Flexible, large-area, and stable perovskite photodetectors have been of interest for their potential applications in various fields, including optics, electronics, and photonic devices. However, due to the inherent instability of perovskite materials, research efforts have been directed towards improving their environmental stability. One of the strategies to enhance stability is to encapsulate the perovskite layers using polymers or metal electrodes, which can better withstand environmental stresses.

In this work, a highly bendable, stable, and large-area (3 cm^2) flexible polystyrene incorporated perovskite photodetector was developed. This composite photodetector exhibited a high responsivity up to 2.73 A/W, a maximum specific detectivity of 6.2 x 10^13 Jones, and a superior switching ratio of 1.0 x 10^4. In addition, the polystyrene-perovskite photodetector yields excellent stability under the combined stresses of moisture, ambient air, and room light, and retains 92% of its original performance for over 30 days. These results demonstrate that this work provides a facile and cost-effective approach to developing high-performance, stable, and highly flexible optoelectronic devices.

**INTRODUCTION**

Large-area, cost-effective, and flexible photodetectors are required in multiple applications such as next-generation wearable optoelectronics, robotics, bio-imaging, illumination monitoring systems, and remote sensing. Organometal halide perovskites, specifically CH_3NH_3PbI_3 (or MAPbI_3) have emerged as outstanding light-harvesting materials in the optoelectronic field due to its long charge carrier diffusion length, low exciton binding energy, broadband absorption, and direct bandgap. The added advantage of their solution based low-temperature fabrication process makes them suitable for application in flexible electronics and photonic devices. Flexibility is primarily defined by the ability to bend these devices to achieve a low form factor or easy integration with a curved surface for mounting on non-traditional spaces. The common strategy in perovskite photodetectors is to reduce the thickness of the film in a single step, which allows for unprecedented mechanical stability, maintaining 85% of its original thickness after 10,000 bending cycles at a bending angle of 120°. Similarly, the solution-processed self-assembled Pt–Au nanochains were developed to provide a simple and fast method of patterning the conductive and flexible electrodes onto the filter substrate. The optimized polystyrene-perovskite photodetector exhibits a high responsivity up to 2.73 A/W, a maximum specific detectivity of 6.2 x 10^13 Jones, and a superior switching ratio of 1.0 x 10^4. In addition, the polystyrene-perovskite photodetector yields excellent stability under the combined stresses of moisture, ambient air, and room light, and retains 92% of its original performance for over 30 days. All these results demonstrate that this work provides a facile and cost-effective approach that paves the way to develop high-performance, stable, and highly flexible optoelectronic devices.
The photocurrent generation from the flexible PS-MAPbI$_3$ photodetector is schematically depicted in Fig. 2a. Our photodetector is based on the photoelectric effect in which the applied voltage results in an electric field that leads to the effective separation of electrons and holes generated on light illumination. Due to the close matching of the work function of Pt–Au (5.2 eV) with the highest occupied molecular orbital (HOMO) of PS–MAPbI$_3$ perovskite (5.4 eV), the collection of holes are favorable by the negative Pt–Au electrode in the symmetric Pt–Au/PS-MAPbI$_3$ (or MAPbI$_3$)/Pt–Au device. Whereas, the electrons are not easily collected (or get trapped) due to the large energy level difference between the Lowest Unoccupied Molecular Orbital (LUMO) of PS–MAPbI$_3$ (3.9 eV) and work function of Pt–Au (5.2 eV). Figure 2b presents the photoresponse from both plain MAPbI$_3$ and PS-MAPbI$_3$ devices with varying concentrations of PS) at 2 V and under the illumination of 1.0 sun. The plain MAPbI$_3$ device showed a photocurrent of 2.8 µA, which enhanced with the incorporation of PS. Specifically, the photocurrent for 0.5 wt % PS-MAPbI$_3$ device is 4.8 µA, and it increased substantially to 11.2 µA for 1 wt % PS-MAPbI$_3$ device. We attribute this to the fewer defects and suppressed charge carrier recombination by the cross-linking of PS within MAPbI$_3$. Further, the addition of PS to 3 and 7 wt % caused a drop in photocurrent to 6.9 and 1.1 µA, respectively. The corresponding light switching on/off ratios is illustrated in Supplementary Fig. 4, and it also follows a similar trend. It is worth noting that the light switching ratio for 1 wt % PS-MAPbI$_3$ device (1.0 × 10$^4$) is 1 order of magnitude higher than that of plain MAPbI$_3$ device (1.0 × 10$^3$). The comparison reveals that among all the above PS concentrations, 1 wt % PS device gives the best performance due to the highest carrier lifetime and mobility, and simultaneously lowest, charge recombination effects, the density of defect traps, and ion migration, and therefore it was characterized in detail. The dark currents and photocurrents were measured to quantitatively compare the performance of photodetectors made with both plain MAPbI$_3$ and 1 wt % PS-MAPbI$_3$. For both the photodetectors, with increasing voltage the dark currents and photocurrents increase (Fig. 2c) due to greater strength of the electric fields, as it leads to more effective charge separation and collection. The 1 wt % PS-MAPbI$_3$ device exhibits a better performance with ~3 times lower dark current and ~4 times higher photocurrent than the plain MAPbI$_3$ devices. This can be attributed to the presence of PS (insulating material), reduced ion-migration effects, and lower carrier recombination in PS-MAPbI$_3$ photodetectors. The photoresponse curves of the optimized 1 wt % PS-MAPbI$_3$ photodetector as a function of different wavelengths (410–710 nm) at a low bias of 2 V are presented in Fig. 2d. The photocurrent of the device was measured to be 26, 22, 28, 40, 45, and 22 nA under 410, 475, 520, 585, 650, and 710 nm light irradiation, respectively. The device shows a consistent and stable response over the cycles under monochromatic light. To quantitatively evaluate the output performance of plain MAPbI$_3$ and 1 wt % PS-MAPbI$_3$ photodetectors, their spectral responsivity (Fig. 2e), detectivity (Fig. 2e), and external quantum efficiency (Supplementary Fig. 5) were measured. The responsivity (R) and detectivity (D$^*$) are defined as

$$R = \frac{I_{ph} - I_d}{P_{in}}$$

(1)

$$D^* = \frac{R}{\sqrt{2qa/A}}$$

(2)

where $I_{ph}$ is the photocurrent, $I_d$ is the dark current, $P_{in}$ is the incident light power, $q$ is the electron charge, and $A$ is the effective area of the photodetector. The responsivity and detectivity of the 1 wt % PS-MAPbI$_3$ photodetector (Fig. 2e) in the spectral range from 400 to 750 nm are 5 and 7.5 times higher than the plain MAPbI$_3$ photodetector, respectively. We observe that the 1 wt % PS-MAPbI$_3$ device attains a maximum responsivity of 0.25 A W$^{-1}$.
and detectivity of $5.7 \times 10^{12}$ Jones under illumination by monochromatic light with a wavelength of 650 nm. The corresponding external quantum efficiency (EQE) of 1 wt% PS-MAPbI$_3$ device is greater than that of the plain MAPbI$_3$ device as shown in Supplementary Fig. 5. The photoresponse of 1 wt% PS-MAPbI$_3$ device irradiated by 650 nm light of different intensities ($0.001$–$10$ mW cm$^{-2}$) is shown in Fig. 2f. The photocurrent increases with the light intensity, which can be ascribed to the increased number of photogenerated charge carriers in response to the higher photon flux$^{26}$. It should be noted that the photocurrent at very low intensity (0.001 mW cm$^{-2}$ or 1 µW cm$^{-2}$) of 650 nm light is 45 nA, which is more than 1 order of magnitude higher than that under dark conditions (1.1 nA). The corresponding responsivity and detectivity are shown in Supplementary Fig. 6. The response time of the 1 wt% PS-MAPbI$_3$ photodetector was also determined and found to be <5 ms, which is the detection limit of our setup (Supplementary Fig. 7). The responsivity and detectivity of plain MAPbI$_3$ and 1 wt% PS-MAPbI$_3$ photodetectors using 2 V at different light intensities by attenuating 1.0 sun illumination (from solar simulator) using neutral density filters is presented in Fig. 2g. The maximum value of $R$ and $D^*$ for 1 wt% PS-MAPbI$_3$ device is 2.73 A W$^{-1}$, and $6.2 \times 10^{13}$ Jones at illumination intensity of 0.001 mW cm$^{-2}$, which is comparable and even higher than the previous reports on the flexible photodetectors (Table 1)$^{3,12,13,20,27–38}$. In contrast, the plain MAPbI$_3$ device achieves only $R$ (0.61 A W$^{-1}$) and $D^*$ (0.86 $\times 10^{13}$ Jones), which implies that the incorporation of 1 wt% PS in MAPbI$_3$ greatly improved the photodetector performance. The higher $R$ and $D^*$ observed under illumination from the solar simulator (a broad-spectrum source) compared to illumination at 650 nm is attributed to the broad absorption spectrum of MAPbI$_3$ which leads to greater absorption of photons of higher energies.

Flexibility and operational stability

As mechanical stability is the major concern for the practical feasibility of flexible photodetectors for wearable electronics, we studied the bending durability of both plain MAPbI$_3$ and 1 wt% PS-MAPbI$_3$ photodetectors. Figure 3a shows the schematic representation of the bending of the flexible PS-MAPbI$_3$ photodetector. The device was bent at varied angles as shown in the inset of Fig. 3b, and the photocurrent was measured during the
bending to ensure the performance reliability of the flexible device. The photocurrent of both flexible devices remains unchanged with the bending angle varying from 0° to 120° (Fig. 3b). Even at the maximum bending angle of 180°, there is a slight decrease in photocurrent which can be due to the reduction of the light-receiving area at the bending state. The photocurrent retained to its original value as the device brings back to zero bending angles. Further, we measured the photocurrent of both devices after bending for 10,000 cycles at a fixed bending angle of 120° and a bending radius of 1.5 mm. For clarity, all the photocurrents were normalized to their respective initial values. As can be seen in Fig. 3c, the photocurrent of the 1 wt % PS-MAPbI3 device maintained 100, 91, and 85% of its initial value after 1000, 5000, and 10000 bending cycles, whereas under the same bending conditions for plain MAPbI3 devices the photocurrent dropped to 90, 68, and 50% of its initial value. The photodetectors are also evaluated for their long-term stability in operation. The plain MAPbI3 and 1 wt % PS-MAPbI3 devices with and without an adhesive layer were tested upon exposure to the ambient environment (in the air at a relative humidity of 35–40% under room light). Figure 3d shows the 1 wt % PS-MAPbI3 devices maintain 92% of its initial photocurrent value after 720 h (30 days). In contrast, the plain MAPbI3 devices completely degrade in just 192 h (8 days) with more than 60% loss within 3 days. Similarly, we also achieved a better stability for the 1 wt % PS-MAPbI3 device without an adhesive layer in the ambient environment as compared to the plain MAPbI3 device (Supplementary Fig. 8). The improved stability of 1 wt % PS-MAPbI3 devices can be attributed to the combined effects of the presence of PS (hydrophobic in nature) which leads to passivation of defects,
and reduced ion-migration effects\textsuperscript{22,39}. Further to ensure the reliability of the photocurrent measurement, the photoresponse of the plain MAPbI\textsubscript{3} and 1 wt % PS-MAPbI\textsubscript{3} devices were recorded as a function of time at 2 V under 650 nm light irradiation (0.1 mW cm\textsuperscript{-2} intensity), as presented in Fig. 3e. Both the devices exhibited very stable performances. Measured for 60 min, the photocurrent of the plain MAPbI\textsubscript{3} device was measured to be at 37 nA, whereas the 1 wt % PS-MAPbI\textsubscript{3} device exhibited a photocurrent of 169 nA. These results demonstrate that our 1 wt % PS-MAPbI\textsubscript{3} device had excellent mechanical durability as well as outstanding environmental stability. The mechanical stability is also attributed to the porous and chain-like structure of the Pt–Au electrode (Supplementary Fig. 2) which can endure repeated bending without failure. It also shows that the Pt–Au electrode remains chemically stable for days and under device operating conditions, without any degradation with the perovskite layer. Ag nanowires in comparison can undergo redox reactions with the perovskite layer with relative ease impacting stability\textsuperscript{30–42}. Ag nanowires also have the challenge of mechanical and thermal stability and junction resistance\textsuperscript{43}. Further the Pt–Au electrodes are made using a room temperature based self-assembly process for synthesis and simple vacuum filtration for control of the deposition process. The conductivity/cost ratio of the Pt–Au electrodes is \textasciitilde63,000 Sm\textsuperscript{-1}/US$, while for Ag nanowire-based electrodes this can range from 8000–123,000 Sm\textsuperscript{-1}/US $, making them comparable in cost. It is also higher than pure Au electrodes (1023 Sm\textsuperscript{-1}/US $)\textsuperscript{31,44}.

In summary, we have demonstrated the feasibility of using PS-incorporated MAPbI\textsubscript{3} networks to develop a porous film on a flexible filter membrane for fabricating large-area ultra-flexible photodetector. Low-cost flexible Pt–Au electrodes were patterned by using the facile solution-processed self-assembled nanochains. The optimum 1 wt % PS-MAPbI\textsubscript{3} device showed extremely high bending stabilities, maintaining a photocurrent of 85% of their original values after 10,000 bending cycles under a large bending angle (\(\theta = 120^\circ\)). The responsivity and detectivity of 1 wt % PS-MAPbI\textsubscript{3} flexible photodetector reached 2.73 A W\textsuperscript{-1} and 6.2 \times 10\textsuperscript{13} Jones, respectively. This is among the best-reported performance with high mechanical durability when compared to other flexible perovskite photodetectors. Moreover, the PS incorporation in MAPbI\textsubscript{3} showed long-term device stability for more than 30 days in the ambient environment. The presented low-cost, ultra-flexible, highly stable, and large-area photodetectors will lay a strong foundation for developing next-generation wearable and portable optoelectronic devices.

**METHODS**

Preparation of Pt–Au nanoparticles chains

Hundred and thirty microliters of 4.5 mg mL\textsuperscript{-1} platinum (IV) tetrachloride (PtCl\textsubscript{4}, Sigma-Aldrich) were mixed with 1 mL citrate capped gold nanoparticles (~10 nm) solution, purchased from BBI Solutions OEM Jones, respectively. This is among the best-reported performance with high mechanical durability when compared to other flexible perovskite photodetectors. Moreover, the PS incorporation in MAPbI\textsubscript{3} showed long-term device stability for more than 30 days in the ambient environment. The presented low-cost, ultra-flexible, highly stable, and large-area photodetectors will lay a strong foundation for developing next-generation wearable and portable optoelectronic devices.

Preparation of MAI precursor

Methylammonium iodide (MAI) was synthesized by dropwise addition of 30 mL of hydroiodic acid (Sigma-Aldrich, 57 wt. % in water) to 27.8 mL of hydrogen iodide (Sigma-Aldrich, 57 wt. % in water) under constant
constant bias of 2 V under 650 nm light irradiation of 0.1 mW cm\(^{-2}\) intensity during the bending process. The inset shows the corresponding optical images of the device under different bending angles. Normalized photocurrent of the flexible devices after different bending cycles at a bending angle of 120° showing the mechanical stability and durability of 1 wt % PS-MAPbI\(_3\) device even after 10000 bending cycles. Normalized photocurrent of the devices with an adhesive layer after aging continuously in ambient air, 35–40% relative humidity, and under room light for 720 h (30 days). Operational stability of the devices at a constant bias of 2 V under 650 nm light irradiation of 0.1 mW cm\(^{-2}\) intensity. Typical measurements were conducted on multiple devices (at least 4) and yield an error range of 3–5% for all the samples.

**Preparation of plain MAPbI\(_3\) and PS-MAPbI\(_3\) solutions**

The 1.35 M of plain MAPbI\(_3\) solution (without PS) was prepared by mixing 230.5 mg of lead iodide (PbI\(_2\)), 79.5 mg of MAI in 53.3 µL of dimethyl sulfoxide (DMSO) and 317.5 µL of dimethylformamide (DMF) for 1 h. To prepare the 0.5, 1, 3, and 7 wt % PS-MAPbI\(_3\) solutions, the required amount of PS (PS, Alpha Chemistry, \(M_\text{w} = 45,000\)) was dissolved in DMSO and DMF and stirred constantly for 30 min. Then, MAI and PbI\(_2\) (1/1 by molar) were added to the above PS solution under constant stirring for 1 h (time for crosslinking) at room temperature. The entire process was carried out in ambient conditions.

**Fabrication of flexible photodetectors**

The polyamide filter membranes with a 25 mm diameter were purchased from GE Healthcare. The self-assembled Pt–Au nanochains solution was used to deposit the flexible and highly conductive Pt–Au electrodes on the polyamide membrane through the vacuum-filtration process. The PDMS mask was used during the vacuum filtration to obtain a certain Pt–Au patterned electrode. The distance between the Pt–Au electrodes was 1.6 mm, the thickness of Pt–Au electrodes was around 550 nm and was kept constant for all samples. 90 µL of plain MAPbI\(_3\) or PS-MAPbI\(_3\) solution was then spin-coated on the polyamide filter membrane patterned with Pt–Au electrodes at 4000 rpm for 30 s. After 8 s of rotation, 200 µL of diethyl ether was dropped onto the center of the flexible polyamide substrate. The obtained films were then heated at 100 °C for 10 min to form the perovskite phase. The thickness of the MAPbI\(_3\) perovskite film with and without PS was around 1 µm (±50 nm). Finally, transparent self-adhesive tape was used to cover the perovskite films. The device configuration was Pt–Au/PS-MAPbI\(_3\) (or MAPbI\(_3\))/Pt–Au on the polyamide filter.

**Characterization and measurement**

The surface morphology of the self-assembled Pt–Au nanoparticles, polyamide filter, and perovskite films was observed by FESEM (FESEM, Zeiss Ultraplus). Transmission electron microscopy (TEM) images were obtained with a LEO 912AB transmission electron microscope. The phase of the perovskite samples was characterized by X-ray diffraction (XRD) using a PANalytical Empyrean diffractometer with Cu-Kα radiation (λ = 1.54 Å). The optical absorption spectra were recorded using a UV–Visible spectrophotometer (Perkin Elmer Lambda 750). The photoelectrical measurements of the device under different light wavelengths and intensities, including I–V, photocurrent, dark current, spectra responsivity, detectivity, and EQE were recorded by a Keysight 3458 A Digital multimeter combined with 6614 C 50 Watt system power supply from Agilent Technologies in ambient condition. All the measurements on Pt–Au/PS-MAPbI\(_3\) (or MAPbI\(_3\))/Pt–Au devices were conducted on the probing station by using a two-probe method. The voltage was applied on the plain MAPbI\(_3\) or PS-MAPbI\(_3\) porous films by connecting one probe to the first Pt–Au electrode on the filter and another probe connected to the second Pt–Au electrode. Simulated AM 1.5 G irradiation (100 mW cm\(^{-2}\)) with a xenon-lamp based solar simulator (Newport Oriel Instrument 67005, 150 W Solar Simulator) was used as the illumination source. The Newport optical filters were used to produce incident light at various intensities. The light intensity was calibrated by an NREL calibrated KGS silicon reference cell to minimize spectral mismatch. All measurements were conducted at room temperature and in ambient air. The active area of the devices was 1.2 cm\(^2\). For the bending test, an MFA motorized miniature linear stage was used to bend the flexible device at a particular angle and to perform 10,000 bending cycles at a constant speed. The long-term stability test of the devices was tested by directly exposing the devices in ambient air with 35–40% relative humidity and under room light. The humidity was
measured by a portable RH sensor and was controlled by adjusting the flow rate of the carrier gas (dry N2).

DATA AVAILABILITY
All relevant data in this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS
R.S. and H.F. contributed equally to this work. V.M. and R.S. planned the idea, R.S. and H.F. conducted the experiments, and all authors analyzed and discussed the results and contributed to the writing of the manuscript.

COMPETING INTERESTS
The authors declare no competing interests.
