

A New Polystyrene–Poly(vinylpyridinium) Ionic Copolymer Dopant for n-Type All-Polymer Thermoelectrics with High and Stable Conductivity Relative to the Seebeck Coefficient giving High Power Factor

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A novel n-type copolymer dopant polystyrene–poly(4-vinyl-N-hexylpyridinium fluoride) (PSPF) with fluoride anions is designed and synthesized by reversible addition–fragmentation chain transfer (RAFT) polymerization. This is thought to be the first polymeric fluoride dopant. Electrical conductivity of 4.2 S cm^{-1} and high power factor of $67 \mu\text{W m}^{-1} \text{ K}^{-2}$ are achieved for PSPF-doped polymer films, with a corresponding decrease in thermal conductivity as the PSPF concentration is increased, giving the highest ZT of 0.1. An especially high electrical conductivity of 58 S cm^{-1} at 88°C and outstanding thermal stability are recorded. Further, organic transistors of PSPF-doped thin films exhibit high electron mobility and Hall mobility of 0.86 and $1.70 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. The results suggest that polystyrene–poly(vinylpyridinium) salt copolymers with fluoride anions are promising for high-performance n-type all-polymer thermoelectrics. This work provides a new way to realize organic thermoelectrics with high conductivity relative to the Seebeck coefficient, high power factor, thermal stability, and broad processing window.

1. Introduction

N-doping has been employed as a crucial process for organic transistors,^[1] solar cells,^[2] organic light-emitting diodes,^[3] and photocatalysts.^[4] Recently, n-doping for use in organic thermoelectrics (OTEs) was studied extensively to control carrier density and electrical conductivity.^[5] OTEs can enable emergent applications in large area and flexible/wearable green energy-harvesting devices, which can convert the heat from the human body into electricity.^[6] Power factor (PF, see below) is commonly used for evaluating the performance of organic thermoelectrics. For example dilute sulfuric acid treated poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS) exhibits high electrical conductivity $\geq 3000 \text{ S cm}^{-1}$, that can equal to or exceed that of indium

tin oxide (ITO) or metal electrodes.^[7] Benefitting from high electrical conductivity, PEDOT:PSS has also been used as a hole-transporting interface material and as electrodes for organic solar cells.^[8] n-type doping results in much lower σ than p-doping with most $\sigma < 1 \text{ S cm}^{-1}$,^[9] and usually uses small molecule n-dopants, such as 4-(1,3-dimethyl-2,3-dihydro-1H-benzimidazol-2-yl)phenyl)dimethylamine (N-DMBI), tetrakis(dimethylamino)ethylene, tetra-*n*-butylammonium fluoride (TBAF), and a polycyclic triaminomethane donor.^[5b,10] To improve the doping efficiency and electrical conductivity, Han Guo et al. reported the air-stable precursor-type molecular dopants for high doping efficiency with a very short doping time of 10 s.^[11]

Recently, most attention was focused on the design and synthesis of novel n-type conjugated polymers. The Lei group reported a new polymer P(PzDPP-2FT) with a zigzag backbone doped with CoCp_2 showing a high electrical conductivity over 120 S cm^{-1} .^[12] The acceptor–acceptor polymer with electron-deficient double $\text{B} \leftarrow \text{N}$ bridged bipyridine unit was proved to be an excellent organic thermoelectric material.^[13] In addition, high electrical conductivity of organic thermoelectrics based on N-DMBI and similar dopants can only be achieved from narrow and limited dopant concentrations.^[14] For example, fluorinated

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The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/adma.202201062>.

DOI: 10.1002/adma.202201062

benzodifurandione-based poly(p-phenylene vinylene) (FBDPPV) doped by N-DMBI exhibits a high electrical conductivity of 12 S cm^{-1} .^[15] Recently, many new benzodifurandione-based oligo(p-phenylene vinylene) (BDOPV)-based polymers were reported for n-type thermoelectrics with conductivity over 10 S cm^{-1} . However, conductivity over 1 S cm^{-1} was only achieved between N-DMBI concentration of 3 and 15 wt%. The thermoelectric performance of polymers is usually evaluated by ZT and power factor (PF)

$$ZT = \frac{S^2 \sigma T}{\kappa} \quad (1)$$

$$PF = S^2 \sigma \quad (2)$$

where S is the Seebeck coefficient, σ is electrical conductivity, T is absolute temperature, and κ is thermal conductivity.^[16] Currently, the common way to enhance the thermoelectric efficiency of polymers is increasing the S and σ ,^[17] because conjugated polymers usually show similar κ .^[18] Though the κ of conjugated polymers is much lower than those of electrically conductive inorganic materials,^[18a] it still can be decreased to enhance the ZT. Polystyrene (PS) usually presents much lower thermal conductivity ($0.03\text{--}0.18 \text{ W m}^{-1} \text{ K}^{-1}$)^[19] than the conjugated polymers ($0.3\text{--}0.5 \text{ W m}^{-1} \text{ K}^{-1}$),^[20] so it can be useful to decrease thermal conductivity while increasing electrical conductivity by introducing PS into dopants.

Compared with small-molecule dopants, polymer-dopant-doped films can achieve higher stability and considerable

electrical conductivity. Seidel et al.^[21] and Yang et al.^[22] reported the polymer dopant poly(ethyleneimine) (PEI)-doped P(NDI2OD-T2) and poly(benzimidazobenzophenanthroline) (BBL) with σ of 0.002 and 8 S cm^{-1} , respectively. Xu et al. reported the conjugated polymer dopants (P(g42T-T)) and (P(g42T-TT)), which doped the ladder polymer BBL at a heterojunction with excellent thermal stability.^[23] To our knowledge, PS-based polymeric fluoride (or other anionic salt) n-type dopants for n-type conjugated polymers, structurally analogous to PSS for PEDOT, have not yet been reported. PSS is a PS derivative with a sulfonic acid group, which makes it an ionic polymer.^[24] Previously, TBAF^[1,25] and the Meisenheimer complexes NDI-TBAF^[26] containing ammonium cation (N^+) and F anion (F^-) were proved to be effective n-type dopants for conjugated polymers. The chemical structure of PSS inspired us to combine PS and the ions of N^+ and F^- for design and synthesis of a polymeric n-type dopant. Pyridine has a similar chemical structure to benzene, and can react with halohydrocarbon to achieve N^+ .^[27] The copolymer dopant PSpF can enhance n-doping ability and maintain the ambient stability of PS. The n-type conjugated polymer PFCITVT (Figure 1) presents excellent n-doping performance with N-DMBI that is similar to that of other BDOPV-based n-type polymers;^[28] here we use it to dope with PSpF for n-type organic thermoelectrics. The highest σ of 4.2 S cm^{-1} and PF of $60 \mu\text{W m}^{-1} \text{ K}^{-2}$ were achieved at room temperature, and high σ of 58 S cm^{-1} was detected at 88°C .

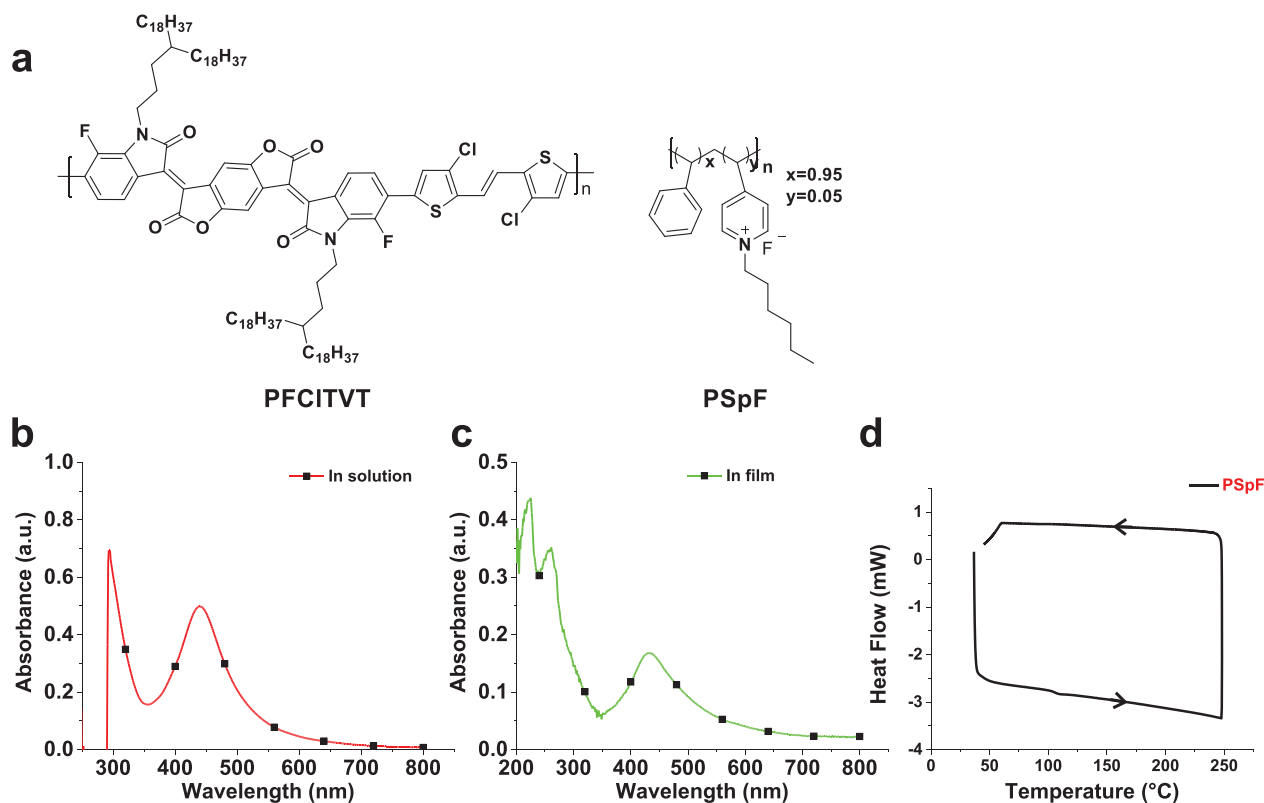


Figure 1. a) Chemical structures of n-type conjugated polymer PFCITVT and dopant polymer PSpF. b,c) UV-vis-NIR absorption spectra of polymer dopant PSpF in solution (b) and in film (c). d) Differential scanning calorimeter (DSC) traces of PSpF measured under N_2 .

2. Results and Discussion

The polystyrene-poly(vinylpyridine) (PS-P) copolymer with 5 mol% pyridine rings was synthesized by RAFT living radical polymerization with molecular weight of 334 kDa.^[29] The copolymer PSpBr containing Br[−] was achieved by nucleophilic substitution with bromohexane (Supporting Information). A PS-P-based polymer dopant PSpF was obtained from PSpBr by ion exchange reaction (Figure 1a). The absorption spectra of PSpF in solution are shown in Figure 1b; two absorption peaks were detected at 294 and 440 nm, respectively. They can be attributed to the absorption of PS^[30] and fluoride polypyridine salt, respectively.^[31] The PSpF film had three absorption peaks at 224, 260, and 431 nm, respectively; the blueshifts of 34 and 9 nm were observed in the absorption of PS and polypyridine salt, respectively. The differential scanning calorimeter (DSC) traces of polymer PS-P and PSpF were measured under N₂ between 40 and 250 °C. The glass transition temperatures (*T*_g) of PS-P and PSpF are 114 and 109 °C, respectively. The relatively lower *T*_g of PSpF is related to the hexyl sidechains on pyridine. The unit “wt%” in this paper means weight ratio of PSpF compared to the conjugated polymer PFCITVT, for example, 100 wt% means equal weights of conjugated polymer and PSpF.

The UV-vis-NIR absorption spectra of pristine and doped PFCITVT films are shown in Figure 2a and Figure S5b, Supporting Information. The pristine film displays two absorption peaks at 465 and 777 nm, which can be attributed to π - π^* transition and intramolecular charge transfer.^[32] With 5 wt% PSpF

doping, stronger absorption was detected in the low energy region of 1000–1800 nm (Figure 2b), contributed by polaron/bipolaron transitions^[33] and similar to N-DMBI-doped films.^[34] However, the absorption of neutral N-DMBI-doped films is usually bleached,^[28a] here the absorption intensity increases with PSpF doping, different from N-DMBI-doped films. When the weight fraction of PSpF increases to 30 wt%, absorption in the low energy region is much stronger and two new weak absorption peaks at 1350 and 1596 nm appear (Figure 2b). With the weight fraction of PSpF increasing from 30 to 75 wt%, the two peaks become stronger and the neutral absorption in the high energy region becomes weaker but is still stronger than for pristine PFCITVT. The absorption result demonstrates that effective doping occurs in films of PFCITVT: PSpF. The electron paramagnetic resonance (EPR) spectra of pristine and doped PFCITVT solution are shown in Figure 2c. There is no radical peak for pristine PFCITVT solution, while an obvious radical peak was detected in 5 wt% PSpF doped solution that is at the similar magnetic field with N-DMBI-doped polymers.^[28a,35] When PSpF fraction increases to 50 and 100 wt%, the EPR intensity is much stronger than 5 wt% PSpF doped solution, and further proves the effective doping by PSpF. The absorption in the region of 1300–1800 nm (near IR, referenced to absorbance at 1200 nm) increases when the doping ratio increases from 5 to 75 wt%, fully as expected, and then decreases when the dopant/polymer ratio is 100 wt%. The EPR spectra are consistent with near IR absorption spectra results. The EPR intensity increases when the dopant ratio increases from 5 to 75 wt%,

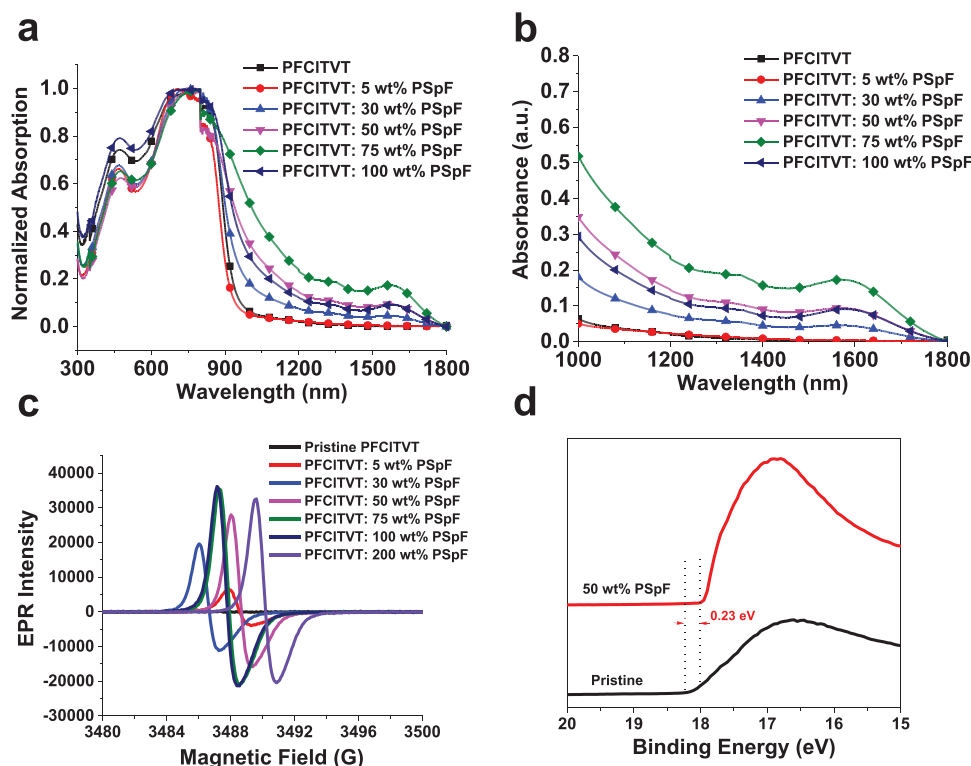


Figure 2. Normalized UV-vis-NIR absorption spectra of pristine PFCITVT and polymer films doped with different weight fractions of dopant in the UV-vis-NIR region (a) and in the NIR region (b). c) EPR spectra of pristine and doped polymer in solution. d) UPS binding energy of the pristine and doped polymer films measured under -5 eV. Herein, “wt%” means weight ratio of PSpF compared to the conjugated polymer PFCITVT, for example, 100 wt% means equal weights of conjugated polymer and PSpF.

then slightly decreased at 100 wt% and decreased more at 200 wt%. The highest spin density was calculated to be $1.35 \times 10^{20} \text{ cm}^{-3}$ based on a Bruker calibration sample. This is of the same order of magnitude as the repeat unit number density. The ultraviolet photoelectron spectroscopy (UPS) spectra are shown in Figure 2d and Figure S5a, Supporting Information. The secondary electron cutoff of PFCITVT doped by 50 wt% PSpF shifts by -0.23 eV , suggesting a downward movement of its Fermi level by 0.23 eV ^[34] which is similar to the TBAF-doped polymer films.^[25a] and could be from associations of the doped polymer with multiple cations of the dopant or a surface voltage induced by the dopant.

The electrical conductivity of doped polymer films was examined by a four-probe method and the Seebeck coefficients were determined by detecting the thermoelectric voltages under different temperature gradients ΔT . All the measurements were performed in the open air. All the doped films exhibit reasonably high σ over 1 S cm^{-1} except polymer films doped by 1 wt% PSpF, indicating PSpF-doped polymer films can give effective electron transport over a broad range of dopant concentration (The F-/PFCITVT ratio is between 4.7 and 188 mol%, Figure S5d, Supporting Information), which is very different from N-DMBI-doped films,^[15,28a,36] suggesting a broad process window for polymer dopant PSpF; polymers with 100 wt% PSpF doping show the highest σ of 4.2 S cm^{-1} (Figure 3a). The Seebeck coefficients for 1, 5, 30, 50, 75, 100, and 200 wt% are

649 ± 75 , 476 ± 7 , -455 ± 10 , -432 ± 31 , -354 ± 28 , -316 ± 11 , and $550 \pm 100 \mu\text{V K}^{-1}$, respectively (Figure 3b); the S are relatively consistent in the PSpF fraction range between 1 and 200 wt% compared to N-DMBI-based devices,^[28a] suggesting high concentration-tolerance of PSpF doping. The highest power factor of $75 (67 \pm 8) \mu\text{W m}^{-1} \text{ K}^{-2}$ was achieved for 200 wt% PSpF doped films with the contribution of relatively high σ relative to S (Figure 3c). PFCITVT doped by 200 wt% PSpF exhibits relatively high electrical conductivity and power factor of 2 S cm^{-1} and $67 \mu\text{W m}^{-1} \text{ K}^{-2}$, respectively. The lowest PF is $28 \mu\text{W m}^{-1} \text{ K}^{-2}$ with 30 wt% PSpF doping; even that PF is still much higher than for most n-type organic thermoelectrics.^[9]

Thermal conductivity measurements on the thin film samples were performed via time-domain thermoreflectance (TDTR) to study the effect of polystyrene-based dopant PSpF on that property.^[37] The thermal conductivities of pristine PFCITVT and PSpF are about 0.25 ± 0.07 and $0.11 \pm 0.04 \text{ W m}^{-1} \text{ K}^{-1}$ (Figure 3d), respectively. The thermal conductivity of PSpF-doped PFCITVT films decreased from 0.22 ± 0.07 to $0.16 \pm 0.04 \text{ W m}^{-1} \text{ K}^{-1}$ when the dopant concentration increased from 5 to 100 wt%, suggesting PSpF can decrease the thermal conductivity of doped polymer films in proportion to its compositional fraction. The sources of uncertainty in our reported values for thermal conductivity measurements on these thin film polymer samples are reported in our prior works.^[20,25a] The highest ZT, assuming isotropic orientation of drop-cast films, is calculated to be about 0.1.

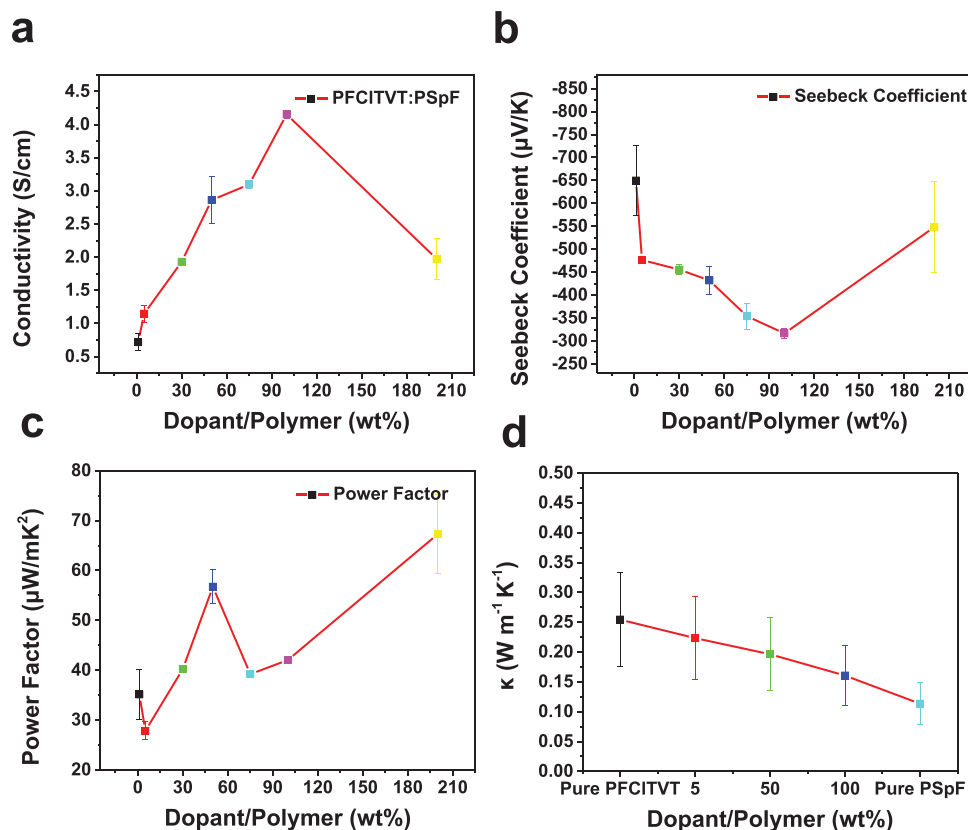


Figure 3. a) Electrical conductivity, b) Seebeck coefficient, c) power factor, and d) thermal conductivity of PFCITVT films doped by various weight fractions of PSpF. The black spot in (a) is 1 wt% PSpF doped, not undoped. Resistance was measured by using a four-probe method with a channel length of 1000 μm and a channel width of 140 μm . Seebeck coefficients were measured with a channel length of 2000 μm and a channel width of 8000 μm .

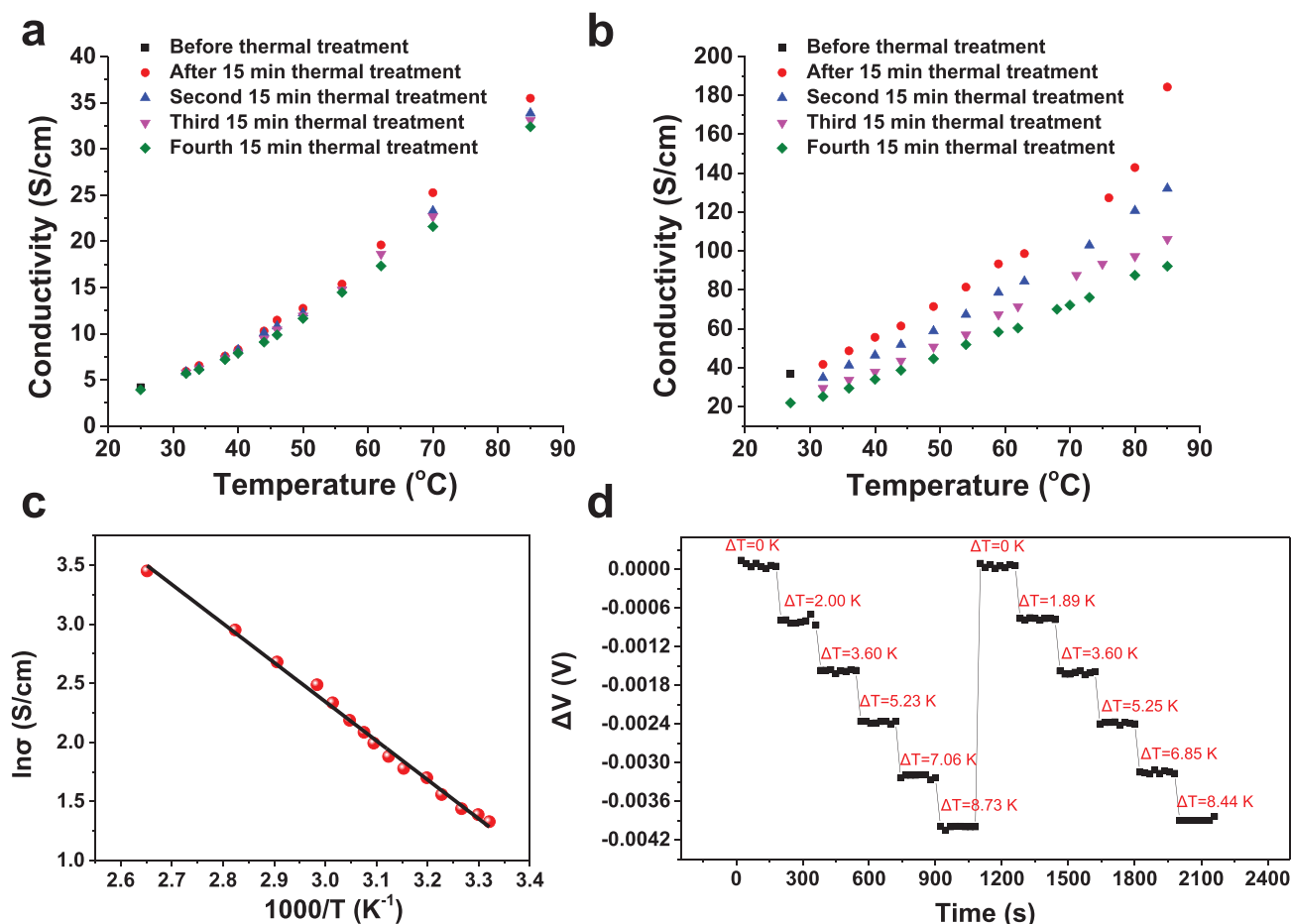


Figure 4. a,b) Thermal air stability of electrical conductivity of PSpF-doped PFCITVT (a) and N-DMBI-doped PFCITVT (b) films after thermal treatment at 120 °C for four-cycles of 15 min in the open air. c) Temperature-dependent electrical conductivity values of PFCITVT film doped with 30 wt% PSpF. d) Time-dependent thermoelectric voltage response under different temperature gradients ΔT .

To explore the relationship of S , PF , and σ , the Seebeck coefficient and power factor as functions of electrical conductivity in this work were compared with reported works which have been summarized by Russ et al. (Figure S6, Supporting Information).^[17] Though the S and PF (Figure S6, Supporting Information) in this work are relatively high, they are still reasonable and very similar to the trend of p-type thermoelectrics based on PEDOT:PSS.^[38]

The thermal stability in the ambient atmosphere is very important for thermoelectric devices. It was explored by recording the electrical conductivity of films with 75 wt% PSpF doping before and after thermal treatment at 120 °C for two cycles of 15 min in the open air. The σ at room temperature was 3.45 S cm⁻¹ before thermal treatment; after 2 cycles of 15 min thermal treatment, the value of 3.39 S cm⁻¹ was achieved, an insignificant 2% decrease (Figure S5c, Supporting Information). The σ values decreased about 1–10% at 28–57 °C, exhibiting excellent thermal stability in the open air. Moreover, the apparent E_a hardly changed in the process. The doped film also shows good ambient stability; the σ was 2.35 ± 0.27 S cm⁻¹ upon 9 days exposure to air, only a 24–40% decrease. Considering that the thickness of the films was only 100–300 nm,

the ambient stability is outstanding. To compare the thermal stability with that from a conventional dopant, 100 wt% PSpF and 50 mol% N-DMBI doped PFCITVT films were measured under the same condition; the result is shown in Figure 4a,b. After four cycles of 15 min thermal treatment, the σ values of PSpF-doped PFCITVT decreased about 4–14% at 32–85 °C, the σ values at room temperature decreased from 4.2 to 3.9 S cm⁻¹, a low decrement of 7% was observed. Meanwhile, a more significant 37–50% decrease was observed in N-DMBI-doped PFCITVT films after four cycles of 15 min thermal treatment, which is much higher than PSpF-doped PFCITVT. The σ value of N-DMBI-doped PFCITVT at room temperature was 36.8 S cm⁻¹, and decreased to 21.8 S cm⁻¹ after four cycles of 15 min thermal treatment; a high decrement of 41% was observed. The stability of PSpF-doped PFCITVT is probably promoted by the fragments of PS in PSpF that could block the access of water and/or oxygen to the mobile electrons.

To estimate the activation energy (E_a) of doped polymer films, temperature-dependent electrical conductivity values of PFCITVT with 30 wt% PSpF doping were recorded in Figure 4c. The PSpF-doped film shows increasing σ values over the range of 25–90 °C. The apparent E_a was calculated according to the

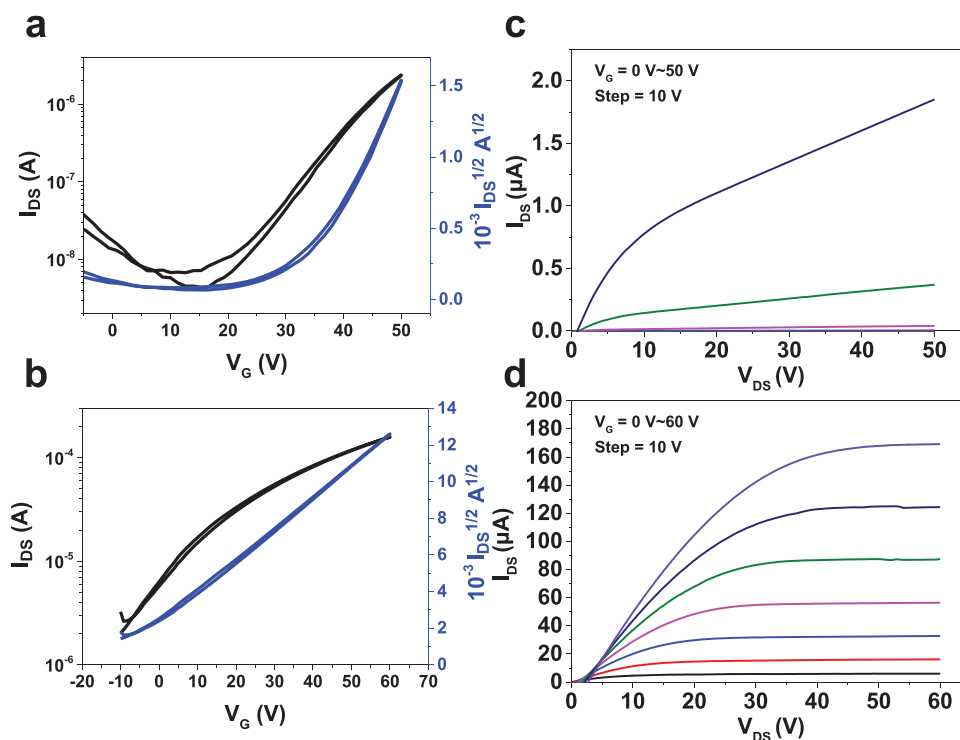


Figure 5. a,b) Transfer curves of pristine OFETs (a) and 1 wt% PSpF doped OFETs (b). c,d) Output curves of pristine OFETs (c) and 1 wt% PSpF doped OFETs (d). The OFETs were prepared with a channel length of 200 μm and a channel width of 8000 μm .

Arrhenius equation, being 282 meV. The value of the activation energy divided by the average temperature of the measurement is $852 \mu\text{V K}^{-1}$.^[39] The value is somewhat higher than the measured Seebeck coefficient due to the barrier to site-to-site hopping, but is of the same order of magnitude as S . The time-dependent thermoelectric voltage responses under different temperature gradients were recorded for 36 min (Figure 4d). These were very stable, suggesting the relatively high Seebeck coefficient can most likely originate from an electron contribution, not from ion contributions.^[40]

Electron mobility plays a key role in electrical conductivity, according to the formula $\sigma = ne\mu$, where n is the carrier density, e is the electron charge, and μ is the corresponding carrier mobility. The σ is positively related to μ and n of polymer films.^[41] To measure the electrical mobility of doped polymer films, organic field-effect transistors (OFETs) with top-gate/bottom-contact configuration were prepared and studied. The dopant PSpF fractions in the OFETs are 1, 2, and 10 wt%. The transfer and output curves are shown in Figure 5, Figures S8 and S9, Supporting Information. The performance of OFETs is summarized in Table S2, Supporting Information. In the transfer curves, PFCITVT with 1 wt% PSpF doping shows much higher I_d than pristine films, while, in the output curves, 1 wt% PSpF doped films show better linear behavior than undoped films in the low V_d region, owing to the reduction of contact resistance.^[25b] PFCITVT with 1 wt% PSpF doping shows a high electron mobility of $0.81 \pm 0.05 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, much higher than the mobility of undoped PFCITVT of $0.24 \pm 0.04 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. This could be from the filling of traps and/or the dopant inducing locally improved order. When the dopant fraction increases to 2 and 10 wt%, the electron mobility

decreases to 0.37 ± 0.01 and $0.13 \pm 0.05 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively, presumably because the unconjugated polymer dopant can disorder the conjugated polymer arrangement. The results are also further supported by a Hall effect measurement. A high electron mobility of $1.70 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ was achieved in 50 wt% PSpF doped PFCITVT films, which is much higher than $0.97 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in pristine PFCITVT films.

Polymer film microstructures were determined by grazing-incidence X-ray scattering (GIXRS). The strong diffraction peaks of (100) and (200) were detected for pristine and 5–75 wt% PSpF doped polymer films, suggesting polymer molecules are in an ordered arrangement when the fraction of PFCITVT is higher than PSpF (Figure S7a, Supporting Information). There is no (010) peak detected in the out-of-plane diffractions, indicating the polymer films have an edge-on orientation packing. With the fractions of PSpF increasing from 0 to 75 wt%, the lamellar d -spacing distance increases from 30.15 to 32.97 \AA (Figure S7b, Supporting Information), possibly indicating some intercalation of polystyrene segments within the non-polar parts of the conjugated polymers. PFCITVT with 100 wt% PSpF doping presents a smaller d -spacing distance (32.17 \AA) than that of 75 wt% PSpF doping and a much weaker (100) peak, suggesting further change and disorder of the polymer arrangements if this d -spacing difference is considered significant. The (200) peak width decreases linearly as the PSpF fraction increases from 0 to 100 wt% (Figure S9, Supporting Information), suggesting PSpF likely can make alkyl side chains more compact.^[42] The surface morphology of polymer films was investigated by atomic force microscopy (AFM). All the films present similar small-size fiber-like aggregates with no preferred direction, suggesting good miscibility of PSpF with

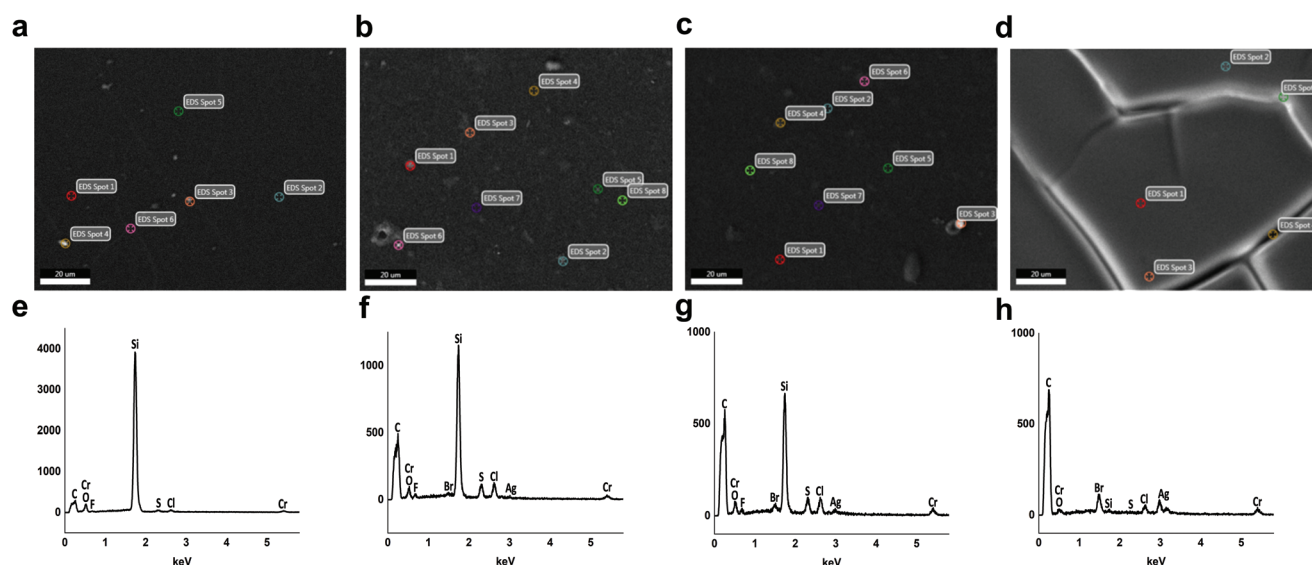


Figure 6. To measure SEM, polymer films were prepared by drop-casting which is the same as for the all-polymer thermoelectrics. a–d) SEM images of: a) pristine PFCITVT, b) 5 wt% PSpF doped PFCITVT, c) 50 wt% PSpF doped PFCITVT, and d) pristine PSpF films. e–h) EDS analysis at the even area of: e) pristine PFCITVT (spot 1), f) 5 wt% PSpF doped PFCITVT (spot 4), g) 50 wt% PSpF doped PFCITVT (spot 5), and h) pristine PSpF films (spot 1). The percentage composition of F in (b) and (c) is higher than (a), suggesting the existence and adduct reaction with PFCITVT of F[−] in PSpF.

conjugated polymers (Figure S11, Supporting Information). The smaller root-mean-square roughness of polymer film with 100 wt% PSpF doping is attributed to the low crystallinity (implying little or no preferred orientation) consistent with the GIXRS result.

To further study the morphology and doping reaction of PFCITVT and PSpF, scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) measurements were done to examine the films prepared by drop-casting on Si/SiO₂ substrates. The micrometer-sized aggregates can be observed in 5 and 50 wt% PSpF doped PFCITVT films (Figure 6), indicating the phase separation between the ionic polymer PSpF and conjugated polymer PFCITVT. There is no F detected in pure PSpF film because F[−] can escape as HF in high vacuum. The content of F atoms increased from 0.14% in pristine PFCITVT to 0.96% in 5 wt% PSpF doped PFCITVT (Tables S3–S5, Supporting Information), due to the reaction of F[−] with BDOPV rings in PFCITVT.^[25a] The results indicate that the F atoms are covalently bonded to the polymer after doping.

We already proposed that the electron-rich F[−] can react with the strong electron-withdrawing unit BDOPV to form a radical anion.^[25a] In the doping process, F[−] is the effective part and PSP⁺ will act as the counterion to compensate for the negative charge on the PFCITVTF[−] polymer chain. In the present case, the electron-rich F[−] can react with the similarly strong electron-withdrawing unit and form anionic Meisenheimer complexes PFCITVTF[−], from which mobile electrons are transferred to other polymer segments, as we have also previously discussed.^[22] The doping reaction scheme is shown in Figure 7. The Meisenheimer complexes are stable because the F is covalently bonded to the polymer PFCITVT, as we confirmed by the EDS measurement. The PSP⁺ cation remains as the counterion for the F-complexes. The PFCITVTF[−] radical anion can be the vehicle for transporting electrons, and PSP cations are relatively stationary and could be local electron traps. The polymer

dopant is non-conducting because of the dominant polystyrene groups, and can enhance the Seebeck coefficients because of the trapping (locally decreased electron energy levels) and possible energy-filtering barriers.

3. Conclusion

The results demonstrate that the copolymer PSpF can be an effective n-dopant for high-performance n-type organic thermoelectrics. A high electrical conductivity of 4.2 S cm^{−1} and power factor of 67 μW m^{−1} K^{−2} were achieved for PSpF-doped polymer films. The OFETs of PSpF-doped thin films exhibit high electron mobility of 0.86 cm² V^{−1} s^{−1}. Moreover, excellent thermal stability and ambient stability were observed for the electrical conductivity of PSpF-doped films. Very stable time-dependent thermoelectric voltage responses under different temperature gradients were recorded. This work opens the way for designing polymer n-type dopants for organic conductors and thermoelectrics with low thermal conductivity, high conductivity relative to the Seebeck coefficient, and high power factor.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work was primarily supported by the National Science Foundation, Division of Chemistry, grant numbers 1708245 and 2107360. The authors thank Baixiang Li and Yuyang Ji for their help with GPC measurement.

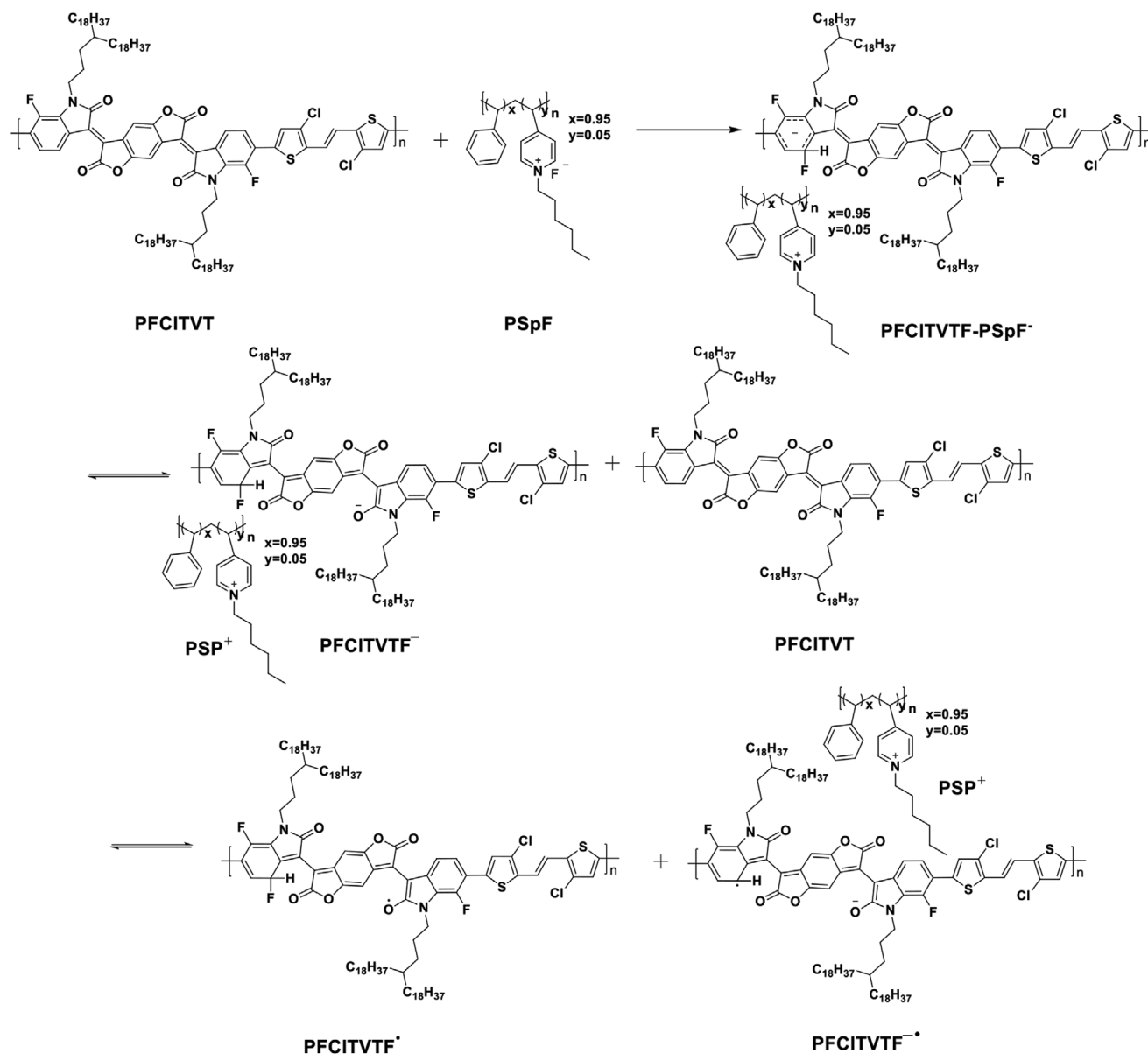


Figure 7. The proposed doping mechanism of polymer dopant PSpF and semiconducting polymer of PFCITVT. Other F⁻ addition sites and radical/anion resonance structures are possible.

The authors acknowledge Lisa Pogue and Tyrel McQueen for assistance and training on the X-ray diffractometer. The authors also appreciate support for thermoelectric measurements from the Office of Naval Research, grant number N00014-20-1-2686.

Keywords

all-polymer thermoelectrics, electrical conductivity, electron mobility, n-type polymer dopants, Seebeck coefficient

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are openly available in the Johns Hopkins University Data Archive.

Received: January 31, 2022

Revised: April 8, 2022

Published online: May 29, 2022

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