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Original Article

A novel and stable ultraviolet and infrared intensity sensor in impedance/capacitance modes fabricated from degraded CH$_3$NH$_3$PbI$_3-x$Cl$_x$ perovskite materials

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Abstract

The present situation of COVID-19 diverted our focus towards utilizing the degraded solar cells for sensor application, this will help in global energy harvesting. So, here is our successful effort to reuse already degraded solar cells as ultraviolet (UV) and infrared (IR) sensor. The spin-coated perovskite (CH$_3$NH$_3$PbI$_3-x$Cl$_x$) has been already tested for visible light spectrum, as an extension to that now it is utilized as UV and IR intensity sensors to cover the whole spectrum. The employed CH$_3$NH$_3$PbI$_3-x$Cl$_x$ material was used after its efficiency loss has been reached to a saturation point in photovoltaic devices. Each deposited layer was investigated from UV to the IR absorption spectrum for deepening study through UV–vis spectroscopy. In the sandwiched architecture possessing FTO/PEDOT: PSS/Perovskite/PC$_61$BM/CdS/Au symmetry, the perovskite film has been employed as an absorbent layer, however, other layers participation also plays a key role. The resultant device yielded very good sensing performance because of the enhanced excitons generation which is attributed to the precise selection of the interfacial materials, e.g. CdS and PC$_61$BM as an ETM and PEDOT: PSS as HTM. The impedance and capacitance of the devices within 0.01–200 kHz under varied UV and IR illumination intensities were investigated. Measurements showed that as the intensity of the light increased i.e., UV (0–200 W/m$^2$) and IR (0–5800 W/m$^2$), impedance decreased while capacitance increased. The current results are attributed to the increase in the concentration of charges i.e., electron-hole pairs generation depending on the built-in capacitance and frequency of the charges.

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1. Introduction

Highly sensitive perovskite-based light detectors are extensively researched in the optoelectronic applications where detection of multicolor light is very much desired, such as day/night-time surveillance, imaging, communication, biological sensing, machine vision, and environmental monitoring [1–3]. Hence, it is desired to detect the ultraviolet (UV) and infrared (IR) spectrum too along with the visible spectrum as an extension to our previous efforts [4]. Although, there the perovskite solar cell was considered for visible light characterization and mesmerizing results were observed. For instance, enormous consumer electronics utilize visible light owing to the high sensitivity of human eyes in the visible spectral region. However, information communication [5], detecting the existence of nearby objects [6], injury treatment such as wounds and ulcers [7] are the applications where the visible spectrum is not effective. The most commonly used commercial light sensors employ crystalline inorganic semiconductors, such as Si, GaAs, GaP, Ge, PbS, and InGaAs [8,9]. However, the photo response of Si-based detectors is limited due to the bandgap of the active material, which ultimately cut-off the long-wavelength at around 1.1 μm [10] and Ge, PbS, and InGaAs detectors possess substantially higher cost and cooling requirements [1]. Hence, the lost cost and low noise sensors covering a larger spectrum, particularly, the UV and IR regions could play an important role in developing the light-sensing.

UV and infrared IR sensors are regarded as an integral part of flame sensors, missile detectors, and real-time measurements. The importance of the UV and IR sensitive devices could be justified because the sunlight is 44% visible light at ground level, 3% UV and rest infrared. 77% of the UV irradiations from the Sun are blocked by the atmosphere. However, in space, at the top of Earth’s atmosphere, about 50% is IR light, 40% is visible light, and 10% UV, for a total intensity of about 1400 W/m² in vacuum. Thus, for future space electronic technology, it is important to fabricate electronic devices that are additionally sensitive to UV and IR. At the same time, it would be practically useful to investigate the effect of UV and IR irradiation on the properties of the perovskite. This could prove useful for the development of the technology of solar cell fabrication on the base of selected organic semiconductor blend. The inorganic semiconductors-based UV/IR sensors are preferred due to their quick response and stability; however, the organic semiconductors-based UV and IR sensors are favored as they are cheap, their deposition techniques are easy, and they possess low cast fabrication and flexibility [11–13]. Hence, the researchers are trying to fabricate devices with hybrid organic-inorganic materials. Recently, the hybrid lead halide perovskites-based devices have emerged as the novel and one of the most promising technology, such as the mixed halide CH3NH3PbI3-xClx perovskite owing to its direct and easily tunable bandgap, solution process ability [14], the longer diffusion length of charge carriers and broad light absorption plus the longer lifetime of the charge carriers and the large carrier mobility [15–17]. Although, the usage of perovskite materials is highly concentrated in photovoltaic cells due its high efficiency typically above 22% [18–20], however, these impressive results have attracted an enormous interest towards the other applications such as in lasers [21], photodetectors [22,23] and light-emitting diodes [24,25].

Though perovskite-based devices have numerous advantages, however, the challenging issue in their commercialization is their fast degradation as compared to other solar cell materials such as organic photovoltaic materials. Although the organic cation is highly desired due to its excellent photovoltaic characteristics such as bandgap, excitons binding energy, and carrier lifetime [26], on the other hand, its chemical instability proves to be a bottleneck in the widespread commercialization of perovskite-based solar cells [27]. The economic viability of the perovskite solar cells is strongly dependent on the degradation rate of the material and despite the high efficiency, the faster degradation increases the overall operational cost [28,29]. However, the utilization of degraded perovskite material in the application such as photodetectors or perovskite-based sensing devices can be a novel route towards the cost-effective and environment-friendly perovskite technology [30,31].

In the context of the aforementioned discussion, the present work as an extension to our previous efforts where the perovskite photodetector was characterized for its visible spectrum [4]. Here, we demonstrate the efficacy of the degraded perovskite solar cell bearing the structure illustrated in Fig. 1, as the efficient and stable sensor to sense UV and IR intensity effects in impedance/capacitance modes. Usage of organic-inorganic hybrid materials as the conductive films in UV or IR sensors requires sturdy absorption, large mean free path, and well-matched Fermi level suitable for both donor and acceptor materials. Key points for the fabrication of UV and IR sensors are a focus on the strong absorption of material, high mobility of carriers, and precise selection of energy levels for a selection of donor and acceptor materials. The perovskite device after reaching to the saturation point in the efficiency loss is no longer useful in the solar cell application which if retains the reasonably good aforementioned properties for other perovskite technologies such as the present work, then it will not only mitigate the major environment lead contamination issue but also reduce the overall fabrication cost and time, making the technology economically more viable. So, the work is comprised of the UV and IR sensors based on degraded perovskite material. In the FTO/PEDOT-PSS/Perovskite/PC61BM/Cds/Au device structure, the symmetry of HTL and ETL with perovskite layer for the device is established to enhance the stability and transport rate of charges.

2. Experimental section

2.1. Materials and device fabrication

Perovskite (CH3NH3PbI3-xClx), Poly (3,4-ethylene dioxythiophene)—Poly(3,4-ethoxy sulfonate) (PEDOT:PSS), Cadmium Sulphide (CdS), and [6,6]-phenyl-C61-butyric acid methyl ester (PC61BM) were purchased from Ossila. The glass substrates with pre-deposited FTO were cleaned in soap water, acetone, ethanol, and distilled water for 10 min each in the ultrasonic bath. They were nitrogen blow-dried followed...
by oxygen plasma cleaning for 5 min. The cleaned substrates were insulated from the sides to avoid the electrodes short-circuiting.

The thin hole transport material (HTM) was deposited by spin-coating the PEDOT: PSS solution over the FTO coated glass for 30 s by Laurell WS-650-23NPP spin coater at 2000 rpm and subsequently annealed at 120 °C for 10 min. Then the absorbent layer was deposited by spin coating the perovskite over the HTM at 3000 rpm for 30 s, however, during the last 12 s, 150 μL chlorobenzene was rapidly dispensed. When chlorobenzene was dropped over the perovskite layer, it immediately changed from a pale yellow color to dark brown which confirms the perovskite formation as mentioned by us in Ref. [4]. Afterward, the slides were further annealed at 100 °C for an hour. Subsequently, the PC61BM (electron transport material (ETM) dissolved in chlorobenzene) was prepared and spin-coated over the annealed perovskite layer at 1500 rpm for 30 s. The devices were transferred to the thermal evaporator chamber where CdS was set down at 10⁻⁶ mbar over PC61BM. Then finally gold electrodes were deposited as shown in Fig. 1.

2.2. Characterizations

All characterizations were performed in the cleanroom and at ambient conditions as shown in Fig. 2. The UV and IR light intensity were measured by Lutron UV-340A and Lingshang LS122 IR solar power meter. The UV and IR lamps were used as a source of light.

Impedance and capacitance in the frequency range of 100 Hz–200 kHz were measured by LCR meter MT 4090. Beckman DU640 UV/V was utilized to perform the UV-absorption spectroscopy.

3. Results and discussions

Fig. 1 represents a schematic of the device architecture of the perovskite-based sensor. Also, the corresponding energy levels of the fabricated multifunctional sensor are presented in Fig. 3 that shows the conduction band, valence band, highest occupied molecular orbital (HOMO) level, lowest unoccupied molecular orbital level, and work functions (WF) of the materials being utilized. It is noteworthy that the energy levels of the materials were obtained from the literature. Further, PEDOT: PSS was used as HTM to enhance the work function of the FTO anode to ensure efficient hole extraction. As well, cadmium sulfide (CdS) was used as an additional ETM material along with the PC61BM.

3.1. Characterization of deposited perovskite film

For the enhanced performance of the devices, deep insight study along with improved morphology with full surface
coverage are critical factors, and hence, they are important to characterize at each layer. For the photosensitive materials, absorbing the solar light is critical for the enhanced device performance, therefore, all layers were characterized by UV–vis spectroscopy to measure the absorption spectrum in the range between 200–1300 nm. It is observed that CH$_3$NH$_3$PbI$_{3-x}$Cl$_x$ perovskite film possesses appreciable absorption in the broader spectrum within 200–900 nm as shown in Fig. 4, indicating the fact that it is one of the suitable materials for photosensitive devices [34]. In addition to that it is observed that perovskite has almost no absorbance after 900 nm, however here PEDOT: PSS layer is aiding it in the absorption of light in the infrared region. Similar results were observed by morphological analysis of the samples through atomic force microscopy of the samples, where depositing perovskite layer over PEDOT:PSS increase the roughness of the sample resulting an increase in the surface area aiding in anti-reflection properties of the device Ref. [4]. Hence, confirming the correct selection of supporting layers.

It is noteworthy that the absorption of each layer should also be considered to analyze the achieved experimental results of the finalized device.

3.2. Performance evaluation of CH$_3$NH$_3$PbI$_{3-x}$Cl$_x$ based sensors in impedance/capacitance modes

The devices were fabricated as illustrated in Fig. 1 and the complete fabrication procedure has been described in "EXPERIMENTAL SECTION" Fig. 2. The resultant device showed high sensitivity to sense ultraviolet and infrared Intensity both in impedance and capacitance modes. In this context, the electronic analysis of the sensors was performed in impedance and capacitance modes to varying UV and IR intensities from 100 Hz up to 200 kHz of frequencies.

The characterization has been repeated four times to confirm the repeatability of the sensors and the plotted graphs are the averaged response of the fabricated sensors. Fig. 5 shows the impedance dependence at different frequencies i.e., (a) 100 Hz, (b) 1 kHz, (c) 10 kHz, (d) 100 kHz, and (e) 200 kHz of the FTO/PEDOT: PSS/Perovskite/PC$_{61}$BM/CdS/Au sensors as a function of the varied UV irradiation intensities, which decreases from 7.4 to 0.27 MΩ depending on the respected frequency range.

It is seen that as the intensity of the UV irradiation increases from dark to 200 W/m$^2$ there is a drop in impedance depending on the frequency of the applied voltage during measurement. The impedance decreased by a factor of 3.33 (at 100 Hz), 3.29 (at 1 kHz), 1.46 (at 10 kHz), 1.35 (at 100 kHz), and 1.28 (at 200 kHz), respectively. This shows that higher fre-
 Frequencies are less affected by the increasing intensity of the UV irradiation, which decreased from a factor of 3.33 in the case of 100 Hz to 1.28 in the case of 200 kHz. The almost steady response has been recorded at higher frequencies from 100 Hz to 200 kHz. This is since at lower frequencies, charges get time to settle down and respond to the change, however at higher frequencies owing to the rapid change the results remain slightly changed and seem to be static.

Fig. 6 represents the capacitance characterization with respect to UV light and it was observed that the capacitance of the samples increased by a factor of 1.28 (at 100 Hz (a)), 1.25 (at 1 kHz (b)), 1.23 (at 10 kHz (c)), 1.224 (at 100 kHz (d)) and 1.222 (at 200 kHz (e)). The response is non-linear to some extent at lower frequencies and quasi-linear when the frequencies are higher. Similar to the impedance mode, the capacitance sensitivity values are higher at low frequencies and are significantly small at high frequencies. However, the capacitance values are generally observed to follow an increasing trend from 1.2 to 245 nF depending on frequency range as the function of increasing UV light intensity.

Moreover, the devices were also characterized by their impedance- and capacitance-IR relationships at varying frequencies. Fig. 7 shows the impedance dependence of the FTO/PEDOT: PSS/Perovskite/PC_{61}BM/CdS/Au samples on IR intensity at different frequencies from 9.1 to 2.16 kHz, respectively. The results show an inverse trend while infrared frequency is increased, e.g., as the IR frequency is increased, the impedance decreases by a factor of 4.15 (at 100 Hz (a)), 3.105 (at 1 kHz (b)), 1.245 (at 10 kHz (c)), 0.76 (at 100 kHz (d)), and 0.355 (at 200 kHz (e)), respectively.

Contrary to the impedance mode, Fig. 8 shows that capacitance increased in direct proportion to that of increasing IR frequency. While the capacitance rises 16.615 (at 100 Hz (a)), 2.29 (at 1 kHz (b)), 0.155 (at 10 kHz (c)), 0.0313 (at 100 kHz (d)) and 0.0118 (at 200 kHz (e)) times, as the IR frequency is increased from dark to 5800 W/cm². Most importantly, results showed in all cases that as the frequency is varying from lower to higher the sensitivity is decreasing in all cases. Therefore, the fabricated sensor can be a useful device to operate at the lower frequency ranges as sensitivity is high, although the total variation is within the range of 0.03–50.5 nF.

It is widely reported in the literature that UV and IR illumination ultimately affects the properties of the materials and ultimately the device response too [35–38]. The effect from the UV and IR illumination could be reversible (concerning physical properties) or irreversible (concerning structures). In this context, the current results concerning the effect of UV and IR on the impedance and capacitance of the FTO/PEDOT: PSS/Perovskite/PC_{61}BM/CdS/Au sensors might be due to the increase in the generation of the electron-hole pairs. It is a reversible process owing to the recombin-
tion of charges, probably, due to the defects in the layers. The sensing responses recorded in Figs. 5 to 8 are probably due to an increment in the reactive current which passes through the capacitor concerning the active current passes through the resistor when the illumination intensities have been increased. For further explanation of this phenomenon, the equivalent mathematical model of the sensor is presented in our previous work [4]. However, at higher frequencies, the light-impedance relationship can be explained by the frequency dependence of the mobility of the charges, which was also investigated by Prins et al. [39].

The sensitivity (S_{Zl} and S_{Cl}) is believed to be a significant way to measure the change in impedance and capacitance as the function of varied UV and IR illumination intensities. The governing equations are presented below:

\[ S_{Zl} = \frac{\Delta Z}{\Delta L} \]  

\[ S_{Cl} = \frac{\Delta C}{\Delta L} \]  

where \( \Delta Z, \Delta C \) refers to the change in impedance and capacitance, while \( \Delta L \) is a change in UV or IR intensities. The measured sensitivities of the UV and IR sensor to impedance and capacitance are tabulated in Table 1 below.

![Fig. 8 - Capacitance response of the fabricated samples on IR radiation at (a) 100 Hz, (b) 1 kHz, (c) 10 kHz, (d) 100 kHz, and (e) 200 kHz frequencies.](image)

| Frequency (kHz) | \( S_{Zl}(\text{UV}) \) (pF/m²/W) | \( S_{Zl}(\text{IR}) \) (pF/m²/W) | \( S_{Cl}(\text{UV}) \) (pF/m²/W) | \( S_{Cl}(\text{IR}) \) (pF/m²/W) |
|----------------|------------------------------------|---------------------------------|--------------------------------|----------------------------------|
| 0.100          | –27.95                             | 265                             | –0.715                         | 2.864                            |
| 1              | –6.965                             | 17.0                            | –0.535                         | 0.394                            |
| 10             | –1.290                             | 10.3                            | –0.214                         | 0.026                            |
| 100            | –0.525                             | 2.20                            | –0.131                         | 0.005                            |
| 200            | –0.380                             | 1.40                            | –0.061                         | 0.002                            |

The results showed that \( S_{Zl} \) and \( S_{Cl} \) led to a drop as the frequencies increases. This might be due to the capacitance which can also be seen from Eq. (3) and well confirmed from the literature [4].

Similar to the reported work by Nahar and Khanna [40] related to the behavior of the sensor in both the capacitive and the resistive mode as a function of relative humidity, the parallel combination of impedance and capacitance can represent the mathematical model of the perovskite-based multifunctional sensor's channel through Eq. (3). Equation (3) represents capacitance (C), as the sum of the channel dielectrics i.e., air-filled pores and the active layer, both of which are sensitive to light. Additionally, the cumulative resistance is presented by \( R \), which is also a function of varied UV and IR light intensities [41].

\[ Z = \frac{R}{1 + jwRC} \]  

Whereas, \( w \) is the circular frequency. The decreased impedance and increased capacitance can be justified because an increase in the illumination intensity also increases the charge carrier's concentration, resulting in a change in the permittivity [42,43]. It is noteworthy that electronic polarizability, i.e., electronic (\( \alpha_e \)), dipolar (\( \alpha_dip \)), ionic (\( \alpha_i \)) and charge carriers (electrons and holes) transfer (\( \alpha_{tr} \)), all of which depends on the concentration of charge carriers and directly affecting the capacitance of the active channel [44-46]. Among all four aforementioned polarizabilities, \( \alpha_{tr} \) is due to the orbital electron's relative displacement, where the participation of charges in the conduction process results in the \( \alpha_{tr} \). However, if we take into consideration only charge carriers transfer polarizability, the Clausius-Mosotti relation can be represented by Eq. (4) as [44].

\[ \frac{\varepsilon - 1}{\varepsilon + 2} = \frac{N\alpha_{tr}}{\varepsilon_0} \]  

where \( \varepsilon \) is the relative permittivity, \( N \) is the total charge carrier's concentration, and \( \varepsilon_0 \) is the permittivity of free space, Fig. 9 represents the experimental results of capacitance transient concerning the response time (\( t_{res} \)) and recovery time (\( t_{rec} \)) for the UV/IR sensor under pulsed UV and IR illumination measured at 100 Hz. When the UV light intensity changed from 0 W/m² to 180 W/m² the averaged \( t_{res} \) and \( t_{rec} \) is 1.4 s and 1.8 s, as shown in Fig. 9a. However, when IR light intensity pulses are introduced at 5500 W/m² from the dark condition as seen from Fig. 9b, then the observed \( t_{res} \) and \( t_{rec} \) are 0.5 s and 0.6 s, respectively. The initial peak in both cases is high, this might be due to the recombination of charges and partially due to the surface charges which settled down later. Each sensor read-
ings were conducted three times. The results were repetitive even after a period of one month with the degradation of 4% from the values measured as fresh samples, therefore all the graphs are based on averaged data.

Fabricating and designing new devices not only need money but also time-consuming. However, if we take an initiative to recycle the already available degraded devices for other applications, it will help in energy harvesting. In addition to that, they will provide a chance to get benefit from already existing devices for a longer period in different ways. So, in a situation where globally or locally, the movement is limited, like a present pandemic of COVID-19 along with the development of new devices, if the old degraded gadgets can be reuse will be beneficial too. Here in this article, we tried to recycle an already degraded perovskite solar cell for its UV and IR sensing properties. Hence, as an extension of our previous work, where we had utilized the degraded solar cell as a visible light sensor from 350 to 850 nm wavelength [4]. This study is performed to deepen the explore of that degraded sample. Here, the device is characterized by 200–1300 nm wavelength to cover the ultraviolet and infrared effect. The experiments result in fascinating results and deep each layer. Details explain how we can design perovskite sensors and solar cells which can cover a large spectrum of light. One of the key obstacles in the development of perovskite-based devices is its degradation due to water, temperature, and light intensity. However, if we can utilize these materials for some other multifunctional applications even after degradation, such as the present work, where degradation does not significantly affect the electric parameters, then the material cost can be significantly reduced owing to the utilization of the materials even after degradation without compromising the performance.

4. Conclusion

In summary, this work performs a detailed investigation of the effects of UV and IR illumination intensity on the impedance and capacitance of a perovskite-based sensor. With the most commonly utilized materials, we engineered a sandwich structured sensor by employing facile fabrication steps and possessing the FTO/PEDOT: PSS/Perovskite/PCBM/CdS/Au symmetry. The impedance and capacitance characterizations were performed at 100 Hz, 1 kHz, 10 kHz, 100 kHz, and 200 kHz. The results obtained confirmed the high response at lower frequencies. An increase in the UV/IR light intensity is directly proportional to the capacitance and inversely proportional to the impedance. However, a sharp decrease in the sensitivities was observed in the impedance and capacitance of the sensors to increase the capacitance. Besides, this phenomenon of enhanced capacitance and decreased impedance can be attributed to the increase in the charge carrier’s concentration in the absorbent layer, with an increase in the UV/IR intensities, which results in a reactive and active current of the devices. Therefore, the present concept offers a simple, facile, and efficient development of the UV/IR responsive sensor with reliable characteristics even after the degradation of the perovskite material. The results presented here thus constitute a further step towards the fulfillment of improved and large-area sensors possessing the high sensing performance while maintaining the low cost and economically viable solution especially in situations like COVID-19, of perovskite-based technologies.

Conflicts of interest

The authors declare no conflicts of interest.

CRediT authorship contribution statement

Tahseen Amin Qasuria: Conceptualization, Project administration, Resources. Noshin Fatima: Methodology, Investigation, Writing - original draft, Writing - review & editing. Khasan S. Karimov: Supervision. Mohd. Adib Ibrahim: Validation.

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