nature portfolio

Peer Review File

Intrinsically Stretchable Fully π-Conjugated Polymers with Inter-aggregate Capillary Interaction for Deep-blue Flexible Inkjet-printed Light-emitting Diodes

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this paper, the authors synthesized anisotropic shape pi conjugated fluorescence polymers for ink-jet printed OLEDs. The main issue in the ink-jet printing technology is “coffee ring” formation during the solvent evaporation process. Coffee ring formation lead to poor film quality. The authors claimed that anisotropic shape pi conjugated fluorescence polymers inhibit the “coffee ring effect”. Unfortunately, this manuscript does not offer any conceptual novelties, and results are not conclusive. Also, there are number of other issues as listed below:

1. What is polydispersity of the polymers. This information is missing. Different polymer chain length can dramatically influence solubility, morphology and hence the thin film quality. Therefore, suppression of “coffee ring effect “due to anisotropic shape of polymer is not conclusive. The authors should provide various control experiments that include effect on molecular weight, polydispersity on coffee ring effect. This will provide conclusive evidence of polymer anisotropic shape with coffee ring effect.

2. What is solubility of theses polymers?

3. What is the stability of the ink?

4. The PLQY of the films are quite low < 50% compared to the state-of the art electroluminescent polymers. This led to very poor OLED device performance < 1% EQE. Is the low eqe is due to poor recombination of electrons or holes?

5. What is the uncertainty of OLED devices characteristics? Device statistics is missing.

6. Unfortunately, the materials developed in this work and their device performance are quite poor compared to the reported work. The time resolved absorption studies and mobility data do not add any insight to the proposed claim and the results are not conclusive. Therefore, I can’t recommend this paper for publication in high impact journal such as nature comm.

Reviewer #2 (Remarks to the Author):

This work proposed an asymmetric substitution strategy to suppress coffee-ring effect and improve the stretchability of deep-blue light-emitting FπCPs for flexible printed PLEDs, namely PFPO and POPOF. The reason behind is the intrinsic stretchability with a strong inter-aggregate capillary interaction. The results are noteworthy and significant. The work mostly supports its conclusion, and the methodology is sound. There is area also enough details for the work to be reproduced. Therefore, this work can be considered for publication after a minor revision by addressing the following comments.

1. The author should rearrange the introduction so that the logic flow is easy to understand. For example, PFFELDs and FπCPs can be introduced first; then the coffee ring effect and intrinsic

stretchability is explained as a difficulty; finally, methodologies to overcome them is proposed, followed by what is achieved in this work. The current introduction is divided into two parts, and the coffee ring effect is mentioned twice as a difficulty, so that the main ideas are hard to catch.

2. The author needs to explain why the variation of viscoelasticity of PFPO in Figure 1a is so large (only this data point has such a huge error bar).

3. The caption of Figure 4 is missing (c) and (d). Please check other typos as well.

4. To demonstrate the stretchability, the author should provide current density and luminance data of the device under stretching conditions.

5. The author needs to compare the efficiency and brightness of the materials proposed in this work with other representative publications.

Reviewer #3 (Remarks to the Author):

In this manuscript asymmetric substitution of conjugated polymers is applied to avoid the coffee stain effect. Three different blue-emitting polyfluorene based polymers with different substitutions are investigated. The difference in printed patterns and mechanical properties is attributed to differences in packing, where a loose packing is beneficial. However, a loose packing is detrimental for charge transport, resulting in very low carrier mobilities. For PLEDs, printability is only of the factors to for commercialization, next to efficiency and stability. In this work, only the first is addressed. Regarding the last two, the performance of the PLEDs is very poor, with EQEs below 1%. Also the PLQY of the polymers presented here is low (35-40%)
To get a better judgement of the achievements, it would be good to make a comparison with a standard polymer as SY-PPV. Like many other emitting polymers it is also asymmetrically substituted and does not crystallize. It has a PLQY of 65% and PLEDs have an EQE of ~4%, which is ten times better than PFPO reported here. Of course, one can argue that it is not fair to compare the performance of a PLED of a yellow emitter with a blue-emitting material, but a comparison of their printability and stretchability could be made, to demonstrate whether the properties realized here are really better.

On a side note, the low PLED performance is, next to the low PLQY also, also the result of a bad hole injection contact. The work function of PEDOT:PSS of ~5.1 eV is far off from the HOMO of the materials of ~6 eV.

Given the very modest optical and electrical properties of the polymers presented here the paper does not meet the high standards of Nat. Comm. with regard to the printability and stretchability a comparison with other asymmetrically substituted polymers should be given.

Responses to the Comments of Referees:

Reviewer 1:

In this paper, the authors synthesized anisotropic shape pi conjugated fluorescence polymers for ink-jet printed OLEDs. The main issue in the ink-jet printing technology is “coffee ring” formation during the solvent evaporation process. Coffee ring formation lead to poor film quality. The authors claimed that anisotropic shape pi conjugated fluorescence polymers inhibit the “coffee ring effect”. Unfortunately, this manuscript does not offer any conceptual novelties, and results are not conclusive. Also, there are number of other issues as listed below:

Answer: Thank you for your comments.

We are also sorry to take your so many troubles for our mistakes. According to your comments, we have corrected our mistakes and polished the whole manuscript. We also added many experiments to support our assumption and results in this revised manuscripts. Therefore, we sincerely hope that you can re-consider our revised manuscript and agree to accept and publish our work in *Nature Communications* journal.

Based on your kind comments, we also provided some detail discussion about the novelty and objectives of our work again as follow:

(1) Conceptual novelties. Emerging printed and flexible electronic devices provided an infinite possibility of artificial optoelectronic components. During the large-area inkjet-printed process, the generation of coffee rings can lead to uneven deposition of the active film, thereby affecting the pattern resolution, device performance and stability. Besides, since flexible optoelectronic devices are highly deformable, active nano-layers in these devices experienced different degrees of tensile and compress strains during the bending, stretching, compressing, and twisting of the devices, which may cause the obvious intralayer fracture and interlayer separation to further result into device failure. These two serious problems are the major obstacles to manufacture the printed and flexible device for the practical electronic equipment. Therefore, obtaining the intrinsically viscoelastic optoelectronic materials with a strong capillary interaction is an effective strategy to obtain a capacity of the suppression of coffee-ring effect and excellent stress tolerance ability for printed and flexible optoelectronics applications. In this work, we propose **an asymmetric substitution strategy to improve the printable and stretchable property of deep-blue light-emitting fully π-conjugated polymers (FπCPs) with a strong inter-aggregate capillary interaction for flexible inkjet-printed polymer light-emitting diodes (PLEDs)**. In a word, we believe that **this is the first work to simultaneously obtain excellent intrinsically inkjet-printable and stretchable capacity of FπCPs for the flexible and printed electronics**. Meanwhile, this work also is the first study to systematically and intuitively tune the interchain aggregation of FπCPs in inkjet-printed ink to suppress coffee rings, induce uniform deposition of thin films and enhance viscoelastic properties, and make our research a milestone in the fabrication of flexible and printed deep-blue LEDs. **The performances of flexible deep-blue PLEDs based on our stretchable film are comparable to the reported ones in *Nature* 2022, 624, 603. *Nat. Mater.*, 2023, 22, 737-745. *Sci. Adv.*, 2023, 9, eadh1504.** Therefore, our work will attract widely and broad attentions in flexible and printed electronics, organic photonics, polymer chemistry, polymer and materials science. The topic and unique conclusion of this work match the research area of “***Nature Communications***”.

(2) Convinced results. The intrinsically viscoelastic FπCPs with an anisotropic shape

aggregate is an effective strategy to obtain strong capillary interactions and avoid the coffee ring effect for printed and flexible optoelectronics applications. The anisotropic shape of the aggregate or particles significantly deforms interfaces, producing strong capillary interactions, which is beneficial for suppressing the formation of coffee ring. In this work, **the asymmetric substitution of novel deep-blue FπCPs can disrupt chain regularity and ordered crystallization capacity of FπCPs, and is favorable for obtaining excellent stretchable properties to enhance the device deformation stability and stress tolerance ability.** As we expected, the elongation at break of freestanding asymmetric FπCPs films (smaller l_{ps}: 6 nm) reached 53% and 72%, much higher than traditional ones (11%), indicating their outstanding intrinsic stretchability and excellent capacity of stress tolerance. This is the key factor to ensure the long-time deformation operation stability of flexible electronic devices. More interestingly, compared to the dense isotropic Gaussian chain stacking of traditional FπCPs, loose anisotropic rod-shaped aggregate of asymmetric FπCPs with a strong capillary interaction is more likely to present a uniform printed deposition, which is beneficial for the formation of high-quality large-area inkjet-printed films. Therefore, flexible and inkjet-printed deep-blue PLEDs were also manufactured with a stable and efficient deep-blue emission (CIE of (0.17, 0.10)) and maximum brightness of 3100 cd/m^2, with comparable performance to that of spin-coated devices. **Even after stretching to 50% strain, the PLED based on the stretchable FπCPs films also displayed a stable device performance, indicated their excellent deformation stability and energy dissipation capacity.** Considering the widespread potential applications of deep-blue flexible and inkjet-printed PLEDs, it is believed that this work will promote the application of PLED in flexible full color displays and wearable devices.

(3) High efficiency flexible inkjet-printed PLED based on stretchable film with excellent deformation stability. We are agreed with you that many high standard works about the flexible OLED are reported and published in the last several years, such as *Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504. However, all these works above are focused on exploring the yellow flexible OLEDs based on the stretchable films. Performance of deep-blue (CIEy< 0.05) emissive stretchable film for the flexible OLEDs needs to further explore. For example, the EQE of blue OLED based on the stretchable films (current efficiency (C. E.) < ~1.6 cd/A and **CIE (0.23, 0.32)** in *Nature* **2022**, 624, 603; TADF materials, EQE< ~**0.8%, in** *Nat. Mater.*, **2023**, *22*, 737-745; C. E. < ~**0.3 cd/A** in *Sci. Adv.*, **2023**, 9, eadh1504.), as displayed in Figure 1. Performance of blue OLED based on stretchable film reported in these high standard journals are very low, much lower than the EQE of our device. We also reasonably believed that the performance of flexible printed OLED based on these stretchable film may further decrease, due to their complicated interchain aggregation and phase separation. In this regard, the EQE of our flexible inkjet-printed deep-blue OLEDs based on our **stretchable film is calculated about 1.39%, much higher than those in these reported works (***Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504.**). More important, even at stretching to 50% strain, the PLED based on the elastic FπCPs films also displayed a stable electroluminescence property and comparable efficiency without any attenuation, demonstrating the excellent dynamic strain tolerance of FπCPs stretchable nanolayers.** Interchain π-π interactions in polymeric materials have a fundamental role in a variety of processes spanning from charge transport to the photophysics of neutral excited species. For example, π-π stacking is essential for high mobility in polymer-based field-effect transistors,

whereas formation of intermolecular excited states in luminescent materials may lead to both reduced photoluminescence efficiency, and a reduced energy gap. These are undesirable, especially for fabrication of blue-emitting devices (and therefore for full-colour displays). Even where interactions are not strong enough to lead to formation of aggregates, close-packing of the chromophores may significantly influence the molecular geometry, altering the colour purity and the emission efficiency. Therefore, **it is reasonably concluded that the complicated interchain aggregation can observe in the rod-coil copolymers and blending materials systems, which may cause the formation of multi-chain excited states and non-uniform morphology. These can further result into low device efficiency, instable EL spectra, poor color purity and non-uniform emission. In a word, to obtain intrinsically stretchable homo-polymer is an effective and convenient strategy to achieve a high performance flexible deep-blue OLEDs.**

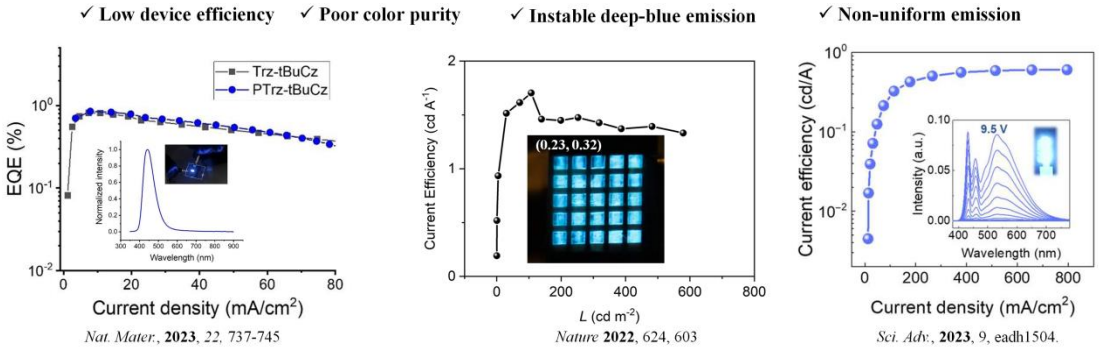


Figure 1. Efficiency of flexible blue OLED based on stretchable film in *Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504.

1. What is polydispersity of the polymers. This information is missing. Different polymer chain length can dramatically influence solubility, morphology and hence the thin film quality. Therefore, suppression of "coffee ring effect "due to anisotropic shape of polymer is not conclusive. The authors should provide various control experiments that include effect on molecular weight, polydispersity on coffee ring effect. This will provide conclusive evidence of polymer anisotropic shape with coffee ring effect.

Answer: Thank you for your comments.

Polymers are mixtures of homologous molecules with different molecular chain lengths. Its molecular weight is the average value of homologues. This non-uniformity in molecular weight is called molecular weight polydispersity. We have supplemented the PDI values in the Supplementary Information. The PDIs of the three polymers in the manuscript are 1.8, 1.7 and 1.9 respectively. The three PDIs are very close. At the same time, their conjugated backbones are the same. By calculating the main chain lengths of PODPF and POPOF, they are close. Therefore, this work can better reflect the impact of changes in polymer stacking methods on coffee rings by controlling variables.

Thank you for your comments and suggestions again.

2. What is solubility of theses polymers?

Answer: Thank you for your comments.

The solubility of the three polymers in toluene can reach more than 50 mg/ml. In fact, for inkjet

printing, higher solubility is not always better. The higher the concentration of the solution, it is easy to cause the nozzle to become clogged. In this work, we only used toluene solutions with a concentration of 5 mg/ml.
Thank you for your comments and suggestions again.

3. What is the stability of the ink?
Answer: Thank you for your comments.
Our ink (5 mg/mL toluene solution) shows good stable emission behavior and inkjet-printable capacity. After the ink was placed in the dark for 2 months, there was no gelling or precipitation, and present a robust deep-blue emission property.

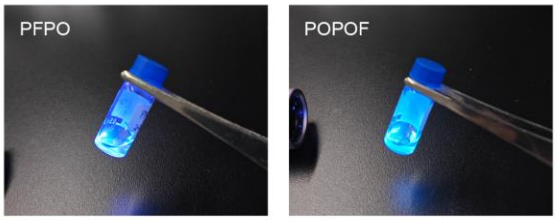


Figure 2. Photo images of two polymer ink under UV lamp (365 nm).

Thank you for your comments and suggestions again.

4. The PLQY of the films are quite low < 50% compared to the state-of the art electroluminescent polymers. This led to very poor OLED device performance < 1% EQE. Is the low eqe is due to poor recombination of electrons or holes?
Answer: Thank you for your comments.
We optimized the device fabrication process and updated the device data (Figures 4b-4c in the manuscript) in the revised manuscript as follows:

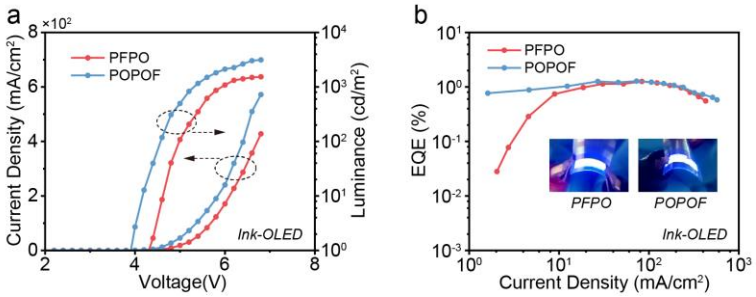


Figure 3. Device performance of flexible OLED on our novel polymers.

The turn-on voltages (V_{on}) of PFPO and POPOF-based flexible inkjet-printed PLEDs were 4.4 V and 4.0 V. The maximum brightness is about 1500 cd/m^2 at 6.8 V and 3100 cd/m^2 at 6.8 V for printed PLEDs of PFPO and POPOF. The maximum external quantum efficiencies of PFPO and POPOF are 1.26% and 1.28%, respectively. This is at the forefront in terms of intrinsic stretchable blue fluorescent emitters. As we discussion above, the EQE of our flexible OLEDs are comparable to reported ones published in *Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504., as showed in Figure 1.

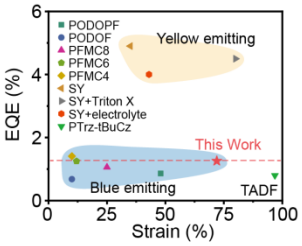


Figure 4. Comparison of the EQE and stretchability given by PFPO (this work) with the corresponding values for previously reported intrinsically stretchable light-emitting polymers based on fluorescence emitters.

It is a fact that our PLEDs present a relatively low efficiency, although it had an excellent stable deep-blue emission. In our opinion, there are some key factors need to consider as follow. We also provided some explanation in this revised manuscript.

1.Unmatched energy level. In general, these flexible conjugated structure result a large bandgap and low HOMO enery level. Low HOMO level caused the unbalance electron and hole injection and result into low charge reconbination.

2. The cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport. In fact, the cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport, which is negative effect on the radiative recombination of holes and electrons in the emissive layer, thus shows a lower EQE. Then, it is ergent to obtain intrinsically stretchable and stable deep-blue conjugated polymer in futures.

3.Configuration structure of our PLEDs need to optimize and further enhance the recombination of electron and hole injection and transport. We need to admit that poor device fabrication capacity is an obstacle to obtain high performance and stable PLEDs. The delicate balance of electrons and holes in the emissive layer for radiative recombination is the key to device efficiency and stability.

Therefore, we are very appreciated your comments about our work. In order to obtain high performance PLEDs, we need to resolve the key problem above. Meanwhile, in our lab, recently, we also obtained a series of high performance and stable deep-blue PLEDs based on the intrinsically stretchable and stable polyfluorene materials. We hope that we can publish some high quality works in these area in future.

Thank you for your comments and suggestions again.

5. What is the uncertainty of OLED devices characteristics? Device statistics is missing.

Answer: Thank you for your comments and suggestions. We are so sorry for these mistakes. In the revised manuscript, we have added device statistics in the supplementary information (Figure S32) as follows:

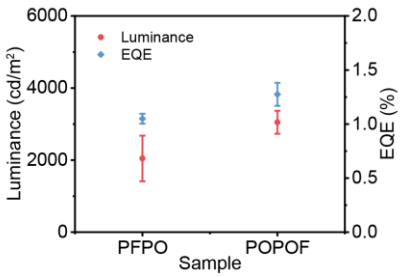


Figure 5. Device statistics of OLED based on two polymers.

Thank you for your comments and suggestions again.

6. Unfortunately, the materials developed in this work and their device performance are quite poor compared to the reported work. The time resolved absorption studies and mobility data do not add any insight to the proposed claim and the results are not conclusive. Therefore, I can't recommend this paper for publication in high impact journal such as nature comm.

Answer: Thank you for your comments and suggestions. In the revised manuscript, we have provided a comparison in the scheme 1f with other reported intrinsically stretchable blue light polymer materials, as well as with super yellow.[1, 2, 3, 4] The materials reported in this work are at a high level in terms of tensile properties. For EQE, it is at the forefront of intrinsically stretchable blue-emitting polymers.

We are agreed with you that many high standard works about the flexible OLED are reported and published in the last several years, such as *Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504. However, all these works above are focused on exploring the yellow flexible OLEDs based on the stretchable films. Performance of deep-blue (CIEy< 0.05) emissive stretchable film for the flexible OLEDs needs to further explore. For example, the EQE of blue OLED based on the stretchable films (current efficiency (C. E.) < ~1.6 cd/A and **CIE (0.23, 0.32)** in *Nature* **2022**, 624, 603; TADF materials, EQE< ~**0.8%, in** *Nat. Mater.*, **2023**, *22*, 737-745; C. E. < ~**0.3 cd/A** in *Sci. Adv.*, **2023**, 9, eadh1504.), as displayed in Figure 1. Performance of blue OLED based on stretchable film reported in these high standard journals are very low, much lower than the EQE of our device. We also reasonably believed that the performance of flexible printed OLED based on these stretchable film may further decrease, due to their complicated interchain aggregation and phase separation. In this regard, the EQE of our flexible inkjet-printed deep-blue OLEDs based on our **stretchable film is calculated about 1.39%, much higher than those in these reported works. More important, even at stretching to 50% strain, the PLED based on the elastic FπCPs films also displayed a stable electroluminescence property and comparable efficiency without any attenuation, demonstrating the excellent dynamic strain tolerance of FπCPs stretchable nanolayers.** Interchain π-π interactions in polymeric materials have a fundamental role in a variety of processes spanning from charge transport to the photophysics of neutral excited species. For example, π-π stacking is essential for high mobility in polymer-based field-effect transistors, whereas formation of intermolecular excited states in luminescent materials may lead to both reduced photoluminescence efficiency, and a reduced energy gap. These are undesirable, especially for fabrication of blue-emitting devices (and therefore for full-colour displays). Even where interactions are not strong enough to lead to formation of aggregates, close-packing of the chromophores may significantly influence the molecular

geometry, altering the colour purity and the emission efficiency. Therefore, **it is reasonably concluded that the complicated interchain aggregation can observe in the rod-coil copolymers and blending materials systems, which may cause the formation of multi-chain excited states and non-uniform morphology. These can further result into low device efficiency, instable EL spectra, poor color purity and non-uniform emission. In a word, to obtain intrinsically stretchable homo-polymer is an effective and convenient strategy to achieve a high performance flexible deep-blue OLEDs.**

As your comment about the milestone work in the fundamental research of the stretchable OLEDs, Bao and co-authors demonstrated the red, green and blue stretchable OLEDs via blending the polymeric emitter with elastic matrix in recent work (Nature, 2022, 624, 603). We are agreed with you that this work is very important in this area. As your comment, blue emission has already been realized for stretchable OLEDs. This work provided an effective strategy to realize the fabrication of flexible optoelectronic device via blending elastic polymer. In fact, it is a common sense that **the key performance parameters of blue OLEDs, both rigid and flexible ones, are the EL spectral stability, color purity, emission uniformity and efficiency, rather than the realization of blue emission. As presented in this work** (Nature, 2022, 624, 603), all key parameters of deep-blue stretchable OLEDs need to further enhance, such as EL spectral stability, color purity, emission uniformity and efficiency, as displayed in Figure 1a-1b. In fact, blend strategy is a universal and convenient method to obtain stretchable active film for flexible optoelectronics, but this method is not suitable for the fabrication of uniform deep-blue stretchable OLEDs. In a word, EQE of the OLED based on our novel materials are comparable to those of OLED based on **stretchable film (> 60% strain).**

As your comment, our device show the very moderate level of EQE values of 1.39% for the flexible inkjet-printed OLEDs. We are agreed with your device efficiency need to further improve. We also believed that the efficiency can be also further enhanced by optimizing device configuration. In general, the EQE of PLEDs is determined by the following equation: $EQE = \phi \times \gamma \times \eta_r \times \eta_{out}$, where ϕ, γ, η_r, and η_{out} represent the quantum efficiency, carrier balance factor, singlet exciton generation rate, and light extraction efficiency, respectively. Due to the high band exciton and wide bandgap, intrinsic low charge density and mobility of blue emitters may also result into a low device efficiency and stability. Therefore, it is a serious challenge to obtain efficient deep-blue flexible OLEDs.

Thank you for your comments and suggestions again.

References

1. Liang JJ, Li L, Niu XF, Yu ZB, Pei QB. Elastomeric polymer light-emitting devices and displays. *Nat Photonics* 2013, 7(10): 817-824.
2. Kim JH, Park JW. Intrinsically stretchable organic light-emitting diodes. *Sci Adv* 2021, 7(9): 9715.
3. Zhuo Z, Ni M, An X, Bai L, Liang X, Yang J, et al. Intrinsically Stretchable and Efficient Fully Pi-Conjugated Polymer via Internal Plasticization for Flexible Deep-Blue Polymer Light-Emitting Diodes with CIE(y) = 0.08. *Adv Mater* 2023: e2303923.
4. An X, Gong H, Chen W, Zhuo Z, Wei C, Sun N, et al. VPERD Assessment of the Mechanical Property of Plasticizing Fully π-Conjugated Polymers with Intrinsic Stretchability for Flexible Deep-Blue Light-Emitting Diodes. *Adv Optical Mater* 2024.

Finally, thank you for your all kind comments and suggestions. We have modified all incorrect sentences in this revised manuscript. We sincerely hope that our paper can be accepted and published in *Nature Communications*.

Reviewer 2:

Reviewer #2: This work proposed an asymmetric substitution strategy to suppress coffee-ring effect and improve the stretchability of deep-blue light-emitting FπCPs for flexible printed PLEDs, namely PFPO and POPOF. The reason behind is the intrinsic stretchability with a strong inter-aggregate capillary interaction. The results are noteworthy and significant. The work mostly supports its conclusion, and the methodology is sound. There is area also enough details for the work to be reproduced. Therefore, this work can be considered for publication after a minor revision by addressing the following comments.

Answer: Thank you for your comments.

We are also sorry to take your so many troubles for our mistakes. According to your comments, we have corrected our mistakes and polished the whole manuscript. Therefore, we sincerely hope that our revised manuscript will be accepted and published in *Nature Communications* journal.

1. The author should rearrange the introduction so that the logic flow is easy to understand. For example, PFFELDs and FπCPs can be introduced first; then the coffee ring effect and intrinsic stretchability is explained as a difficulty; finally, methodologies to overcome them is proposed, followed by what is achieved in this work. The current introduction is divided into two parts, and the coffee ring effect is mentioned twice as a difficulty, so that the main ideas are hard to catch.

Answer: Thank you for your comments and suggestions. According to your comments, we have reorganized the logical sequence of the introduction section to enhance readability in this revised manuscript as follow:

“Emerging printed and flexible electronics technology provided a powerful engine to promote the development and innovation of modern electronic equipment, such as high-quality displays and large-area solid-light, bionics electronic skin, and parabrain circuits. Flexible printed polymer light-emitting diode (FPPLED) is an essential component of flexible display equipment owing to their low-cost processing, potentially mechanical flexibility, and color tenability. As the critical element in flexible display, the morphological uniformity and optoelectronic batch stability of printed light-emitting polymer layers is prerequisite to ensure their outstanding performance and excellent operational stability. As a typical light-emitting polymer, traditional fully π-conjugated polymers (FπCPs) chains always easily tend to self-assemble into rigid and brittle semi-crystalline structure, due to strong face-to-face π-π interaction, which result into extremely weak inter-chain entanglement with low viscosity in the precursor ink, and lead to the formation of coffee rings. In fact, the ubiquitous nature of the coffee-ring effect has made it difficult to avoid in inkjet-printed processing of FπCPs for the fabrication of FPPLED. The generation of coffee-rings often results in uneven deposition of printed films, affecting the resolution of the patterns and the optoelectronic performance of the flat panel displays. In addition, independent aggregates produce uneven deposition by capillary flow, inducing the complicated photo-physics of the interchain excited species and altered the color purity and the emission efficiency, which is undesirable for the fabrication of deep-blue PLEDs. Up to date, many physical and engineering strategies have been implemented to inhibit coffee rings, such as doping surfactants in solvents, using mixed viscous solvents, changing the substrate's surface energy, etc. which is not suitable for the deep-blue printed PLEDs because of their low capacity of defect tolerance. Therefore, rationally designing deep-blue emitting FπCPs to weaken the capillary flow during the evaporation process of ink droplets and suppress the formation of coffee rings is the key to realizing large-area deep-blue

flexible PLEDs manufactured by inkjet printing.

In the printed processing, the weak inter-aggregate or intermolecular interactions caused a serious capillary flow outward from the center of the ink drop, bring suspended aggregate and molecules to the edge and form the coffee ring during evaporation proceeds (Scheme 1a). Interestingly, the anisotropic shape of the aggregate or particles significantly deforms interfaces, producing strong capillary interactions, which is beneficial for suppressing the formation of coffee ring (Scheme 1b). Besides, FπCPs with potential intrinsic viscoelasticity can reasonably weaken capillary flow through strong inter-chain entanglement and inhibit the formation of coffee rings, making them ideal candidate materials for printed electronic devices. Therefore, obtaining the intrinsically viscoelastic FπCPs with an anisotropic shape aggregate is an effective strategy to obtain strong capillary interactions and avoid the coffee ring effect for printed and flexible optoelectronics applications. Here, we used an asymmetric substitution strategy to design and synthesize intrinsically stretchable FπCPs with a strong inter-aggregate capillary interaction to fabricate flexible printed deep-blue PLEDs. In solution, the anisotropic shape aggregate with a relatively loose stacking is more likely than dense Gaussian chain stacking to induce uniform deposition of polymers and form high-quality films during the evaporation of the solution. At the same time, this loose stacking makes it easier for the molecular chains to slip under the action of external force, resulting in the polymer having better tensile properties (Scheme 1e). Therefore, designing FπCPs with loose anisotropic aggregate is an effective strategy to prepare active layers for fully printed flexible devices."

Thank you for your comments and suggestions again.

2. The author needs to explain why the variation of viscoelasticity of PFPO in Figure 1a is so large (only this data point has such a huge error bar).

Answer: Thank you for your comments and suggestions.

In fact, there are many factors that affect the viscosity of polymers, such as molecular weight, solvent, etc. Since PFPO has a relatively large molecular weight, it has a higher viscosity. For errors, we only truthfully reflect the experimental conditions. Compared with PODPF and POPOF, we believe that the large change in PFPO viscosity comes from the polydispersity of the polymer itself on the one hand, and the error generated during the testing process on the other hand. However, this does not affect our comparison of the viscosity trends of the three materials.

Thank you for your comments and suggestions again.

3. The caption of Figure 4 is missing (c) and (d). Please check other typos as well.

Answer: Thank you for your comments and suggestions. We are so sorry for these mistakes. In the revised manuscript, we have updated Figure 4 and corrected the aforementioned errors as follows:

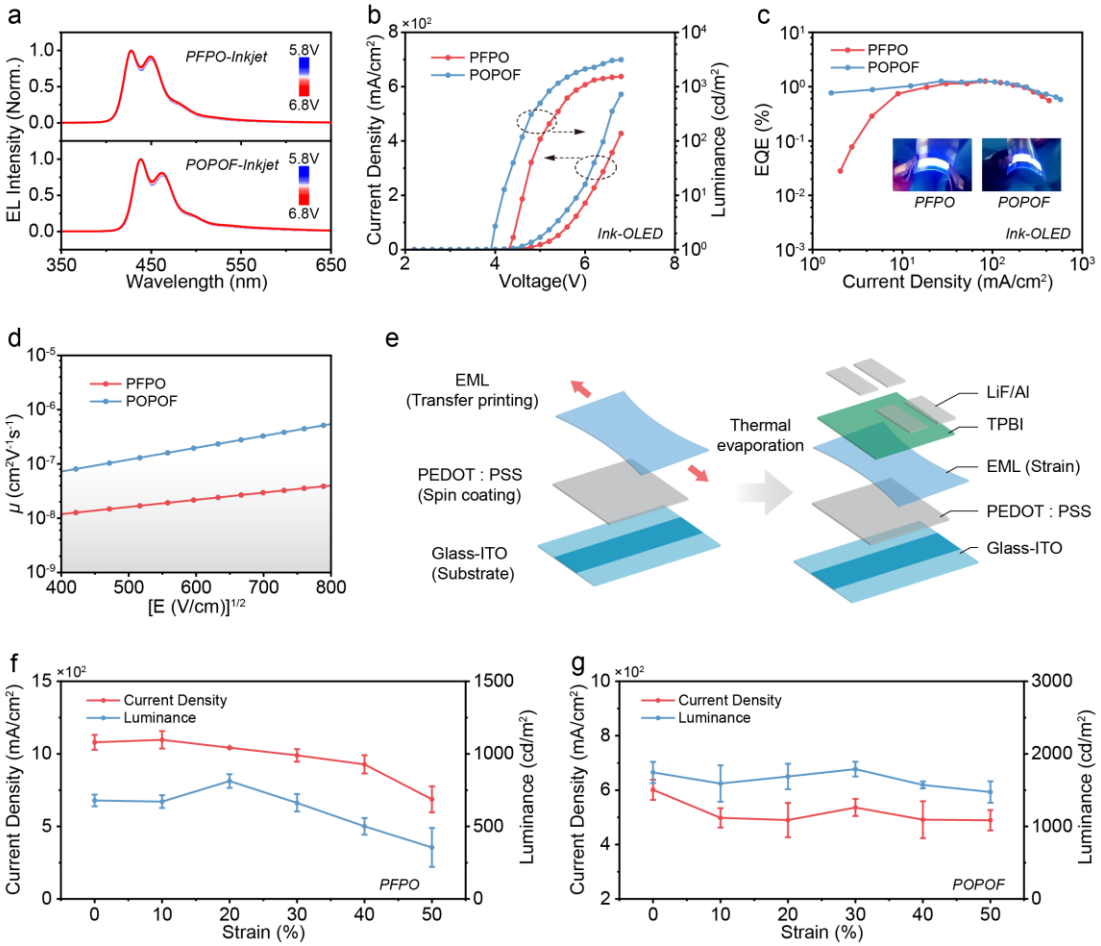


Figure 1. Performance of PFPO and POPOF-based flexible inkjet-printed PLEDs. (a) EL spectra of inkjet-printed PLEDs at different applied voltages. (b) *J-V-L* curves of corresponding inkjet-printed PLEDs. (c) EQE and (d) carrier mobility characterization of inkjet-printed PLEDs. Insets are optical photos of their flexible device prepared by inkjet printing. (e) Schematic diagram of preparation of emitting layer stretching device. Relative highest current density and luminance statistics of PLEDs using (f) PFPO and (g) POPOF films with increased strain (data are represented as mean values ± s.d. from four devices).

Thank you for your kind comments again.

4. To demonstrate the stretchability, the author should provide current density and luminance data of the device under stretching conditions.

Answer: Thank you for your comments and suggestions. We are so sorry for this mistake. In the revised manuscript, we have provided the brightness and current density of the film under stretching state in Figure 4f-g, with an explanation of the data, as follows:

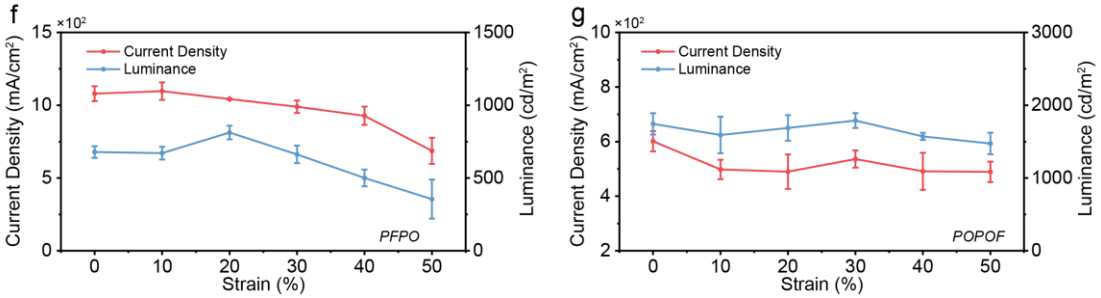


Figure 2. Relative highest current density and luminance statistics of PLEDs using PFPO and POPOF films with increased strain (data are represented as mean values ± s.d. from four devices).

"Under normal circumstances, the molecular chain conformation and stacking mode changes as the film is stretched, which can affect the device performance. To confirm the practical application of PFPO and POPOF in intrinsically stretchable PLEDs, PLEDs at different stretching degrees were prepared using transfer printing technology (Figure 4e). As shown in Figure S29, both PFPO and POPOF devices exhibit excellent spectral stability as the strain increases, which is crucial for stretchable devices. Additionally, the strain-brightness-current density curves are shown in Figure 4f, 4g. The current density of PFPO is almost constant at low strain but exhibits a slow decline at high strain, attributed to subtle defects generated in the PFPO film at high strain. Interestingly, brightness of PFPO increases at low strain, reaching a maximum of 800 cd/m² at 20% stretch (Figure S30). This is attributed to a more balanced carrier injection and improved interface contact. Subsequently, brightness of PFPO shows a decreasing trend due to a reduction in the effective emitting area caused by the generation of subtle defects in the film. It is worth noting that at 50% stretch, both the current density and brightness of POPOF demonstrate good stability. This once again supports the idea that the more compact molecular chain stacking and the inhibition of cyclic main chain formation in POPOF enhance charge transfer capabilities, resulting in superior stretch stability of optoelectronic performance."
Thank you for your kind comments again.

5. The author needs to compare the efficiency and brightness of the materials proposed in this work with other representative publications.
Answer: Thank you for your comments and suggestions. In the revised manuscript, we have provided a comparison in the scheme 1f with other reported intrinsically stretchable blue light polymer materials, as well as with super yellow.[1, 2, 3, 4] The materials reported in this work are at a high level in terms of tensile properties. For EQE, it is at the forefront of intrinsically stretchable blue-emitting polymers.

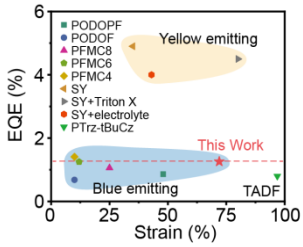


Figure 3. Comparison of the EQE and stretchability given by PFPO (this work) with the corresponding values for previously reported intrinsically stretchable light-emitting polymers based on fluorescence emitters.

Thank you for your comments and suggestions again.

References

1. Liang JJ, Li L, Niu XF, Yu ZB, Pei QB. Elastomeric polymer light-emitting devices and displays. *Nat Photonics* 2013, 7(10): 817-824.

2. Kim JH, Park JW. Intrinsically stretchable organic light-emitting diodes. *Sci Adv* 2021, 7(9): 9715.
3. Zhuo Z, Ni M, An X, Bai L, Liang X, Yang J, et al. Intrinsically Stretchable and Efficient Fully Pi-Conjugated Polymer via Internal Plasticization for Flexible Deep-Blue Polymer Light-Emitting Diodes with CIE(y) = 0.08. *Adv Mater* 2023: e2303923.
4. An X, Gong H, Chen W, Zhuo Z, Wei C, Sun N, et al. VPERD Assessment of the Mechanical Property of Plasticizing Fully π-Conjugated Polymers with Intrinsic Stretchability for Flexible Deep-Blue Light-Emitting Diodes. *Adv Optical Mater* 2024.

Finally, thank you for your all kind comments and suggestions. We have modified all incorrect sentences in this revised manuscript. We sincerely hope that our paper can be accepted and published in *Nature Communications*.

Reviewer 3:

In this manuscript asymmetric substitution of conjugated polymers is applied to avoid the coffee stain effect. Three different blue-emitting polyfluorene based polymers with different substitutions are investigated. The difference in printed patterns and mechanical properties is attributed to differences in packing, where a loose packing is beneficial. However, a loose packing is detrimental for charge transport, resulting in very low carrier mobilities. For PLEDs, printability is only of the factors to for commercialization, next to efficiency and stability. In this work, only the first is addressed. Regarding the last two, the performance of the PLEDs is very poor, with EQEs below 1%. Also the PLQY of the polymers presented here is low (35-40%)

To get a better judgement of the achievements, it would be good to make a comparison with a standard polymer as SY-PPV. Like many other emitting polymers it is also asymmetrically substituted and does not crystallize. It has a PLQY of 65% and PLEDs have an EQE of ~4%, which is ten times better than PFPO reported here. Of course, one can argue that it is not fair to compare the performance of a PLED of a yellow emitter with a blue-emitting material, but a comparison of their printability and stretchability could be made, to demonstrate whether the properties realized here are really better.

On a side note, the low PLED performance is, next to the low PLQY also, also the result of a bad hole injection contact. The work function of PEDOT:PSS of ~5.1 eV is far off from the HOMO of the materials of ~6 eV.

Given the very modest optical and electrical properties of the polymers presented here the paper does not meet the high standards of Nat. Comm. with regard to the printability and stretchability a comparison with other asymmetrically substituted polymers should be given.

Answer: Thank you for your comments.

We are also sorry to take your so many troubles for our mistakes. According to your comments, we have corrected our mistakes and polished the whole manuscript. At the same time, we have optimized the device preparation process and improved the device efficiency. Besides, we aldo added many experiment to support our assumption. Therefore, we sincerely hope that our can re-consider our revised manuscript and agree to accept and published our work in *Nature Communications* journal.

We are agreed with you that many high standard works about the flexible OLED are reported and published in the last several years, such as *Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504. However, all these works above are focused on exploring the yellow flexible OLEDs based on the stretchable films. Performance of deep-blue (CIEy< 0.05) emissive stretchable film for the flexible OLEDs needs to further explore. For example, the EQE of blue OLED based on the stretchable films (current efficiency (C. E.) < ~1.6 cd/A and **CIE (0.23, 0.32)** in *Nature* **2022**, 624, 603; TADF materials, EQE< ~**0.8%, in** *Nat. Mater.*, **2023**, *22*, 737-745; C. E. < ~**0.3 cd/A** in *Sci. Adv.*, **2023**, 9, eadh1504.), as displayed in Figure 1. Performance of blue OLED based on stretchable film reported in these high standard journals are very low, much lower than the EQE of our device. We also reasonably believed that the performance of flexible printed OLED based on these stretchable film may further decrease, due to their complicated interchain aggregation and phase separation. In this regard, the EQE of our flexible inkjet-printed deep-blue OLEDs based on our **stretchable film is calculated about 1.39%, much higher than those in these reported works. More important, even at stretching to 50% strain, the PLED based on the elastic FπCPs films also displayed a stable electroluminescence property and comparable**

efficiency without any attenuation, demonstrating the excellent dynamic strain tolerance of FπCPs stretchable nanolayers. Interchain π-π interactions in polymeric materials have a fundamental role in a variety of processes spanning from charge transport to the photophysics of neutral excited species. For example, π-π stacking is essential for high mobility in polymer-based field-effect transistors, whereas formation of intermolecular excited states in luminescent materials may lead to both reduced photoluminescence efficiency, and a reduced energy gap. These are undesirable, especially for fabrication of blue-emitting devices (and therefore for full-colour displays). Even where interactions are not strong enough to lead to formation of aggregates, close-packing of the chromophores may significantly influence the molecular geometry, altering the colour purity and the emission efficiency. Therefore, **it is reasonably concluded that the complicated interchain aggregation can observe in the rod-coil copolymers and blending materials systems, which may cause the formation of multi-chain excited states and non-uniform morphology. These can further result into low device efficiency, instable EL spectra, poor color purity and non-uniform emission. In a word, to obtain intrinsically stretchable homo-polymer is an effective and convenient strategy to achieve a high performance flexible deep-blue OLEDs.**

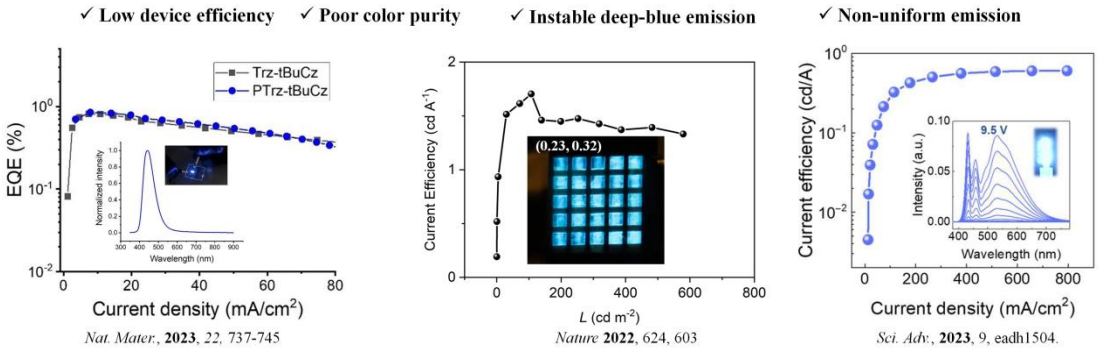


Figure 1. Efficiency of flexible blue OLED based on stretchable film.

It is a fact that our PLEDs present a low efficiency, although it had an excellent stable deep-blue emission. In our opinion, there are some key factors need to consider as follow. We also provided some explanation in this revised manuscript.

1. Unmatched energy level. In general, these flexible conjugated structure result a large bandgap and low HOMO enery level. Low HOMO level caused the unbalance electron and hole injection and result into low charge reconbination.

2. The cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport. In fact, the cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport, which is negative effect on the radiative recombination of holes and electrons in the emissive layer, thus shows a lower EQE. Then, it is ergent to obtain intrinsically stretchable and stable deep-blue conjugated polymer in futures.

3. Configuration structure of our PLEDs need to optimize and further enhance the recombination of electron and hole injection and transport. We need to admit that poor device fabrication capacity is an obstacle to obtain high performance and stable PLEDs. The delicate balance of electrons and holes in the emissive layer for radiative recombination is the key to device efficiency and stability.

Therefore, we are very appreciated your comment about our work. In order to obtain high performance PLEDs, we need to resolve the key problem above. Meanwhile, in our lab, recently, we also obtained a series of high performance and stable deep-blue PLEDs based on the intrinsically stretchable and stable polyfluorene materials. We hope that we can publish some high quality works in these areas in future.

1. The performance of the PLEDs is very poor, with EQEs below 1%.

Answer: Thank you for your comments.

We are agreed with your comment about the device performance, which need to further improve. We optimized the device fabrication process and updated the device data (Figures 4b-4c in the manuscript) in the revised manuscript as follows. The turn-on voltages (V_{on}) of PFPO and POPOF-based flexible inkjet-printed PLEDs were 4.4 V and 4.0 V. The maximum brightness is about 1500 cd/m^2 at 6.8 V and 3100 cd/m^2 at 6.8 V for printed PLEDs of PFPO and POPOF. The maximum external quantum efficiencies of PFPO and POPOF are 1.26% and 1.28%, respectively. This is at the forefront in terms of intrinsic stretchable blue fluorescent emitters.

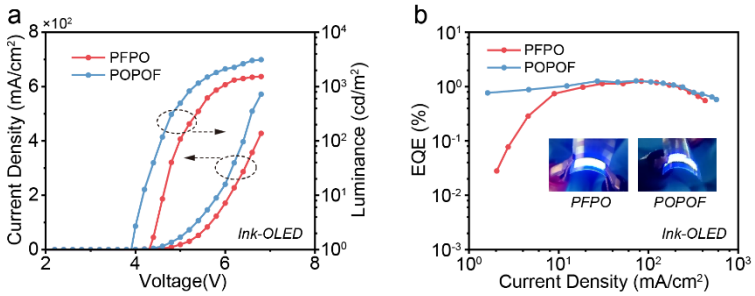


Figure 2. Device performance of flexible OLED on our novel polymers.

In a word, we are agreed your comment about the property and performance of the polymers presented in our work. And as we discussion above, our device performance are comparable to those of flexible OLED based on stretchable films (*Nature* **2022**, 624, 603. *Nat. Mater.*, **2023**, 22, 737-745. *Sci. Adv.*, **2023**, 9, eadh1504.). However, we need to explain again about our novelty and topic of our work again here. In this work, we propose **an asymmetric substitution strategy to improve the printable and stretchable property of deep-blue light-emitting FπCPs with a strong inter-aggregate capillary interaction for flexible inkjet-printed deep-blue polymer light-emitting diodes (PLEDs).** In general, emerging printed and flexible electronic devices provided an infinite possibility of artificial optoelectronic components in application span from soft artificial intelligence and implantable bioelectronics to high quality flexible and stretchable displays and smart wearable. On the one hand, during the large-area inkjet-printed process, the generation of coffee rings can lead to **uneven deposition of the active film, thereby affecting the pattern resolution, device performance and stability.** On the other hand, since flexible optoelectronic devices are highly deformable, active nano-layers in these devices experienced different degrees of tensile and compress strains during the bending, stretching, compressing, and twisting of the devices, which may cause the obvious **intralayer fracture and interlayer separation to further result into device failure**. These two serious problems are the major obstacles to manufacture the printed and flexible device for the practical electronic equipment. Therefore, **obtaining the intrinsically viscoelastic optoelectronic materials with a strong**

capillary interaction is an effective strategy to obtain **a capacity of the suppression of coffee-ring effect and excellent stress tolerance ability** for printed and flexible optoelectronics applications. As promising and potential candidates for flexible printed electronics, however, traditional fully π-conjugated polymers (FπCPs) always present a serious coffee-ring effect and mechanical brittle behavior, due to their rigid and plane backbone structures, which are the most obstacles to limit their application in these areas. Based on these challenge and problem in flexible and printed electronics, we demonstrated an **asymmetric substitution strategy** to obtain robust deep-blue light-emitting FπCPs with a strong **inter-aggregate capillary interaction** for flexible printed polymer light-emitting diodes (PLEDs). This asymmetric substitution strategy can disrupt chain regularity and ordered crystallization capacity of FπCPs, and is favorable for **obtaining excellent stretchable properties to enhance the device deformation stability and stress tolerance ability**. As we expected, the elongation at break of **freestanding asymmetric FπCPs films** (smaller l_{ps}: 6 nm) reached 53% and 72%, much higher than traditional ones (11%), indicating their **outstanding intrinsic stretchability and excellent capacity of stress tolerance**. This is the key factor to ensure the long-time deformation operation stability of flexible electronic devices. More interestingly, compared to the dense isotropic Gaussian chain stacking of traditional FπCPs, **loose anisotropic rod-shaped aggregate** of asymmetric FπCPs with a **strong capillary interaction** is more likely to present a uniform printed deposition, which is beneficial for the formation of **high-quality large-area inkjet-printed films**. Therefore, **flexible and inkjet-printed deep-blue PLEDs** were also manufactured with a stable and efficient deep-blue emission (CIE of (0.17, 0.10)) and maximum brightness of 3100 cd/m^2, with comparable performance to that of spin-coated devices. Considering the widespread potential applications of deep blue flexible and inkjet-printed PLEDs, it is believed that this work will promote the application of PLED in flexible full color displays and wearable devices. In a word, we believe that this is the **first work to simultaneously obtain excellent intrinsically inkjet-printable and stretchable capacity of FπCPs for the flexible and printed electronics**. Meanwhile, this work also is the **first study to systematically and intuitively tune the interchain aggregation of FπCPs in inkjet-printed ink to suppress coffee rings, induce uniform deposition of thin films and enhance viscoelastic properties**, and make our research a milestone in the fabrication of flexible and printed LEDs. **Our work will attract widely and broad attentions in printed electronics, flexible electronics, organic photonics, polymer science**. The topic and unique conclusion of this work match the research area of "***Nature Communications***". Thank you very much for your re-consideration for publication in ***Nature Communications*****.**

Thank you for your comments again.

2.The PLQY of the polymers presented here is low (35-40%)

Answer: Thank you for your comments.

Compared with small molecules, the PLQY of luminescent polymers is relatively low, which is determined by the properties of the polymer itself. For small molecules, it is easier to crystallize during the film preparation process and it is more difficult to form defects. On the contrary, defects in polymers are easier to form, which is detrimental to luminous efficiency, so the PLQY of polymers is generally low.[1, 2] The PLQY of our polymers is comparable to previous work LCPs in solid states.

Thank you for your comments again.

References

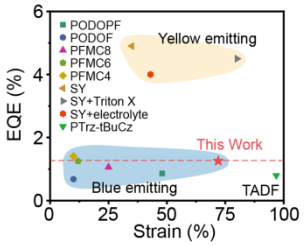
1. St-Onge V, Cui M, Rochon S, Daigle J-C, Claverie JP. Reducing crystallinity in solid polymer electrolytes for lithium-metal batteries via statistical copolymerization. *Communications Materials* 2021, 2(1).

2. Ariu M, Lidzey DG, Sims M, Cadby AJ, Lane PA, Bradley DDC. The effect of morphology on the temperature-dependent photoluminescence quantum efficiency of the conjugated polymer poly(9, 9-dioctylfluorene). *Journal of Physics: Condensed Matter* 2002, 14(42): 9975-9986.

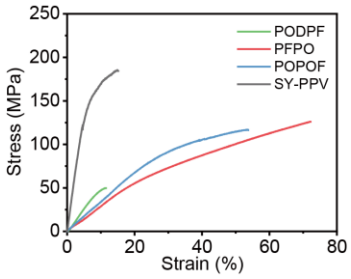
3. To get a better judgement of the achievements, it would be good to make a comparison with a standard polymer as SY-PPV. Like many other emitting polymers it is also asymmetrically substituted and does not crystallize. It has a PLQY of 65% and PLEDs have an EQE of ~4%, which is ten times better than PFPO reported here. Of course, one can argue that it is not fair to compare the performance of a PLED of a yellow emitter with a blue-emitting material, but a comparison of their printability and stretchability could be made, to demonstrate whether the properties realized here are really better.

Answer: Thank you for your comments and suggestions.

First, we admit that the performance of our device is not outstanding. Due to limited laboratory conditions, the preparation of the emitting layer is completed in the air, which is an objective reason for poor device performance. However, in the revised manuscript, we have provided a comparison in the scheme 1f with other reported intrinsically stretchable blue light polymer materials, as well as with super yellow.[1, 2, 3, 4] The materials reported in this work are at a high level in terms of tensile properties. For EQE, it is at the forefront of intrinsically stretchable blue-emitting polymers.

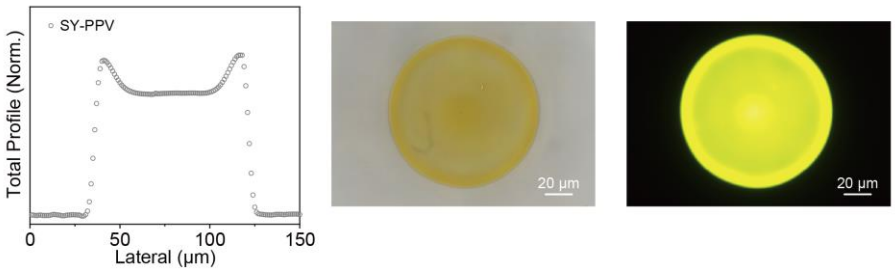


Building upon this, in order to rigorously compare the tensile strength of SY-PPV, we conducted tensile tests on SY-PPV under the same conditions. The experimental results indicate that the fracture elongation of SY-PPV is similar to previous literature reports and is only close to PODPF, significantly lower than PFPO and POPOF.



Finally, we conducted inkjet printing tests using SY-PPV. Compared to PODPF, individual ink droplets exhibited uniform morphology and demonstrated good capability in suppressing coffee ring formation, possibly attributed to the double-sided asymmetric substitution structure of

SY-PPV. Meanwhile, it is also a common sense that the SY-PPV also presents a gelation processing in the toluene solution, which may also result into a proper ink viscosity to obtain slight printable property (Ulrich S.Schubert et al *Adv. Funct. Mater.*, 2007, *17*, 277. Show-An Chen et al *Macromolecules* 2008, *41*, 6500-6504).



Thank you for your comments and suggestions again.

References

1. Liang JJ, Li L, Niu XF, Yu ZB, Pei QB. Elastomeric polymer light-emitting devices and displays. *Nat Photonics* 2013, 7(10): 817-824.

2. Kim JH, Park JW. Intrinsically stretchable organic light-emitting diodes. *Sci Adv* 2021, 7(9): 9715.

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4.On a side note, the low PLED performance is, next to the low PLQY also, also the result of a bad hole injection contact. The work function of PEDOT:PSS of ~5.1 eV is far off from the HOMO of the materials of ~6 eV.

Answer: Thanks for your comments and suggestions. We cannot agree more with you. It is a fact that our PLEDs present a low efficiency, although it had an excellent stable deep-blue emission. In our opinion, there are some key factors need to consider as follow. We also provided some explanation in this revised manuscript.

1.Unmatched energy level. In general, these flexible conjugated structure result a large bandgap and low HOMO enery level. Low HOMO level caused the unbalance electron and hole injection and result into low charge reconbination.

2. The cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport. In fact, the cyclic main chain of PFPO and the loose chain stacking of POPOF are not conducive to charge transport, which is negative effect on the radiative recombination of holes and electrons in the emissive layer, thus shows a lower EQE. Then, it is ergent to obtain intrinsically stretchable and stable deep-blue conjugated polymer in futures.

3.Configuration structure of our PLEDs need to optimize and further enhance the recombination of electron and hole injection and transport. We need to admit that poor device fabrication capacity is an obstacle to obtain high performance and stable PLEDs. The delicate balance of electrons and holes in the emissive layer for radiative recombination is the key to device efficiency and stability.

Therefore, we are very appreciated your comment about our work. In order to obtain high performance PLEDs, we need to resolve the key problem above. Meanwhile, in our lab, recently, we also obtained a series of high performance and stable deep-blue PLEDs based on the intrinsically stretchable and stable polyfluorene materials. We hope that we can publish some high quality works in these area in future.
Thank you for your kind comment.

Finally, thank you for your all kind comments and suggestions. We have modified all incorrect sentences in this revised manuscript. We sincerely hope that our paper can be accepted and published in *Nature Communications*.

Finally, we are sincerely thanks three reviewers. We have checked through carefully and corrected the errors in this revised manuscript. We sincerely hoped that our work can be accepted and published in *Nature Communications* journal.

Thank you for your kind helps again.

All changes to the manuscript have been highlighted. The revised manuscript, Supporting Information, and a list of changes & our responses to the referees' comments have been submitted through my Author Centre.

Thank you very much.

With best wishes.

Sincerely yours,

Professor Wei HUANG

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have addressed all my concerns. They have made attempts to improved the quality of the manuscript.

Reviewer #2 (Remarks to the Author):

The responses from the authors addressed my concerns, and the quality of revision is substantially improved. I think this paper could be accepted.

Reviewer #3 only provided confidential remarks to editors.