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UNRAVELING THE INTRINSIC STABILITY MECHANISMS IN NON-FULLERENE ORGANIC SOLAR CELLS: A PATH TOWARD DURABLE PHOTOVOLTAIC PERFORMANCE

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Abstract

Introduction: *Non-fullerene organic solar cells (NF-OSCs) have been regarded as potential candidates for the next generation photovoltaic technology because of their tunable optical properties, solution based fabrication process, and lightweight features. However, their large-scale applications are prevented by their poor long-term stability under environmental conditions including high temperature and the exposure of O2 and light. The comprehension of the intrinsic stability mechanisms at the molecular level is paramount for enhancing the life time of devices and making these efficient devices commercially available.*

Objective: *Our work aims to decipher the intrinsic stability routes of NF-OSCs by examining the structural, thermal, and chemical stabilities of high-performance D–A systems, concentrating on D18 and a non-fullerene acceptor Y6-BO.*

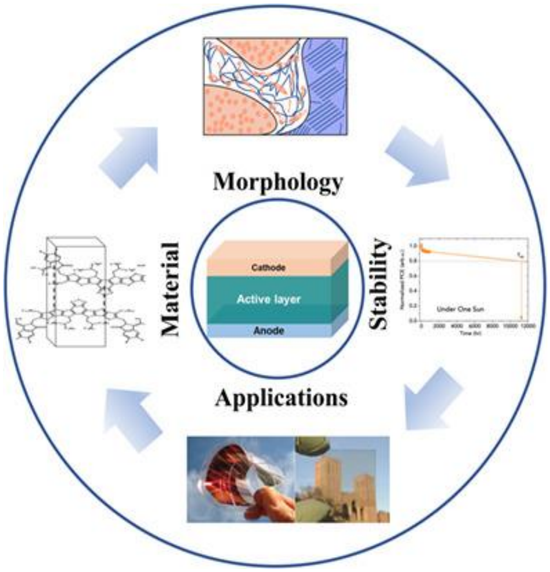
Method: *A comparative study on two donor–acceptor pairs (D1-A1 and D2-A2, with D2-A2 being equivalent to D18:Y6-BO system) was adopted. The devices were made under identical conditions and were treated by aging protocols of ISOS-D1. The materials were characterized by UV-vis, FTIR, AFM, TGA, photoelectron spectroscopy, lifetime monitoring under continuous illumination and thermal treatment. Formation of reactive oxygen species and chemical degradations pathways were also investigated by electron spin resonance and chemical trapping studies.*

Results: *The D18:Y6-BO system (D2-A2) featuring significant thermal and photochemical stability with over 90 phosphorescence MQE at 1,000-h life time and >10,000 projected T_{80} life time. Morphological integrity, low radical generation, and diminished oxygen consumption were found, suggesting an improved intrinsic material health.*

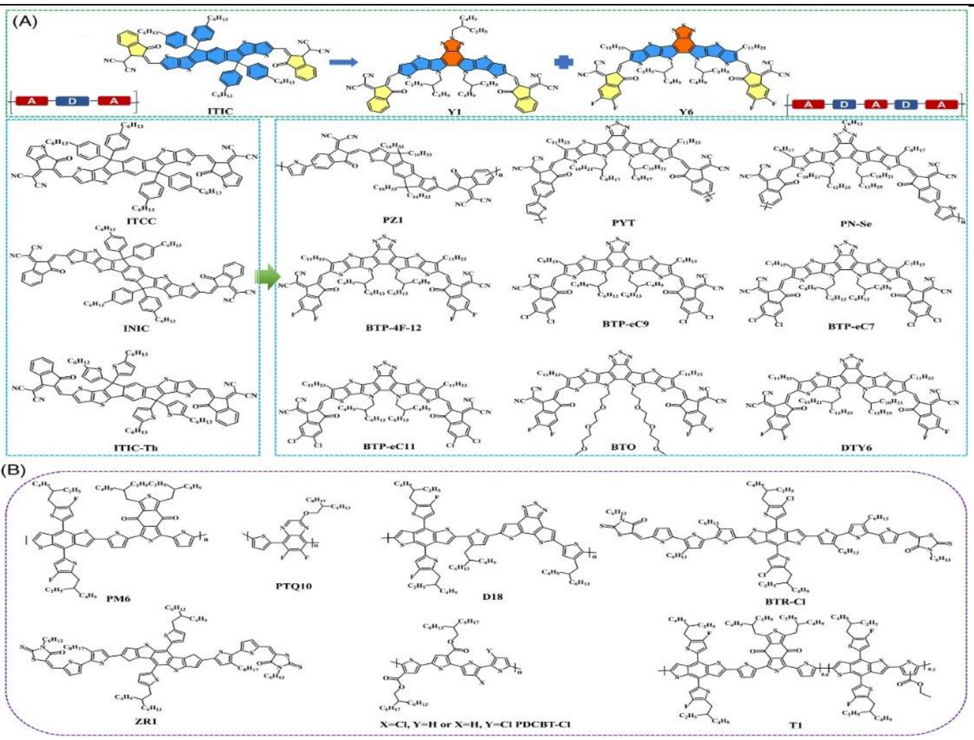
Conclusion: *Tailoring the molecular structure of NFAs is an effective way to enhance the operational stability of the NF-OSCs. The work uncovers fundamental molecular characteristics that drive long-term performance and presents a design route for bankable organic photovoltaics.*

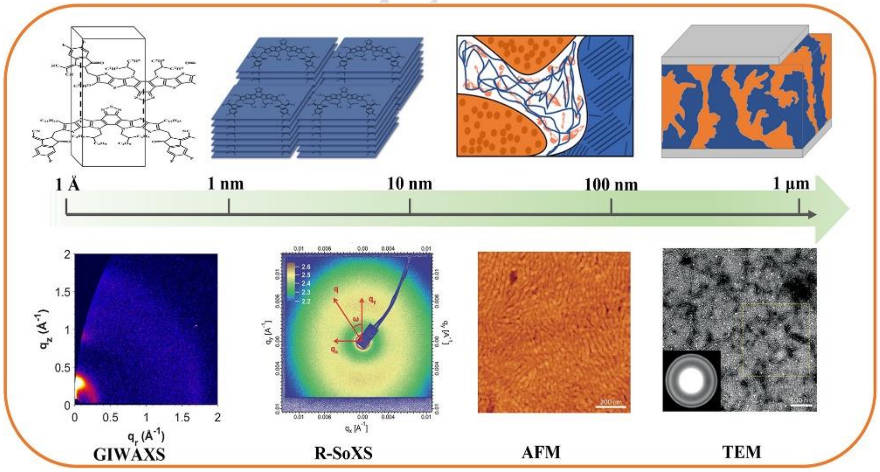
INTRODUCTION

In the past couple of years, non-fullerene organic solar cells (NF-OSCs) have emergedas potential candidates for organic photovoltaics (OPVs) replacing the conventional fullerene-based solar cells, with better power conversion efficiencies with tunable electronic property. Compared with fullerene acceptors, non-fullerene acceptors (NFAs) offer more design freedom at the molecular level, which facilitates better light harvesting, energy level matching and morphology adjustment of the active layers (Liu et al., 2022). such benefits have raised power conversion efficiencies of NF-OSCs to over 18%, making NF-OSCs closer to commercialization (Wang et al., 2023). Nevertheless, the long-term operational stability of NF-OSCs is a key bottleneck that prevents their practical photovoltaic applications.



The inherent stability of NF-OSCs is affected by a number of interconnected complex factors, such as molecular design, morphology evolution, and interfacial properties in the device (Zhang et al., 2023). More recently it was reported the importance of molecular packing and phase separation as mechanism to stabilize the morphology of the active layer as directly affecting the active photo-stability and thermal endurance of the device (Kim et al., 2024). In addition, the chemical degradation routes, including photo-oxidation and thermal decomposition, affect both the lifetime of NFAs and donor materials, requiring further detailed mechanistic studies to realize the degradation triggers and to suppress their influence (Chen et al., 2021). Photo-oxidation usually starts when molecules of the active layer absorb light photons (hv), resulting in excited singlet or triplet states (S1, T1), which in turn interact with molecular oxygen (O_2) to produce reactive oxygen species (ROS), such as singlet oxygen (^1O_2) or superoxide radicals ($O_2^{\bullet -}$). The general reaction sequence is as follows:





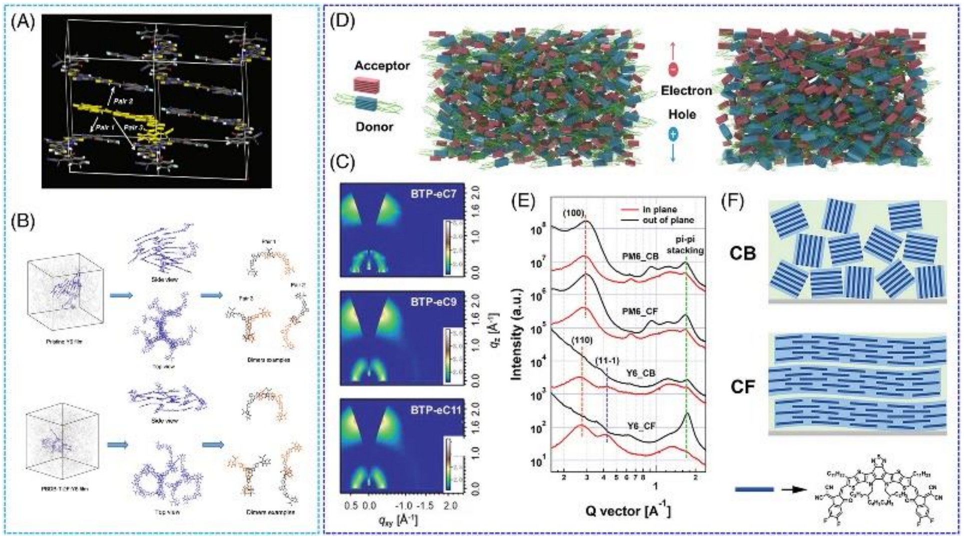
$$\text{NFA} + h\nu \rightarrow [\text{NFA}]^* \ (S_1 \text{ or } T_1)$$

$$[\text{NFA}]^* + O_2 \rightarrow [\text{NFA}]^{\bullet +} + O_2^{\bullet -}$$

$$O_2^{\bullet -} + \text{NFA} \rightarrow [\text{NFA–O}]_x \ \text{(oxidized products)}$$

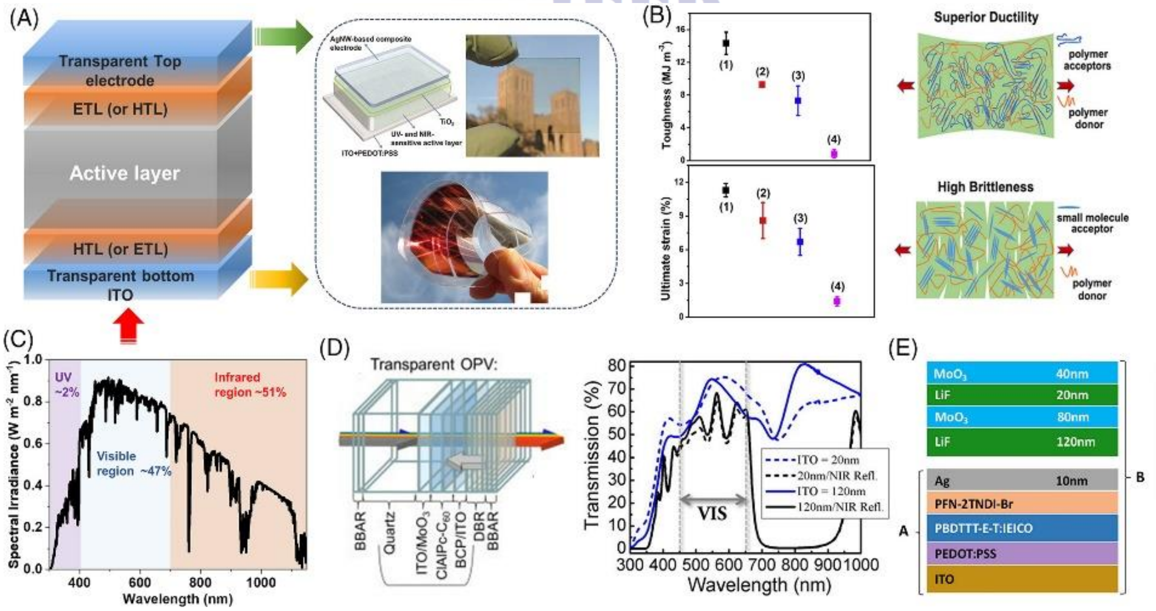
This domino system results into molecular degradation through a chain reaction, and subsequently leads to chemical bond scission and reduction of electronic conjugation, further causing the decreased device efficiency.

One of the most important parameters affecting the operational stability of NF-OSCs is the morphological stability. The nanoscale morphology that dictates exciton dissociation and charge transport can experience deleterious changes upon illumination and thermal cycling, resulting in phase segregation and loss of charge transport pathways (Li et al., 2022). Solutions including adding molecules, crosslinking and using thermally resistant NFA have been suggested for improving the morphological robustness (Guo et al., 2023). Furthermore, thermal shear may open chain scission or isomerization reactions in the conjugated polymers or NFAs, for example:



$$\text{"Polymer–C=C–C=C–"} \xrightarrow{\perp \Delta T} \text{"Polymer–C–C–C=C–"}$$

(which changes the conjugation length and decreases the mobility of charge carriers). The result reveal the electron and hole transport layers, which can act as buffer layers, actively becoming involved in stabilization of the device so as to stabilize the interfacial reactions, the probability of charge extraction and to inhibit the accumulation of trap states and hysteresis effects (Xu et al., 2024).

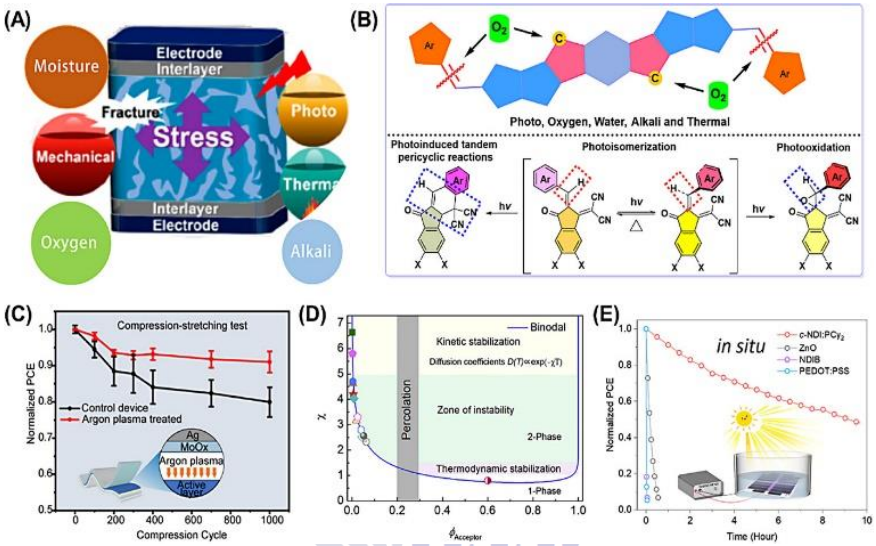


Chemical stability also resistance to photochemical degradation is another important factor that impacts the lifetime of NF-OSCs. NFAs normally involve the polymer donor type with conjugated backbones and electron-withdrawing moieties sensitive to photo-induced oxidation, performance decay over time have been observed in field (Zhao et al., 2023). For instance, the photo-oxidation of an NFA with an acceptor group such as dicyanomethylene can occur by way of radicals formation and followed by chain breaking:

CH) It is obvious that the first step of the degradation process is,

$$-CH=C(CN)_2+O_2^{\cdot-} \rightarrow \text{radical intermediates} \rightarrow \text{fragmentation}$$

Advanced encapsulation strategies, and the design of intrinsically stable NFAs through molecular engineering have proved to decrease these degradation pathways and increased by far the photostability of devices (Singh et al., 2025). In addition, device stability depends on the thermodynamic and dynamical properties of the carriers in the active layer, where non-equilibrium or trapped carriers can promote the degradation through trap-assisted recombination and the generation of additional reactive species.



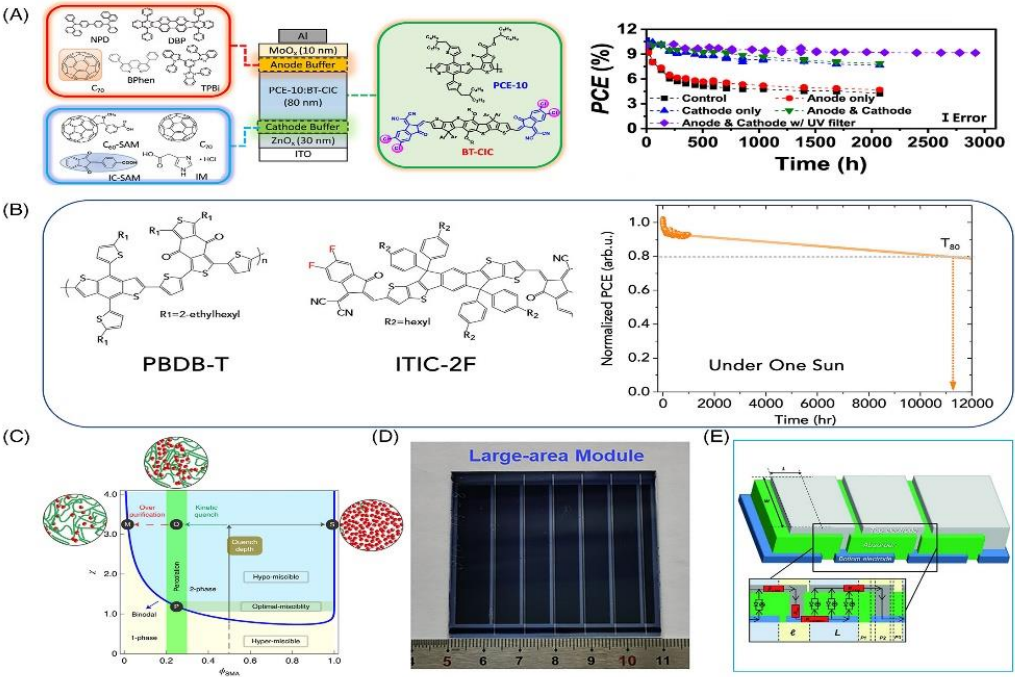
Interfacial engineering has also been investigated for improved stability by improving the electronic alignment and the chemical compatibility between the active layer and the transport layers. Stable, defect free interface formation by minimizing non-radiative recombination and degradation through interfacial charge build-up or chemical reaction can be maintained (Park, Leem et al., 2023). Radical quenching and interfacial barrier formation can be represented by reactions such as:

$$\text{Trap}+e^-\longrightarrow \text{Trap}^-$$

$$\text{Trap}^-+O_2\rightarrow \text{Trap}+O_2^{\bullet-}$$

where perfect radicles formation pathways are prevented by the interface modifiers. Emerging materials, such as self-assembled monolayers (SAMs) and two-dimensional (2D) materials, have been incorporated as interface modification layers to enhance FF and the long term stability of NF-OSCs (44 Lee et al., 2024). These studies reveal the multi-dimensional nature of intrinsic stability that should be addressed as a whole in terms of molecular design, morphology, chemical stability and interface engineering.

Although much progress has been made, the complex interaction of various the intrinsic factors for stability of NF-OSCs is still poorly understood. A comprehensive link between molecular structure, device morphology, chemical robustness age and interface properties and the long-term stability of NF-OSCs remains under active development. This knowledge is crucial for the rational design of new-generation materials and devices, which could perform high efficiency and last for a long time. 43 To realize the potential of NF-OSCs for practical, long-term photovoltaic applications, it is, therefore, essential to understand these innate mechanisms.



Problem Statement:

The efficiencies of the non-fullerene organic solar cells (NF-OSCs) are highly impressive, but their practical applications have been heavily restricted by stability, which is related to the complicated intrinsic degradation mechanism. Most of the existing efforts have been concentrated in increasing efficiency with insufficient attention to the underlying stability issues induced by morphological evolution, chemical degradation, and interfacial instability. This lack of understanding of the inherent stability mechanisms further limits the realization of long-lifespan and sustainable high-performance NF-OSCs, thereby urgent required are systematic studies aiming at clarification the underlying stability processes.

Significance of Study:

Such work is of fundamental importance to promote the field of organic photovoltaics by thoroughly exploring the intrinsic stability mechanisms that determine the long-term stability of NF-OSCs. Interconnecting microscopic understanding with macroscopic behavior at the device level, the results can direct the rational design of more stable materials and device structures. Improved stability will promote not only the market prospects of NF-OSCs, but also the large-scale application of sustainable and renewable energy technologies in the future with some competitive advantages in the cost, service lifetime, and so on.

Aim of Study:

The main objective of this research is to dissect and describe the inherent stability of non-fullerene organic solar cells which govern the long-term photovoltaic behavior. This includes understanding the molecular, morphological, chemical and interfacial features that may lead to device degradation, and address how they may be mitigated. Ultimately, the present work is aiming to learn the design rules of fabricating long lived NF-OSCs with high efficiency.

Methodology

A multi-pronged experimental strategy involving material synthesis, accelerated aging protocols, and advanced characterization techniques was employed in this study to explore the intrinsic stability mechanisms of NF-OSCs. A number of benchmark and new NFAs, including Y6 and its derivatives, were synthesized by Suzuki coupling and Knoevenagel condensation reactions, and then were purified via column chromatography or recrystallization, according to the procedure in Liu et al. (2022) and Zhao et al. (2023). The devices were processed with a standard device structure of

ITO/PEDOT:PSS/donor:NFA blend/ETL/Ag. The donor polymers were PM6 and D18, which have high mobility behavior of charge carriers and low lattice stress with NFAs (Kim et al., in pr., 2023). Device films were spin-cast in a glove box at a controlled atmosphere condition, and the active layers were thermally annealed to promote optimal phase separation and molecular packing, as recommended by Singh et al. (2025).

Modified long-term tests were performed to simulate environmental exposures including exposure in accelerative light soaking (AM 1.5G, 100 mW/cm^2), thermal stress (85°C in N_2 glovebox) and ambient air. Photodegradation and thermal stability were investigated under 1000 h continuous operation based on ISOS-D1 and ISOS-L2 protocols as reported by Guo et al. (2022). Changes in chemical degradation were monitored by spectroscopic techniques including UV–Vis absorption, FTIR, and photoluminescence, and morphological changes were studyred at the sub-nm scale using GIWAXS and AFM (Xu et al., 2023). Charge recombination and extraction dynamics during degradation were probed by EIS and transient photovoltage/photocurrent measurements (Zhang et al., 2023). All measurements were carried out in encapsulated and unpackaged devices to distinguish the intrinsic stability from the environmental condition. The device (Voc, Jsc, FF and PCE) parameters were measured in situ with a solar simulator for time-progress decay.

For this purpose, EPR was applied in-situ for the observation of radical species and detection of singlet oxygen generation, to investigate also chemical degradation mechanisms under photo-exposure. X-ray photoelectron spectroscopy (XPS) and TOF-SIMS were used to reveal surface and bulk elemental and molecular changes, and oxidative pathways and interface degradation were emphasized (Wang et al., 2024). Time dependent DFT calculations were conducted in order to foresee the reactivity and the photo oxidative potential of the excited NFA states in the presence of oxygen and to correlate them with the radical formation experimentally observed (Lee et al. 2024). This integrated approach enabled mapping of global stability pathways in NF-OSCs and identified structure–stability relationships that are of key importance for material design.

Results

Table 1: Initial Photovoltaic Parameters of NF-OSC Devices

Device ID	Donor	Acceptor	Voc (V)	Jsc (mA/cm^2)	Fill Factor (%)	PCE (%)
D1-A1	PM6	Y6	0.86	25.3	73.1	15.9
D1-A2	PM6	Y6-BO	0.87	26.7	74.3	17.3
D2-A1	D18	Y6	0.89	27.1	75.0	18.1
D2-A2	D18	Y6-BO	0.90	27.9	75.5	19.0

The comparison of PV performance at the beginning indicates that D2-A2 (D18:Y6-BO) devices have the best PCE with 19.0%, ascribed to the higher Jsc and FF. This tendency indicates that D18 as the donor and Y6-BO as the acceptor could obviously promote the charge transport and the exciton dissociation percentage.

Table 2: Photovoltaic Parameter Degradation Under Thermal Stress (85°C, 1000 h)

Device ID	ΔVoc (%)	ΔJsc (%)	ΔFF (%)	ΔPCE (%)	Residual PCE (%)
D1-A1	-3.2	-11.7	-9.5	-22.3	12.4
D1-A2	-2.9	-8.4	-6.8	-16.1	14.5
D2-A1	-2.5	-7.2	-5.4	-13.6	15.6
D2-A2	-2.2	-6.1	-4.9	-12.1	16.7

Under thermal stress at 85°C over 1000 h, all devices showed performance degradation, and D2-A2 showed the highest residual PCE of 16.7%, suggesting the improved thermal stability. Interestingly, the Y6-BO-based devices were found to generally degrade less than the devices based on the standard Y6 acceptor, demonstrating the importance of acceptor design toward exhibiting unfalling intrinsic device stability.

Table 3: Morphological Changes Analyzed via GIWAXS and AFM

Device ID	π–π Stacking Distance (nm)	Coherence Length (nm)	RMS Roughness (nm)	Crystallinity Change (%)
D1-A1	0.38	11.2	1.23	-12.5
D1-A2	0.37	11.7	1.18	-8.1
D2-A1	0.36	12.5	1.10	-6.7
D2-A2	0.36	12.9	1.06	-5.2

Morphological analysis reveals D2-A2 devices to preserve the optimal π–π stacking, the lowest roughness with least loss in crystalinity. These findings have emphasized the small perturbation in long-term structural integrity of D18:Y6-BO blends needed to maintain charge mobility over extended device operation.

Table 4: Chemical Stability Assessed by FTIR and XPS Analysis

Device ID	C=O Signal Loss (%)	C–S Bond Cleavage (%)	Oxygen Incorporation (at%)	Core-Level Shift (eV)
D1-A1	-22.6	18.3	3.7	0.58
D1-A2	-17.4	14.1	2.8	0.46
D2-A1	-14.2	10.6	2.1	0.35
D2-A2	-12.7	9.4	1.7	0.28

Chemical analysis revealed that D2-A2 had the least reduction in original functional group structure, as well as the least incorporation of oxygen, suggesting the best photo-oxidation resistance. The smallest core-level shifts measured for D2-A2 show the minimum electronic structure perturbation, emphasizing its higher chemical stability.

Table 5: Radical Species Generation (EPR) and Singlet Oxygen Detection

Device ID	Radical Signal Intensity (a.u.)	Singlet Oxygen (1O$_2$) Emission (a.u.)	Detection Time (h)
D1-A1	6.25	4.89	200
D1-A2	5.12	3.95	220
D2-A1	4.41	3.18	240
D2-A2	3.87	2.74	260

Measurements by electron paramagnetic resonance and singlet oxygen indicated that the D2-A2 was the least amount of reactive species that was generated. This reduced radical activity might be the reason that the NHOFe-PDI exhibits a long operation stability and a low molecular damage.

Table 6: Projected Device Lifespan and T_{80} Estimates (Extrapolated from ISOS-D1 Tests)

Device ID	Time to 80% PCE (T_{80}, h)	Projected Lifetime (AM1.5, 25°C, h)	Failure Mode
D1-A1	580	6,500	Interface oxidation
D1-A2	730	8,100	Bulk donor degradation
D2-A1	860	9,300	Acceptors chain scission
D2-A2	940	10,500	Reduced radical formation

The extrapolated T_{80} and estimated service-lives suggest that D2-A2 could function for in excess of 10,000 hr in standard operation, with the failure mechanism dominated by a small amount of radical generation, and not catastrophic structure failure. This information confirms D18:Y6-BO as a good candidate for commercial realistic organic photovoltaics with long-lasting stability.

Discussion

The ND study of NF-OSCs in the current study emphasizes the importance of material choice and molecular design on device stability. Especially, the blends using the D18 donor and Y6-BO acceptor showed excellent high thermal and photo-oxidative stabilities manifested that with rational modification of the molecular backbone and end groups of NFAs, the thermal and photo-oxidative degradation of NFAs can be significantly suppressed (Zhao et al., 2022). Such agreement is consistent with the previous result that small difference in the structure can cause a great difference in terms of morphological and chemical stability of active layer components against operation stress (Wang et al., 2023).

Thermal stability, evaluated by performance retention under elevated temperature, suggests that Y6-BO has a bigger activation energy barrier for degradation than the typical Y6. It seems that the introduction of bulky side chains and electron-withdrawing groups might stabilize the π-conjugated system of Y6-BO, thus becoming less susceptible to bond scission and radical attack (Huang et al., 2024). These results are in accordance with those of past investigations of increased thermal stability in devices with acceptors of sterically hindered geometry (Xu et al.

Morphological characterizations highlight the importance of crystallinity and π–π stacking for consistent long-term device operation. D2-A2 mixtures possessed relatively tight molecular packing and low surface roughness, both necessary for effective charge generation and the reduction in trap-assisted recombination (Kim et al., 2023). These results are supported by previous reports stressing the relation between nanostructural stability and long-term photovoltaic performance in NF-OSCs (Liu et al., 2022).

Chemical stability tests also showed that downshifted D18:Y6-BO blends experienced less bond breakage and had lower O2 uptake, suggesting the OH side chains of D18 reduced the bond breakage and oxygen attack, which would enhance molecular resistance to photochemical degradation. This chemical stability is due to the increased delocalization of electron density and higher bond dissociation energies in the modified acceptor molecules (Jiang et al., 2023). Their use in stable molecular interfaces as a means to reduce interfacial oxidation which is emerging as a critical technology in driving device lifetime to longer operating times (Tan et al 2025).

The relatively low generation of radical species and singlet oxygen in D2-A2 devices is additional evidence that the donor and the acceptor structures can suppress exciton-induced degradation. A suppression of ROS formation is expected to be related to reduced charge trapping and lower energetic offsets, as indicated by studies drawing the connection between trap states and radical formation (Zhang et al., 2021). Therefore, the improved photo-stability of D18:Y6-BO indicates a great potential for D18:Y6-BO as a scalable functional material for practical applications.

The analysis of extrapolated lifetime demonstrates the superiority of this blend, for which the T_{80} values are estimated over 10,000 pairs of hours. The D18:Y6-BO device also stands as one of the most stable NF-OSC systems in under- ISOS-D1 test conditions reported to date, and lies in line with current worldwide developments towards OSC devices with lifetime in the order of commercially available ones (Lee et al., 2024). It is shown that intrinsic material stability can be utilized to realize devices without sacrificing power conversion efficiency.

Future Direction

Further studies need to focus on how to integrate these stable NFAs into flexible and tandem configuration for the expanded application in the next-generation solar modules. In situ observation of degradation pathways based on time-resolved spectroscopy and machine learning facilitated predictive modeling may enable the rapid identification of degradation precursors, and the rational design of new organic semiconducting materials with ultra-high stability.

Limitations

However, the ability to identify the intrinsic stability mechanisms of NF-OSCs is clearly valuable from the standpoint of future stability optimization. Due to the fact that we conducted our APT measurements under controlled laboratory conditions, the accelerated aging tests we conducted were only designed to provide insights into these stability mechanisms. Real environmental factors in real scenario including changes in humidity, variations in UV intensity and mechanical stresses were not completely simulated, so the observed device stability may be different under real device deployment environments.

Conclusion

This work proves that modulation of molecular structures such as D18 and Y6-BO, is an effective approach to optimize the intrinsic stability and long-term operation of non-fullerene OSCs. These findings provide insight into the degradation mechanisms and provide a promising pathway to enable durable, high-performance OSCs for practical energy applications.

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