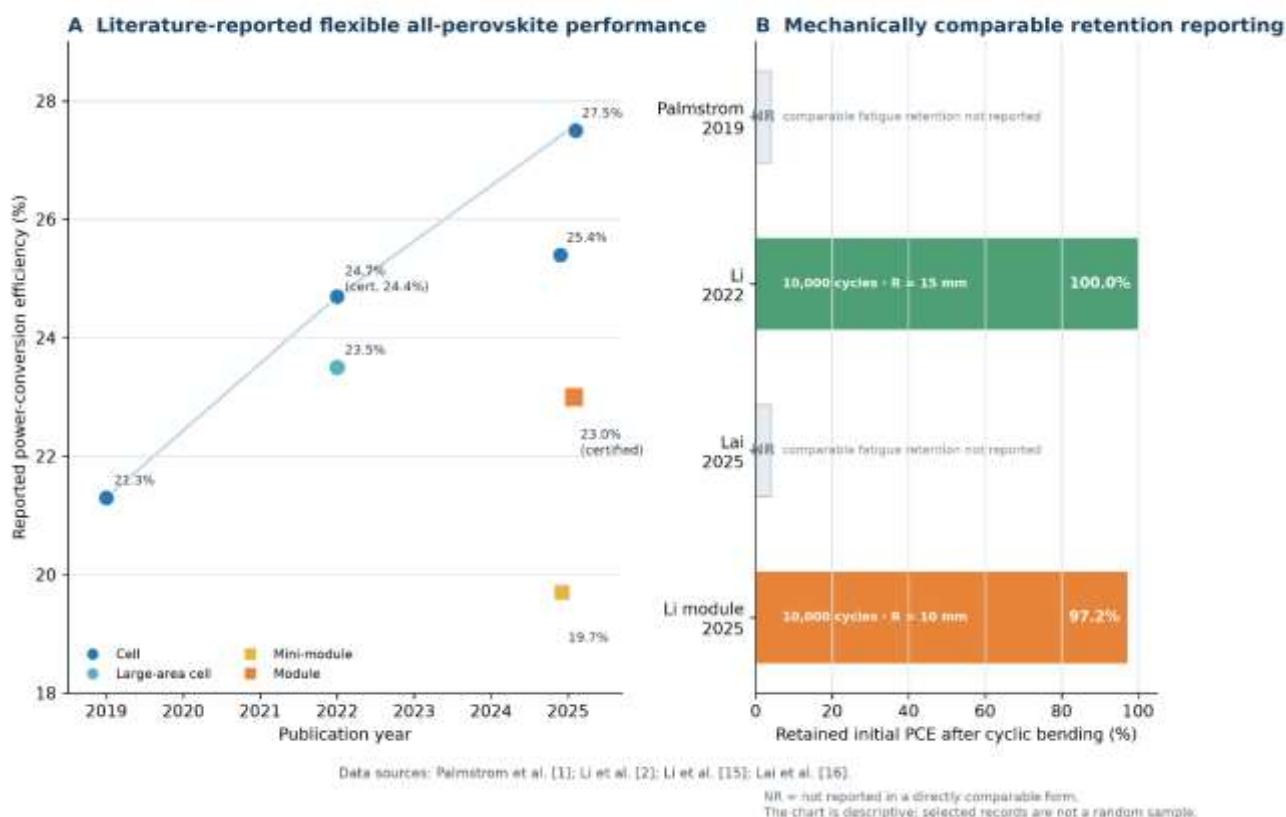
<http://amresearchreview.com/index.php/Journal/about>**Flexible all-perovskite tandem benchmarks: efficiency, scale, and bending durability**

Figure 5. Literature-derived statistical benchmark of flexible all-perovskite tandem efficiency, device scale, and bending durability. Panel A compares reported cell and module power-conversion efficiencies, while Panel B distinguishes directly comparable 10,000-cycle retention results from studies that did not report a comparable fatigue metric.

Strain, neutral axis, and residual stress

The bending strain at the surface of the device is approximately $\epsilon = h/(2R)$ for a single layer, and for multilayers the bending strain at each interface is dependent on the position of the neutral axis based on the stiffness and thickness of each layer [4–5]. Two consequences follow. First, a radius of reporting alone is not enough: the thickness of the stack and the layer-wise moduli are required to estimate actual interfacial strain [5,43]. Second, the vectorial sum of residual stress due to deposition, drying and thermal mismatch induces a bias in fractures toward the interfaces with the least adhesion or lowest fracture energy [4–5]. Therefore, co-optimization of energy dissipation across the bulk, interfaces and device layout is essential; introducing the neutral axis in the most brittle or defect prone region is a foretold failure accelerator [5].

A flexible Tandem using all perovskites provides solid evidence to support reasonable targets. In order to maintain initial performance after 10,000 bends at 15 mm radius, Li et al. used a molecule-bridged NiO contact that increased adhesion and decreased

the interfacial recombination [2]. Large-area flexible tandem modules maintained 97.2% of their initial PCE after 10,000 bends with a radius of 10 mm and were subjected to testing between -40 and 85 °C thermal cycling and continuous one-sun illumination [15]. Palmstrom et al. previously showed flexible tandem PCE of 21.3% that correlated to an ALD-AZO recombination barrier, while also enhancing the solvent compatibility and decreasing water-vapor transmission [1]. Although there are differences in the layer configuration and thickness of these studies, and they are not complete stress histories, they suggest that the mechanical strength of a buried interface with a good adhesion and controllable residual stress is as important as the absolute tensile strength for any single layer.

Flexible single-junction research provides greater insight into the mechanics. Surface flaws were reduced by a low-dimensional perovskite capping, which was also found to increase the fracture threshold of tension-only bending, thereby separating out all the mechanisms for crack initiation from complex alternating-bend artefacts, and suggesting tension-only bending protocols for interpretability [4]. Zwitterionic elastomeric matrices relieved the residual tensile strain, yet provided efficiency [6]. The PEDOT:PSS vertical composition was engineered to introduce a tuned wettability-induced stress after the coating, which enhanced the reproducibility [16]. All of these are examples of IMCC coupling: Wetting/electronic passivation changes can also affect neutral axis location, residual stress and fatigue.

Fracture, delamination and fatigue protocols

The lack of an accepted ISOS-equivalent for mechanical testing is explicitly mentioned by Wang et al. and they suggest that the strain should be reported instead of the radius only and that the grain boundaries, the interfacial fracture energy, the reversible bonds and the design of the neutral-axis should be treated as a connected system [5]. To avoid ambiguous failure signatures, Dong et al. suggest performing tension-only cyclic tests, while also warning about the mixing of cracking, peeling and apparent closing of cracks when using convex/concave bending [4]. For mechanical protocols, within the IMCC framework, they should be combined with electrical bias and environmental controls: for instance, cyclic bending at a specified strain under 1 sun illumination at maximum power point (MPP) and under controlled humidity/oxygen to capture coupled mechanochemical failure. The definitions that include mechanical dimensions and failure criteria are explicitly missing from the ISOS procedures for light/thermal/humidity stress, but they are provided as a starting scaffold [14]. A word of caution for rigid-substrate studies that relate accelerated tests to outdoor performance: there may be a dependency on the sequence or combination of stressors for failure modes and rate [32, 33]. Therefore, we recommend:

Report: substrate/stack thickness; layer sequence; moduli (measured or literature value); calculated maximum interfacial strain and neutral-axis position; bending mode (tension-only vs alternating), speed, dwell, and cycle number; environment (temperature, humidity, oxygen), and bias condition (open circuit, MPP).

Set failure criteria prior to testing: T90 or T80 under user-defined conditions; Failure criterion based on crack density by optical/electrical mapping; Failure criterion based on delamination length by adhesion testing/microscopy.

Make distinction between damage regimes: elastic recoverable; stable damage (no new cracks per cycle); unstable damage (accelerating crack density); and catastrophic failure; correlate electrical signature (e.g., series resistance rise vs shunting) and separate IMCC couplings.

Operando electrical, optical, structural, and chemical diagnostics

The flexible tandems under-measure coupled mechanochemical degradation. Causality will be possible by conducting operando diagnostics that probe the electronic performance, structural evolution, and chemical state simultaneously under a controlled strain. Practical approaches include:

Distributed resistance and photovoltage mapping during bending cycles, to determine the contact degradation from absorber cracking and MPP tracking during cyclic bending to capture functional fatigue [2,4–5,15].

Time-resolved PL and EL under strain to identify nonradiative recombination centers caused by microcracks or ion migration and correlation with the passivation strategies from rigid studies [19–20].

In situ curvature-controlled X-ray scattering for monitoring the grain reorientation, microstrain, and crack initiation, and link to low dimensional capping effectiveness in flaw sealing [4]. In the case of polymeric contacts, a depth-sensitive spectroscopy has been performed to ensure the tuned vertical composition is maintained after bending [16].

Operando XPS or NEXAFS to track oxidation states (e.g., Sn(II)/Sn(IV)), interfacial redox of transport layers, and chemisorption of ambient species, based on rigid NBG stabilization strategies [17–20]. Use of capture layers or microextraction cells for post mortem lead mobility mapping under defined damage, to determine sequestration efficiency with and without cracks as per lead management frameworks [8,10,12].

Although deployments of all sizes are relatively rare, there have been recent flexible tandem modules that have integrated mechanical and thermal cycling and illumination [15]. Equivalent fatigue-retention data, however, do not exist for several other high-efficiency reports [1,16] as is revealed by Figure 5B. Before one can compare efficiency and durability between laboratories, however, standardized multi-stressor workflows are needed.

A minimum comparable reporting protocol

We suggest a minimum reporting protocol (Table 5) which is compatible with ISOS and extended for mechanics and IMCC in order to facilitate comparability and speed up data-driven learning:

Device class: explicitly differentiate between flexible all-perovskite tandem, flexible single-junction and rigid all-perovskite; do not mix data across device classes [1–2,4,6,15–20,42–46,62,86].

Stack and mechanics: give the complete layer sequence, thicknesses, moduli or storage moduli at the testing temperature, estimate of neutral axis, calculated interfacial strains, residual stress proxies (such as curvature after deposition), and energy needed for adhesion or fracture (metrics measured if applied) [4–5,43].

Test conditions: bending mode, bending radius, number of cycles, bending rate, dwell time, environment (T, RH, O₂), electrical bias (MPP, open circuit), illumination

spectrum, intensity [2,4,5,14,15]

Failure definition and metrics: PCE retention thresholds (T90/T80), crack-density or delamination-length criteria, and correlated shifts in series/shunt resistances [4-5,14].

Multi-stressor matrix; if single stressor, state and justify; if combined, specify sequence and interactions—learning from lab–field linkage studies [32–33].

Sustainability and safety: If chemicals that capture leads are used, report on the efficiency of the lead capture; if low-toxicity solvents are used, report on solvent metrics; and test the capture after mechanical damage, as per emerging guidance [8–10,12,31].

Table 5 is a minimum comparable protocol derived from the reporting gaps revealed in Figure 5. It demands complete stack geometry, strain instead of only radius, tensile/compressive orientation, cycle profile, coupled environmental conditions, electrical operating point, and the definition of a failure metric to enable mechanical retention to be compared between laboratories [1–5,14–16,31–34,43].

Table 5. Proposed minimum reporting protocol for flexible all-perovskite stability

Reporting block	Required variables	Recommended measurement	Why it matters
Device identity	Full stack, absorber composition/band gap, area, aperture definition, encapsulation, flexible substrate	Cross-section and process record	Prevents mixing incomparable architectures
Initial photovoltaic performance	Forward/reverse J–V, stabilized MPP, EQE-integrated current, statistical distribution	Calibrated simulator and MPP tracking	Distinguishes champion scans from reproducible output
Mechanical geometry	Total and layer thicknesses, moduli where available, bending radius, calculated strain, neutral-axis location	Multilayer strain model, profilometry	Radius alone cannot compare devices of different thickness
Fatigue mode	Convex/concave or tension-only, cycle frequency, dwell, mandrel geometry, sample orientation	Automated controlled-radius bending	Avoids conflating crack opening, peeling, and crack closure
Environment	Temperature, RH,	Controlled chamber	Chemical

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nt	oxygen, illumination spectrum/intensity, electrical bias, preconditioning	with logged traces	kinetics strongly depend on simultaneous conditions
Operando diagnostics	MPP/PCE, sheet resistance, EL/PL mapping, impedance, temperature, curvature	Synchronized electrical/optical/mechanical acquisition	Connects performance loss to mechanism rather than endpoint only
Failure definition	T95/T90/T80, catastrophic failure, area loss, delamination or barrier breach	Survival curves and failure-mode classification	Enables reliability comparison and uncertainty analysis
Statistics and data	Sample count, distribution, exclusions, raw time series, uncertainty	Open metadata and machine-readable files	Supports reproducibility, meta-analysis, and physics-informed ML [32,120–121,127,129–132,135–139]

Source: Authors' proposed reporting protocol based on flexible tandem and stability-standard literature [1–5,14–16,31–34,43].

Scalable low-temperature fabrication and modules

Flexible all-perovskite tandems require process windows which maintain the integrity of the subcell optoelectronics, the integrity of the buried interfaces, and the orthogonality of solvents while operating at temperatures suitable for polymer substrates. This section brings together recent scalable advances and cross domain process control analogies.

Coating, drying and crystallization control

Controlled crystallization at low temperature, in the absence of unstable intermediate states that could lead to the formation of defects or dissolution at the interfaces, is at the core of scalable flexible tandems. A high crystallinity (27.5%) with low trap density and void-free buried interfaces were obtained by in situ additive coating under continuous gas quenching, and a flexible module with an aperture of 20.26 cm² was certified with 23.0%, high crystallinity and 95.8% geometric fill factor, a slot-die

WBG module had an aperture of about 804 cm² [15]. A very rare case of co-optimization of a crystallization strategy, a scalable coating technique and a mechanically compatible flexible substrate. The process window has been rendered polymer substrate compatible by reducing thermal budgets and speeding up solvent removal.

At small scale, low-temperature contact deposition and adhesion of nanocrystals is possible with a molecule-bridged NiO contact, and this results in 24.7% flexible tandem performance with bending durability [2]. Nanoscale composition gradients were optimized using phase engineering of PEDOT:PSS in order to achieve better wettability and solvent tolerance that was especially important for stacked solution processing [16]. The rigid tandems require Sn–Pb inks with stabilization via comproportionation and long carrier lifetimes, which are obtained by defect control, and narrow-bandgap flexible subcells that are processed at reduced temperature without being oxidized by Sn(II) [17–18]. Chemically robust films with controlled grain boundary chemistry can be obtained under flexible constraints as confirmed by flexibly treated single-junction NBG studies with SnF₂ and carbonyldiimidazole (CDI) treated polymer substrates [39–40].

Rheology and crystallization cross-domain analogies can be used to parameterize coating windows. Clay-filled and starch-filled polymers demonstrate that the hydrodynamics of the shear flow affects both the viscosity and nucleation and crystal growth kinetics of the particles (analogy) [110–112], with the number of nucleations and orientation of crystals influenced by the particle loading. It is well known that curing behavior in fly ash–lime compacts is dependent on the ionic additives and the humidity/temperature history, therefore, it is possible to control or steer multi-component ionic systems by the post-deposition humidity/temperature history (analogy) [123]. Such interactions between additives, temperature and time in perovskite deposition can be explored efficiently with fewer experiments than full factorial approaches (analogy) can provide, by using design of experiments frameworks, e.g. Box–Behnken designs used in sustainable catalysis [119]. For process control, grey-box models and surrogate-assisted optimization methods for thermal and separation processes provide blueprints for the integration of first-principles drying/solidification models with data-driven corrections to ensure the quality of the film throughout reels and seasons (analogy) [125–132].

Solvent orthogonality and recombination-layer protection

The design of orthogonal solvents, and the robustness of the recombination layers, are important factors for stacking solution-processed WBG and NBG subcells on flexible substrates. In flexible all-perovskite tandems, an ultrathin PEIE nucleation layer was implemented to conformally coat ALD AZO onto the C60 layer and provided both electrical contact and a solvent/process barrier, allowing for the use of common non-orthogonal perovskite solvents for the top subcell and a water vapour transmission reduction of approximately one order of magnitude [1]. The molecule-bridged NiO architecture also achieved low interfacial recombination and strong adhesion, which were beneficial to the protection of the buried recombination contact during the following coating process [2]. In the case of the gas-quench strategy, films of WBG were obtained with no voids at the buried interface in the module scale, as WBG is

naturally devoid of solvents that may penetrate during the deposition of the top-cell. The fragility of recombination layers is further demonstrated by rigid all-perovskite module work, which emphasizes the need for sequences of deposition to be performed in a way that minimizes damage to the layers and the choice of aggressive solvents and plasma treatment [34]. Interpenetrating interfaces, which spread loading and offer multivalent bonding, can also provide spreading of chemical potential drops, decreasing the danger of delamination or redox reactions at abrupt discontinuities in composition (rigid, mechanistic study) [46]. Last, but not least, solvent sustainability and worker-exposure considerations should also enter into the selection of orthogonality: solvent hazard scores and recovery potential are not interchangeable with orthogonality, and the selection of solvent should consider wetting and drying properties as well as environmental metrics [31]. The proposed multi-stressor reporting protocol in Table 5 thus incorporates solvent orthogonality and barrier-integrity checks, since the selection of the solvent impacts on the residual stress stored during repeated thermal cycles and moisture uptake. This is in support of the statistical evidence gap depicted in Figure 5B and connects process chemistry directly to mechanical comparability [1–2,15–16,31,34].

Patterning, interconnections, geometric fill factor and yield

Low-loss interconnections free of brittle seams or high resistance scribe lines are the key relationship between what is translated from the coupon scale flexible tandems to modules. A certified efficiency of 23.0% at a 95.8% geometric fill factor (GFF) and 20.26 cm² area was obtained for the flexible all-perovskite modules using gas-quenched WBG layers and intelligent scribing and interconnection, demonstrating that high GFFs without compromising interconnect reliability are achievable [15]. The interconnection resistance and damage to the recombination layer are cross-platform bottlenecks, clearly demonstrated by the 21.7% perovskite module on a rigid substrate which is scalable, and the damage to the recombination layer is expected to be worse on flexible substrates if proper mechanics and solvent control are not used. Flexible modules, failures are frequently caused by localized module shunts or open circuits at scribe lines, pinholes caused by debris and barrier fractures in the edges of the modules. Monitoring and predictive maintenance of fouling and defect accumulation in heat exchanger and thermal processes can be transferred to predicting coating/scribing defect hotspots from upstream process parameters (analogy) [127–129]. Under ambient drift (analogy) [128–130] uncertainty-aware surrogates allow for fast scribe energy or overlap rule re-calibration. Backward compatibility (reverse interconnection) and possibility of separation at the downstream end of the circuit (analogy) should be considered when designing conductors and interconnections: Gravity-based separation has proven effective in recovering copper from scrap cables, and may be applied to size/density separation of flexible interconnects to reduce the load on the recycling chain. In addition, the pattern geometries can also mechanically interact with the stack, leading to narrower busbars and more aggressive scribes, creating an increased stress concentration and potential for delamination without toughened interlayers [5,43].

Roll-to-roll integration and quality control

Flexible all-perovskite Tandems must be aligned across hundreds of meters, with low

thermal budgets, during the roll-to-roll (R2R) coating, drying, interconnection, encapsulation and in-line quality control steps. With regard to the large area flexible tandem demonstrations, there is a convincing development towards this direction with the ambient-processed WBG layers slot-die coated on PET, gas quenched and combined with other flexible modules with hundreds of square centimeters of active area [15]. The processing insights gained from rigid modules (multi-head slot-die, vapor/solution hybrid and low-damage sputter/ALD conditions) are scalable to the R2R lines, which can be operated at temperatures suitable for polymer webs [34]. As flexible perovskite technology is reviewed, it is reiterated that device designs need to be mechanics-aware and that the devices must be encapsulated in a way that is fatigue-tolerant for R2R [42–43].

Quality control (QC) should not be thought of as a process of coupon inspection in the end but as an in-line multi-sensor model based system. Grey-box and reduced-input ML strategies, which allow estimation of latent film-quality descriptors based on small number of sensors, achieve a compromise between instrumentation requirements and prediction accuracy (analogy) [121,126–132]. Re-useable biosensor platforms provide design approaches for thin, flexible and reconfigurable sensing layers that would be incorporated into web carriers for on-going monitoring of humidity, solvent residue or ionic contamination (analogy) [133]. In terms of energy systems, exergy analysis and energy-recovery analysis on industrial lines promote heat integration (e.g., solvent-vapor heat recovery) to minimize the energy embodied in the R2R fabrication process, if the recovery of the heat from the solvents is safe [31,125,141]. Water reuse modules (e.g., lamella settlers) and membrane separations associated with other water-intensive sectors will provide an opportunity to close the water loop and decrease freshwater use in manufacturing (analogy) [114–115,118].

Flexible tandems are heading towards circular production, and two sustainability strands must be worked equally to integrate R2R. Mechanical damage modes that are specific to flexible modules need to be accounted for in a lead-containing and/or recovery plan. Chemical sequestration ability of encapsulated chemical-capture layers (e.g., supramolecular chelators) should be assessed after specified mechanical fatigue and barrier puncture in stacks as well as in pristine encapsulated layers [8–12]. Secondly, periodic rejuvenation strategies (modules are deliberately refurbished periodically), have been found to hold promise in enabling early market entry as circularity infrastructure matures, but demand that laminates, adhesives, and interconnects can be disassembled without causing catastrophic damage to the polymer substrate [29]. System-level analyses indicate that transformation to all-perovskite tandems can reduce sector-wide climate impacts, further encouraging flexible and lightweight form factors, with reliable and end-of-life issues being credibly taken care of [30]. The recycling processes that have been developed for the rigid modules, such as high-yield recovery of lead and aqueous-based separation, set performance targets and processes that need to be "re-engineered" for tandem-specific mixed Pb/Sn chemistries and multilayer polymer webs [7,13]. Besides the above mentioned end-of-life options for polymer-rich flexible modules there are thermochemical routes to consider, such as catalytic pyrolysis of polymer fractions, which should be assessed for their net environmental and resource flows (analogy) [142]. Third, for any R2R step, solvents should be assessed for health and

environmental parameters as well as process performance and recovery systems should be designed in from the beginning [31].

These developments and analogies inspire a manufacturing roadmap that aims for a co-optimization of nanocomposite function, mechanical design and chemical robustness under multi-stressor testing. The gap in efficiency between these two in Fig. 5A and the reporting incompleteness in Fig. 5B explain the need for transparent metadata and uncertainty-aware quality control, and Table 5 explicitly lays out a minimum set of experiments that must be reported. In this IMCC-centric perspective, "scalable and stable" no longer is a label for a lab recipe but is the key design constraint when considering the selection of nanocomposites, interfaces and processing sequences.

Sustainability, safety and circularity

If flexible all-perovskite tandems are to be commercialized, they must also be sustainable and safe, along with efficient and producing good yields. Environmental performance, in an Interface – Mechanics – Chemistry Coupling (IMCC) view, is not an external postscript, or a packaging problem: cracks or delamination (mechanics) open up fast diffusion pathways; interfacial redox and ion migration (chemistry) changes lead speciation and mobility; and the interfaces that stop recombination or water ingress must be designed to be disassembled and recycled. The ability to integrate the perovskite recombination layer with the barrier layer [1] and the solvent orthogonality [1] are elements that are already in place in literature as enabling characteristics for the flexible all-perovskite tandem architecture, and molecules used to create contacts also offer enhanced adhesion [2] and are utilized in ambient processes with validated mechanical and thermal stress testing [15]. However, the outcome of sustainability is still not strongly linked to the physics of the devices and the realities of scale-up. This section discusses the concepts of lifetime of lead contamination and lifetime of lead sequestration, choice of solvents and critical materials, repair and closed-loop recovery, and application-specific sustainability in the context of the IMCC lens and the translation roadmap in Figure 6.

Highlights

When combined with light, heat, humidity, and mechanical stress, sequestration efficiency and sequestration lifetime should be considered as primary device metrics, in addition to PCE [10].

Design recombination and encapsulation interfaces for high yield recovery of WBG and NBG streams for operational barriers and disassembly aids [1,7,13].

Make a change from using solvent screening based on only toxicity to a solvent–substrate–barrier compatibility analysis and recovery plan, accounting for manufacturing [31].

Match module rejuvenation with lead capture architecture to prevent the risk of leakage to occur when modules fail in the market [8,10,29].

Standardize the leakage test of damaged modules to the water-contaminant test (methodological analogy) after cyclic bending and puncture [143–144].

Lead containment and sequestration lifetime

Lead management in flexible tandems demands integration of materials chemistries,

device architecture and mechanics. Although physical encapsulation and barriers to oxygen and water help minimize ingress/egress, flex-induced microcracks can overcome the purely physical barrier [8]. As shown on rigid and flexible platforms, a well-designed junction can even serve as a processing barrier, minimizing cross talk from solvents and transmission of water vapor through the active tandem stack, thereby acting as an electronic contact as well [1]. Similarly, flexible modules can be stacked together in a recombination layer to share loads, to retard moisture diffusion and to preserve the integrity of the interface between the module and the substrate during bending, provided that both adhesion and fracture energy are optimized [1,4–5].

Lead sequestration can be incorporated into the stack to ensure that no lead reaches a breach should it get there. Supramolecular complexes and zwitterionic groups were reported to bind to Pb^{2+} and, in some instances, to deliver multiple functional properties like defect passivation and mechanical toughening (in single-junction rigid and flexible contexts, respectively) [6,12]. The feasibility of multifunctional additives for creating dynamic electrostatic and hydrogen-bond interactions that enabled the self healing of flexible single-junction devices and maintained performance after bending is a good example of the potential for nanocomposites to perform both mechanical and ion capture functions [6]. In flexible modules, these ideas need to be converted to thin, transparent layers to fit into low temp processing that don't disrupt recombination or current matching in tandems.

One of the main results of recent sustainability studies is the separation of sequestration efficiency and sequestration lifetime [10]. Efficiency of sequestration is the amount of lead immobilized per leakage event; sequestration lifetime is the time of the immobilization under environmental exposure. These need to be measured not only on pristine encapsulated devices, but on a realistic mechanical fatigue as well. Flexible tandem modules, for example, have been found to maintain performance across thousands of bends in a 10–15 mm radius and have been shown to be thermally robust (up to 85 °C (room temperature) to –40 °C), but there is currently no harmonized protocol that expands on the cyclic bending, controlled breach, humidity, and illumination experiments while simultaneously measuring time-resolved lead egress. A lack of a universally accepted ISOS-equivalent mechanical standard has been cited [5] and flexible perovskite stability reporting has been evolving [14,32–33]—these now need to be expanded to include damage-module leakage under strain [10].

The analytics need to be improved too. Water-quality fields offer methodological examples of powerful approaches to contaminant quantification and standardization, such as those in place for detection and regulation of polycyclic aromatic hydrocarbons (PAHs) in water [143]. Likewise, multivariate statistical methods developed for the surveillance of stream pollutants can be used to assist in sampling, outlier detection and source apportionment of perovskite module leachates as a function of bending history (methodological analogy) [144]. Uncertainty propagation in Monte Carlo to quantify the risks should include mechanical cycling histories, environmental conditions, and uncertain breach sizes, as in agroecosystems metal contamination risk assessment (methodological analogy) [145]. Lastly, the capacity of chemical capture systems to operate under realistic cycling, as well as the possibility

for their saturation in this context, should be modeled and tested, as requested in ref. [10] in this context for integrated isolation, immobilization, and recycling infrastructure.

In flexible single-junction cells, some groups have noted that decreasing the magnitude of the leakage leads to a decrease in the amount of leakage. Some groups have noted that in flexible single-junction cells decreasing the magnitude of the leakage leads to a decrease in the amount of leakage for certain cells by increasing the thermal diffusivity and modifying the interlayer chemistry. These should be viewed as proof-of-concept for the design of materials using the guidance of the IMCC, but not yet as complete tandem-module solutions. The recombination junction is a logical location for secondary capture agents for tandems since it is located in the center, mechanically protected by subcells and chemically orthogonal [1–2]. Any capture chemistry that is put in here, however, should not affect the recombination resistance or optical loss.

Solvents, critical materials and manufacturing footprint

The selection of solvents is still an important factor that affects worker exposure, emissions reduction, and circularity of the module. The health and environmental hazards of common perovskite solvents have been quantified and the importance of considering their toxicity, volatility and process performance has been emphasized [31]. The solvent orthogonality between subcells is especially sensitive for flexible tandems due to the mechanical fragility of the ultrathin layers and the low thermal budgets of the polymer substrates. It is already known that using ultrathin polymeric nucleation layers can significantly broaden the solvent window for the processing of a top cell in both rigid and flexible tandems with the low-temperature atomic-layer-deposited oxide barrier [1]. More recently, large-area IGWBG films and modules have been processed in an ambient environment using solvent systems compatible to slot-die coating [15], indicating that ambient processing of flexible substrates could be carried out without a performance loss, provided that the nucleation and drying kinetics of the film and/or module is controlled along the IMCC axes.

But sustainability is also driven by critical materials demand. The use of indium-based transparent conductors is commonplace, but costly and subject to long-term supply problems. All-perovskite tandems have been shown at scale with rigid interconnect architectures and crack-resistant, low-resistance conductors, but further into the future there are still transparent electrode architectures and low-resistance, crack-resistant conductors for flexible modules to be demonstrated at high efficiency. A mini-module level report of barrier-integrated transparent electrodes for flexible perovskite cells suggests a means to decrease the number of layers and interfaces while simultaneously enhancing moisture resistance [44]. The selection of any electrode or barrier materials will then be reconsidered on the basis of the sheet resistance, transmittance as well as fatigue resistance, adhesion energy, vapor transmission, and recyclability.

Manufacturing footprint is not just about materials, it's about energy used, yield generated and emissions produced. However, as per the life-cycle assessment (LCA) study, there is a significant reduction in the energy payback time and the greenhouse-gas (GHG) emissions of perovskite modules with enhanced reliability and high

recovery value [9,11]. Projections of PV sector impacts on climate also indicate that all-perovskite tandems could decrease impacts, provided that they are made durable and at scale [30]. Circular business models with periodic module rejuvenation have been suggested to speed up the introduction in the market of tandems, if the externalization of lead leakage/rework waste for rejuvenation is avoided [10,29]. Emissions monitoring and predictive control on the factory floor can take advantage of data-driven models developed in other sectors; for example, machine-learning predictors of ozone emissions with interpretable features provide a means to integrate sensor data with process setpoints (methodological analogies) [138]. Solvent-recovery and waste-treatment investments in perovskite lines can be guided by methods that can be transferred from techno-economic assessment practices in production of bioenergy and hydrogen, including methodological analogies such as coupling yield, co-product valorization and process selection under uncertainty [146-149].

Repair, delamination, recovery and closed-loop design

Closing the loop requires designing today's interfaces for tomorrow's disassembly. Recovery of lead and transparent conductors with high efficiency and minimal secondary pollution has been demonstrated on rigid glass modules; recycled PbI₂ and transparent conductors have been used to remake devices without performance loss [7]. Aqueous-based recycling protocols capable of producing materials with device-level performance parity have also emerged [13]. However, the flexible-stack problem is materially different: polymer substrates, ultrathin oxides, adhesives, and tandem-specific recombination layers complicate separation. Thermal delamination used for glass modules is often incompatible with low-T_g substrates [7]. Therefore, flexible tandems need "delamination-on-demand" interfaces—adhesives and interlayers whose adhesion can be switched by mild triggers such as temperature windows narrow enough to preserve polymer integrity. Interfacial engineering strategies that strengthen adhesion and reduce defect density in operation can be re-tuned to enable controlled release at end-of-life [3,46]. The fracture-mechanics framework used to interpret bending fatigue—where stress scales with elastic modulus and thickness and failure depends on flaw size and fracture toughness—also applies to delamination design: recovery processes should seek to concentrate fracture at benign, pre-defined interfaces while protecting critical semiconductor junctions [4].

Closed-loop recovery must also maintain the chemical identity of both subcells. Wide-bandgap (WBG) and narrow-bandgap (NBG) absorbers differ in halide content and, for NBG, in Sn/Pb ratio and oxidation state; mixed streams complicate purification [17–20]. Tandem-specific protocols should thus target selective dissolution or delamination that separates WBG from NBG layers before aggregation, as emphasized in recent recycling advances [13]. Mechanical pre-processing (e.g., roll-slit segmentation) could expose designed "release planes," while aqueous or weakly acidic chemistries recover lead halides with high selectivity [7,13]. Critically, such protocols must be validated for flexible encapsulation stacks, where adhesive/barrier synergy is engineered to both resist moisture during life and to debond cleanly during recovery.

Lead capture during accidental damage and during scheduled rejuvenation is a separate design target. Chemical capture materials integrated into encapsulants have

been recommended as part of a layered containment approach [8,10]. Multifunctional flexible nanocomposites that toughen interfaces and immobilize lead are promising, but their compositional and loading thresholds must be connected to measurable transport, modulus, optical, and chemical-reactivity limits to avoid trade-offs that increase shunting or reduce transparency [6]. This review adopts a function-first nanocomposite taxonomy precisely to make these trade-offs legible for recovery and safety decisions.

Decision frameworks from other fields can help structure multi-criteria choices among adhesives, barriers, and capture chemistries. Complex fuzzy-bipolar soft-set methods illustrate how to encode competing criteria and stakeholder preferences formally (methodological analogy) [150]. Likewise, multivariate pollutant-trend analyses and standardized sampling from hydrology can inform how to validate delamination effluents and post-damage drainage (methodological analogy) [143–144]. Risk models of metal(oids) in soil–crop systems demonstrate the value of Monte Carlo propagation of uncertain sources and sinks to derive defensible, conservative designs for capture capacity and buffer volumes (methodological analogy) [145].

Application-specific sustainability

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Data-driven optimization and digital manufacturing

Defects in process control and quality assurance of flexible all-perovskite tandems may result from small fluctuations in the wetting and drying of the interfaces, in interfacial chemistry, and in placement of the neutral axis, and affect electronic, mechanical, and chemical failure modes. High-dimensional, partially observable solution space. Optimization based on data can speed up the discovery to manufacturing process provided it is based on physics, uncertainty-aware, and is tightly coupled to interoperable datasets that record the IMCC state variables. Figure 6 illustrates a translation ladder in which modules with embedded sensing and model-based control are translated from a coupon to a roll-to-roll form.

Physics-informed and uncertainty-aware machine learning

There is a growing number of public data sets for perovskite devices, but these data sets are typically missing the metadata to recreate the performance for flexible devices or to learn transferable models. Flexible-device datasets should include at a minimum, thicknesses, moduli (where known), and interfaces designed for delamination; substrate and encapsulation thicknesses and moduli to be used to compute neutral-axis position and tensile/compressive strain; bending protocols (radius, calculated strain, orientation, waveform, rate, dwell, cycle count) [4–5]; environmental conditions and ISOS-like stressors, noting when combined; electrical operating point during tests; lead-sequestration efficiency and sequestration lifetime under pristine, fatigued and punctured conditions [10]; solvent systems and drying conditions; barrier permeation data [31]; yield and defect statistics for scaled coating [15,34]; and adhesion and fracture metrics for buried interfaces [3,46]. Modules should include interconnection resistance, recombination-layer integrity after scribing and edge-seal geometry.

For interoperability, controlled vocabularies and units are required, and for the design of metadata, feature engineering, and data quality control, templates are available based on standardized ontologies from environmental monitoring and pollutant-analysis workflows (methodological analogies) [143–144]. Methodological analogy:

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Risk-centric datasets must include Monte Carlo representations of manufacturing and operation uncertainty, for propagation of variance to performance and safety quantities [145]. Formal decision sciences (methodological analogy) [150] may encode multi-objective trade-offs among material toxicity, cost, availability, mechanical compliance and recyclability in decision support data layers.

In-line sensing, surrogate models, and digital twins

In-line sensing should be able to measure the states of critical layers and interfaces in real-time in order to realize roll-to-roll flexible tandems. Film thickness and drying fronts can be monitored via optical monitoring (e.g., reflectance/ellipsometry), non-radiative recombination/s and shunts can be detected by using PL and lock-in thermography, web tension and curvature can be measured by using mechatronic sensors, and the species of solvents can be measured by using vapor sensors for safety and recovery. A key challenge is to convert sensor streams into actionable control variables, which should be done by surrogate models – fast, approximate predictors trained from detailed simulations and experiments. The gradient-boosted trees, including XGBoost, have successfully captured interpretable relationships in emission models and shown the possibility of combining multi-sensor data to develop confident predictions (methodological analogy) [138]. Surrogate models can be used for flexible tandems to predict the likelihood of damage to the recombination layer when a new solvent is suggested, or to predict loss of adhesion after a tension–compression protocol, allowing proactive changes to be made to the solvent quench line, the line temperature or line speed.

These surrogates and sensors are linked to mechanical models of film formation, ion transport and fracture via digital twins. They would then be changed from lab scale (environmental control spin coaters) to pilot scale (environmental control in-line optics and permeation monitors; slot-die, blade, or spray coating; end-of-line module testing) and ultimately to manufacturing scale (roll-to-roll coating with integrated solvent-recovery, emissions control and end-of-line module testing) [15,34]. If recombination interfaces also serve as water-vapor barrier surfaces, the twin needs to monitor: electronic recombination parameters, and water-vapor transmission parameters, computing expected value of the water-vapor sequestration lifetime based on the rolling environmental forecast [1,10]. Multi-objective metaheuristics can be used to optimize control policies while maintaining safety constraints (maximum allowable concentration of solvent vapor, maximum capacity of solvents for capture) and production targets, as has been done in hybrid scheduling of energy systems (methodological analogies) [135,151–153]. Whereas the twin allows for propagation of process choices to cost and environmental indicators, as is common in hydrogen/bioenergy analysis, it should be embedded in the techno-economic module to propagate process choices to cost and environmental indicators [146]. In all layers, limits on workers' exposure to solvents and particulates must be binding, as required by the solvent-impact assessments [31].

Research gaps and a testable roadmap

The evidence presented here calls for a disciplined approach to thinking from device-

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centric to system-centric: flexible all-perovskite tandems are systems that must be designed, manufactured, monitored, repaired, and recovered. The gaps are summarized in a synthesizing table (Table 6) with methods and target outcomes that can be tested, and Figure 6 places these gaps on the translation ladder. [1–20,29–34,39–40,42–46,62,86].

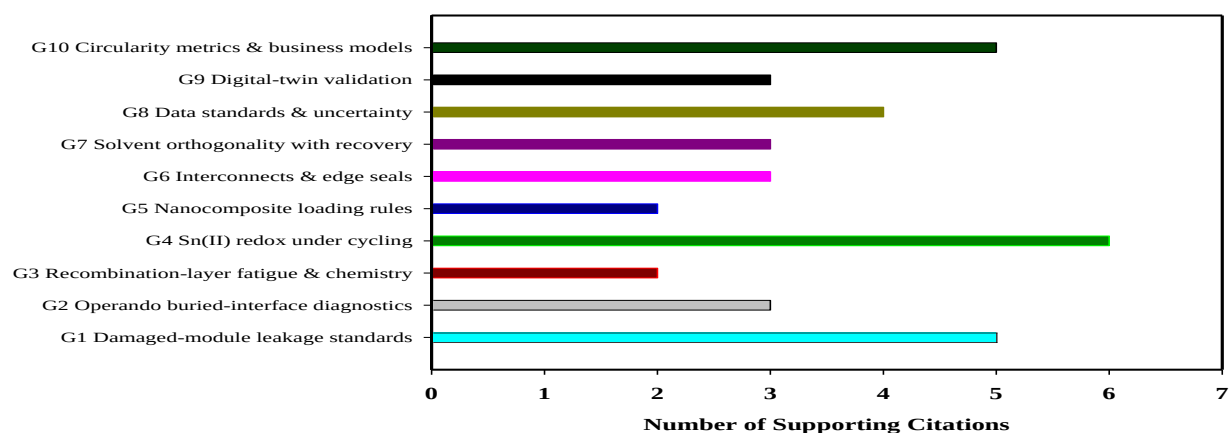


Figure 6: Research gaps by translation-ladder stage (source: Section 11.1) [1–20,29–34,39–40,42–46,62,86].

Figure 6. The ten research gaps, mapped in the translation ladder, with the size of the gaps determined by the supporting-citation density (Section 11.1). The standards and diagnostics for damage modules, the operando validation of buried-interface diagnostics, the recombination-layer fatigue and chemistry, the Sn(II) redox under cycling, the loading rules for nanocomposites, the interconnects and edge seals, the solvent orthogonality with recovery, the data standards and uncertainty, the digital-twin validation, and circularity metrics and business models are all covered in this technical group.

Ten clearly numbered research gaps

Damaged-module leakage standards. Multi-stressor protocol does not currently exist that combines cyclic bending (with reported strain), controlled breach/puncture, humidity/oxygen, illumination and maximum power point operation to quantify the lifetime of the egress and sequestration of lead in flexible all-perovskite tandems [5,10,14,32–33].

Buried-Interface diagnostics under strain (Operando). Flexible tandems are typically not measured under operando conditions of adhesion, interfacial fracture energy, ion accumulation and chemical speciation during cyclic deformation [3–5].

Fatigue and chemistry of the recombination layer. The recombination function in any tandem must be efficient and have a low loss component to it, as well as being a solvent barrier and not experiencing fatigue when under combined solvent and strain conditions, and pathways for degradation remain poorly characterized [1–2].

mechanical and thermal cycling redox of Sn(II). Most strategies for NBG sub-cells are optimized without the consideration of strain transfer, low temperature processing or scalable drying [17–20,39–40].

Rules for loading/composites based on function. No dose–structure–function maps

exist for multifunctional additives for passivation, stress dissipation, and lead sequestration that incorporate thresholds for transparency, percolation, modulus, roughness and chemical reactivity [6,12].

Interconnects and edge seals are flexible. Further development and standardized testing is needed for low resistance, chemical orthogonality, and fatigue resistance in flexing modules for tandem interconnects and edge seals [15,34,44].

Orthogonality of the solvent used and recovery. There is a lack of documentation of solvent systems that are suitable for low-temperature flexible substrates and interlayers, that provide uniform films in slotdie/blade/spray processes, and that can be recovered at high yield and low emissions in tandems [1,15,31].

Data standards and uncertainty. Full IMCC metadata and uncertainty quantification are rarely included in the datasets of flexible tandems, and there is limited interoperability between labs and scales [5,14,32–33].

Digital-twin validation. There are very few studies that validate digital twins which couple in-line sensing, physics-based modeling, and surrogate learning for control of flexible-tandem manufacturing and for prediction of safety-relevant metrics (e.g., sequestration lifetime) [10,15,34].

Circularity metrics and business models 10). Technologies which combine PCE retention, mass-specific power, recovery yield, sequestration lifetime, solvent recovery and number of rejuvenation cycles into one flexible-module circularity metric do not exist, nor are there validated models of how rejuvenation interacts with leakage risk and producer responsibility [9–11,29–30].

Six falsifiable hypotheses aligned with Table 6

H1. Recombination-junction barrier co-design reduces ingress and slows mechanochemical degradation. In flexible all-perovskite tandems, integrating a conformal low-temperature oxide at the recombination junction atop an organic interlayer will reduce water-vapor transmission across the active stack relative to an organic-only junction of equal optical thickness, without increasing recombination losses. Falsification: measure WVTR and recombination-layer resistance before/after cyclic bending and solvent exposure; failure to observe a reduction in WVTR at constant or improved recombination performance falsifies the claim [1–2].

H2. Sequestration-lifetime is a separable, designable property. Embedding a chelating supramolecular complex within the encapsulant leads to a time-dependent lead-release profile under standardized breach–fatigue testing that is sublinear in breach area over an initial regime, indicating true immobilization rather than mere dilution. Falsification: design a puncture–bending protocol and quantify lead egress; linear scaling with breach area at early times would falsify [8,10,12]. Cross-check for flexible single-junctions to control for stack-chemistry effects [6].

H3. Physics-informed models outperform black-box learners in extrapolation. For predicting flexible-tandem yield during slot-die coating across solvent compositions and line speeds, a physics-informed surrogate that encodes drying kinetics and dimensionless numbers will show lower prediction error on out-of-distribution conditions than a parameter-matched deep neural network trained on the same data. Falsification: define OOD test sets; lack of improvement in OOD error falsifies [15,136,139,154].

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H4. Neutral-axis engineering measurably extends sequestration lifetime under cyclic strain. Shifting the neutral axis into the active stack via asymmetric encapsulation reduces the rate of lead egress under cyclic bending with controlled breaches relative to symmetric encapsulation at equal barrier thickness. Falsification: execute identical breach–bending tests with quantified strain and measure lead release vs cycles; absence of reduction falsifies [4–5,10].

H5. Function-first nanocomposite rules predict safe loading windows. For a given multifunctional additive family, there exists a percolation-limited loading window in which mechanical toughening and lead chelation improve without increasing optical parasitics or shunt risk; this window can be predicted from independently measured modulus, refractive index, particle size, surface functionality, and halide reactivity. Falsification: failure of predicted windows to avoid parasitic increase or shunting across at least two additive families falsifies [6,12].

H6. Module rejuvenation is compatible with capture-first designs. Implementing barrier-integrated capture chemistries and designed release-interfaces enables at least one rejuvenation cycle (absorption layer replacement) on flexible tandem mini-modules without increased lead leakage during rejuvenation relative to first-life recovery. Falsification: side-by-side rejuvenation with and without capture-first designs; any increase in leakage for capture-first designs falsifies [7,10,13,29].

Near-, medium-, and long-term translation milestones

Near term (0–2 years)

Publish an ISOS-Mechanical extension with standardized breach–fatigue–operation protocols and validated analytical methods for lead quantification, drawing on water-contaminant standards (methodological analogy) [14,32–33,143–144]. Include required reporting of strain, bending mode, environment, and operating point [4–5].

Demonstrate flexible all-perovskite tandem coupons with recombination-junction barrier stacks that preserve efficiency after combined solvent exposure and bending, and document WVTR and adhesion vs cycling [1–2].

Release IMCC-complete datasets with uncertainty estimates and interoperable metadata schemas; provide physics-informed surrogate baselines for at least one scalable coating workflow [15,136,139].

Medium term (2–5 years)

Validate digital twins on pilot slot-die lines for flexible tandems with in-line sensing, physics-informed surrogates, and control policies optimized by constrained metaheuristics; demonstrate predictive control of yield and barrier integrity [15,34,135,138,151–153].

Quantify sequestration lifetime in flexible tandem mini-modules with integrated capture chemistries under standardized breach–fatigue–operation; report failure envelopes and saturation behaviors [8,10,12].

Report selective delamination protocols that separate WBG and NBG layers on flexible stacks with high recovery yield, compatible with aqueous or weakly acidic chemistries; benchmark circularity metrics that include solvent recovery and rejuvenation readiness [7,11,13,29].

Long term (5+ years)

Certify flexible all-perovskite tandem modules with validated multi-stressor durability (light–heat–humidity–mechanics) and documented sequestration lifetime; integrate module health sensing for predictive maintenance [2,15,32].

Deploy capture-first, release-on-demand encapsulation architectures at manufacturing scales with closed-loop solvent and lead recovery; embed producer-responsibility mechanisms and field leakage tests based on standardized damaged-module protocols [10,31].

Integrate flexible tandem production into decarbonized energy systems with coordinated storage and power-to-fuels pathways; evaluate system-level environmental and economic impacts using updated LCAs that include rejuvenation and circular operations (contextual analogies) [9,29–30,146–149].

Table 6 converts the review’s research gaps into falsifiable hypotheses, proposed measurements, and translation gates. The table is intended as a decision framework: a candidate material or interface should advance only when it demonstrates concurrent electronic, mechanical, chemical, manufacturing, and safety benefits rather than a single champion metric [1–20,29–34,39–40,42–46,62,86].

Table 6. Research-gap roadmap with testable hypotheses and translation gates

Research gap	Testable hypothesis	Minimum experiment	Success criterion	Translation gate
Sn-rich buried-interface redox under bending	A chemically anchored antioxidant/adhesive layer suppresses Sn(IV) formation while increasing fracture energy	Operando XPS-compatible aging or periodic depth profiling during MPP + cyclic strain	Lower Sn(IV)/Sn(II), lower Voc and higher interfacial fracture energy than controls	Reproducible on slot-die-coated NBG films
WBG phase segregation coupled to stress	Controlled compressive residual strain raises the barrier to halide segregation without increasing delamination	Spatial PL/GIWA XS under calibrated tensile and compressive strain with illumination	Smaller reversible/irreversible PL shift and no loss in adhesion	Stable in tandem current-matched operation
Nanocomposite dose–structure–	An optimum loading window exists where	Systematic loading series with dispersion,	Pareto improvement in PCE, fracture	Same optimum persists across

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function rules	passivation/toughening gains precede percolation or optical penalties	roughness, PLQY, mobility, modulus, and fatigue mapping	resistance, and aging	coating scale and batch
Flexible interconnect fatigue	A low-lateral-conductivity, ductile recombination junction localizes shunts and resists cyclic cracking	Four-point transport, cross-sectional microscopy, and tandem fatigue under MPP	Stable out-of-plane recombination with low lateral shunt propagation	Demonstrated in laser-scribed mini-module
Barrier and edge-seal fatigue	Neutral-axis-positioned hybrid barrier retains low permeation after repeated strain	WVTR/TR before and after multi-radius fatigue plus damp-heat	Minimal permeation increase and no edge pumping	Continuous lamination compatible with R2R
Coupled qualification	Multi-stressor damage is non-additive and can be predicted from a small set of operando state variables	Designed experiment varying light, heat, RH/O ₂ , bias, and strain with uncertainty-aware model	Validated lifetime prediction on unseen stress profiles	Field-correlated acceleration factor
Circular flexible module	Reversible adhesives and selective sorbents permit high-yield recovery without sacrificing service reliability	Damage/leakage test followed by low-temperature delamination and material recovery	High Pb capture/recovery, reusable substrate/electrode, comparable remanufactured PCE	Cost and LCA advantage over disposal [7,104–105,140]–[13]

Source: Authors' research roadmap synthesized from the literature cited throughout this review [1–20,29–34,39–40,42–46,62,86].

Conclusions

Flexible all-perovskite tandems sit at the intersection of delicate buried interfaces, demanding mechanical loads, and chemically reactive absorbers. The Interface–Mechanics–Chemistry Coupling framework advanced here makes these interdependencies explicit, enabling a sharper critique of what constitutes progress. The most convincing device demonstrations to date—barrier-enabled recombination junctions on flexible stacks [1], molecule-bridged contacts that also strengthen adhesion [2], and ambient-processed large-area modules with documented bending and thermal cycling [15]—succeed because they resolve at least two IMCC axes at once. Sustainability and safety will only be achieved when the third axis is fully integrated: sequestration efficiency and sequestration lifetime must become design objectives, measured under combined stressors with standard protocols [5,10,14,32–33]. Manufacturing must be digitized—sensed, modeled, and controlled—using physics-informed, uncertainty-aware tools that respect the constraints of flexible stacks and the realities of solvent management [15,31,136,139]. Circularity must be built in through release-on-demand interfaces and capture-first encapsulation that enable high-yield recovery of both WBG and NBG streams [7,13,29].

The breadth of the bibliography across environmental analytics, machine learning, optimization, hydrogen systems, and decision science provides methodological and contextual precedents—not direct photovoltaic evidence. When used transparently as such, these fields offer robust test designs, risk quantification, and control strategies that perovskite researchers can adapt. The next generation of flexible all-perovskite tandems will be judged less by short-term champion efficiencies and more by whether IMCC-grounded designs deliver durable power, safe failure modes, and high-yield recovery at scale. The roadmap in Figure 6 and the hypotheses in Table 6 are intended to make that judgment testable.

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