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### Supplementary data

### **Supplementary Information**

# Lightweight flexible self-powered photo-supercapacitors with good stability through photoelectrochemical deposition of tellerium on PPy-V<sub>2</sub>O<sub>5</sub> films as new visible light active dual photoelectrode

Mohamad Mohsen Momeni<sup>a\*</sup>, Hossein Mohammadzadeh Aydisheh<sup>a</sup>, Byeong-Kyu Lee<sup>b\*</sup>, Ali Naderi<sup>b</sup>

<sup>a</sup> Department of Chemistry, Isfahan University of Technology, Isfahan 84156-83111, Iran

<sup>b</sup> Department of Civil and Environment Engineering, University of Ulsan, Daehakro 93, Namgu, Ulsan, 680-749, Republic of Korea

<sup>\*</sup>Corresponding author. Email address: mm.momeni@cc.iut.ac.ir

<sup>\*</sup>Corresponding author. Email address: bklee@ulsan.ac.kr

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### Synthesis of V<sub>2</sub>O<sub>5</sub> Nanowire:

Vanadium pentoxide ( $V_2O_5$ ) nanowires were prepared by a one-step hydrothermal method. First, 0.5 g of commercial pure  $V_2O_5$  powder was mixed with 25 mL of distilled water under stirring at 25 °C for

30 min. Next, 5.0 mL of 30%  $H_2O_2$  was added to the solution and stirring was further continued for an hour. The resulting orange solution was transferred into a 50 mL Teflon-lined autoclave and kept at 180 °C for 12 hours. Having been cooled to ambient temperature, the orange precipitate was washed with distilled water and annealed at 400 °C for 1.0 hour in air.

#### Chemical synthesis of polypyrrole

Polypyrrole (PPy) was prepared by a chemical polymerization method. First, a 2.0 mM pyrrole solution was prepared by dissolving distilled pyrrole in 60 mL of aqueous 1.0 M H<sub>2</sub>SO<sub>4</sub>. Next, the pyrrole solution was cooled on an ice bath, followed by the slow addition of 60 mL of ammonium persulfate solution (APS) as the oxidizing agent, with a molar ratio of 1.2:1 to pyrrole over 40 min. The reaction mixture was kept at ambient temperature for 12 hours while being magnetically stirred. Last, the resulting PPy particles were filtered, washed with deionized water two times, and dried at 50 °C for 1.5 hours.

#### Fabrication of flexible Te@PPy-V<sub>2</sub>O<sub>5</sub> photo electrode:

The flexible Te@PPy-V<sub>2</sub>O<sub>5</sub> photoelectrode with supercapacitive activity, flexibility, transparency, and photosensitivity was prepared. Under optimal conditions, 0.058 g of synthetic V<sub>2</sub>O<sub>5</sub> nanowires, 0.031 g of PVDF powder (Kynar 761 type), as the binder, were dispersed in 15 mL of N-methyl-2-pyrrolidinone (NMP) for 60 min. to yield a homogeneous solution. The mixture obtained was then added dropwise to clean indium tin oxide (ITO) substrate, followed by drying at 50 °C to form a thin and flexible PPy-V<sub>2</sub>O<sub>5</sub> photoelectrode. Last, to fabricate Te@PPy-V<sub>2</sub>O<sub>5</sub> electrode, Te was deposited on the PPy-V<sub>2</sub>O<sub>5</sub> substrate by a single-step electrodeposition and photoelectrodeposition method in a three-electrode setup. The working electrode comprised flexible PPy-V<sub>2</sub>O<sub>5</sub> electrodes as

well as Ag/AgCl and platinum sheet as the reference and counter electrodes, respectively. The Te growth solution was prepared by dissolving 0.14 M TeO<sub>2</sub> in aqueous 0.12 M HNO<sub>3</sub>. This solution was kept under stirring at 80 °C until becoming transparent. Afterward, both electrodeposition and photoelectrodeposition were performed by chronoamperometry in a *three-electrode* setup. A -1000 mV voltage was applied for varying time intervals (5.0, 10, 15, and 20 min.). Upon completion of the deposition, the Te@PPy-V<sub>2</sub>O<sub>5</sub> samples were carefully separated from the three-electrode system, thoroughly washed with distilled water to remove any lightly adhered tellurium from the flexible electrode, and dried in a vacuum oven at 60 °C.

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Sample	<b>Deposition Time (min)</b>	Methode	Temperature (°C)		
5.0 min E	5.0	Electrodeposition	80		
5.0 min PE	5.0	Photoelectrodeposition	80		
10 min E	10	Electrodeposition	80		
10 min PE	10	Photoelectrodeposition	80		
15 min E	15	Electrodeposition	80		
15 min PE	15	Photoelectrodeposition	80		
<b>20</b> min E	20	Electrodeposition	80		
20 min PE	20	Photoelectrodeposition	80		

 Table S1 Tellurium electrodeposition and photoelectrodeposition conditions (pH=3.5) and sample

 labelling

### Sample characterization

The phase purity and crystalline structure of the electrodes were determined by X-ray diffraction (XRD) (XRD, Philips XPert diffractometer) using Cu K $\alpha$ -filtered radiation. The morphology of the

electrodes prepared was investigated using high-resolution field-emission scanning electron microscopy (FE-SEM; QUANTA FEG 450 instrument). The elemental analysis of the samples fabricated was determined by energy dispersive X-ray spectroscopy (EDS) using EDX coupled with FESEM. A JEOL JEM-3010 instrument was used to obtain transmission electron microscopic (TEM) images using bright-field diffraction contrast mode at an applied voltage of 300 kV. A Kratos Axis Nova X-ray photoelectron spectrometer equipped with a monochromated Al Ka X-ray source (hu = 1486.6 eV) operating at 15 kV and 10 mA was used to perform X-ray photoelectron spectroscopy (XPS). All the binding energies (EB) were referenced with respect to C1s at 284.6 eV.

### Electrochemical and photoelectrochemical measurements

An Origaflex-OGF500-Potentiostat/Galvanostat instrument (Origaflex electrochemical workstation; France) was used to carry out all the electrochemical analyses including cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS). Using a three-electrode system and aqueous 6.0 M LiCl electrolyte, the initial electrochemical studies were performed to determine the mechanism of charge storage. Platinum wire and Ag/AgCl served as the reference and counter electrodes, respectively. Specific gravimetric capacitance (Cs, F/cm<sup>2</sup>) was determined based on the galvanostatic charge/discharge (GCD) tests (equation 1).

$$C_{sp} = \frac{I\Delta t}{S\Delta V} \tag{1}$$

In which, I,  $\Delta t$ ,  $\Delta V$  and S corresponds to the current (A), discharge time (s), potential window (V) and surface area (cm<sup>-2</sup>), respectively.

Energy and power densities ( $E_g$ , Wh/cm<sup>2</sup> and P, W/cm<sup>2</sup>, respectively) of the electrodes were calculated using the specific capacitance of the materials, (Eqs. 2 and 3, respectively):

$$E_{max} = 0.5 \ C (\Delta V)^2 / 3.6 \tag{2}$$

$$P_{max} = \frac{E_{max}}{\Delta t} \times 3600 \tag{3}$$

where  $C_{sp}$ ,  $E_{max}$ ,  $P_{max}$ , I, S,  $\Delta t$ , and  $\Delta v$  are the specific capacitance (mF/cm<sup>2</sup>), energy density (Wh/cm<sup>2</sup>), power density (W/cm<sup>2</sup>), discharge current (mA), area of the active material (cm<sup>2</sup>), discharge time (s), and a voltage difference between the upper and lower potential limits (V), respectively [1, 2]. The flexible solid-state photosupercapacitor (FSSPC) was fabricated by sandwiching PVA-LiCl gel between the negative and positive electrodes. Before sandwiching, enough care was taken to assure the fabrication of a fine PVA-LiCl layer on one side of both electrodes. The layer cast in the middle of the electrodes hinders the short circuit. To prepare the gel, first, 1.0 g of PVA was dissolved in 10 mL of distilled water under continuous stirring at 80 °C to form a transparent solution. Next, 1.0 M LiCl was added dropwise to the PVA polymer solution while being continuously stirring. The resulting solution was stirred at room temperature to form a homogeneous and viscous gel electrolyte.



**Fig. S1** SEM images with two magnifications showing the top views of (a) 5.0 min PE, (b) 10 min PE, (c) 15 min PE and (d) 20 min PE samples.

#### Cross-section and transmittance and reflectance curve

Reflectance refers to the amount of light reflected from a surface or optical component, while transmittance indicates the amount of light transmitted through it. Fig. S2b presents the optical transmittance and reflectance spectra for Te@PPy-V<sub>2</sub>O<sub>5</sub>/ITO. The data shows that the trends of transmittance, T( $\lambda$ ), and reflectance, R( $\lambda$ ), for Te@PPy-V<sub>2</sub>O<sub>5</sub>/ITO can be categorized into distinct stages across a wavelength range of 200-850 nm. Initially, T( $\lambda$ ) increases sharply as wavelength ( $\lambda$ ) increases, reaching a maximum value of approximately 8.5 at  $\lambda$  = 350 nm, after which it gradually declines to a plateau level of about 7.8 at higher wavelengths. Conversely, the increase in T( $\lambda$ ) is attributed to the presence of tellurium (Te), while R( $\lambda$ ) exhibits an opposite trend. Notably, when T( $\lambda$ ) reaches its maximum, R( $\lambda$ ) shows minimum values in the wavelength range of 330-380 nm, as illustrated in the figure.





Fig. S2 (a) SEM images showing a cross-section of the 10 min PE sample and (b) transmittance and reflectance versus wavelength for this sample.









Fig. S5 SEM image, EDX mapping of elements C, O, N, V and Te in Te@PPy- $V_2O_5$  and its EDX spectrum.

Element	Bond	Peak position/eV	FWHM/eV
	V 2p <sub>3/2</sub>	517.31	1.67
V	V 2p <sub>1/2</sub>	524.68	1.92
	O 1s	529.9	1.58
	Сβ	282.51	1.42
	C-C	282.87	181
С	=C-N <sup>+</sup>	284.91	1.82
	C=O	288.87	1.09
	Te 3d <sub>5/2</sub>	575.76	1.73
Те	Te 3d <sub>3/2</sub>	586.2	1.55
	O <sub>2</sub> -2	531.22	2.69
Ο	-OH	533.21	1.58
	Н-О-Н	534.02	1.54
Ν	С=Н	399.37	1.75

Table S2 The peak positions and FWHM for all the XPS peak fittings



**Fig. S6** Specific capacitance of different electrode based on GCD curves (a) in dark (b) under light. **Effect of electrolyte concentration** 

The electrolyte, which consists of a salt and a solvent, plays a central role in electrochemical investigations. It provides ionic conductivity and facilitates charge equalisation at each electrode in the cell [3]. Aqueous electrolytes are superior to organic electrolytes due to their better compatibility and higher ionic conductivity [4, 5]. First, we investigated the electrochemical performance of the Te@PPy-V<sub>2</sub>O<sub>5</sub> electrode using different concentrations (1 M, 3 M, 6 M and 8 M) of aqueous LiCl electrolyte, as shown in Fig. 7. The cyclic voltammetry (CV) curves for this electrode were plotted in a potential window from -0.8 V to 0.3 V using LiCl electrolytes with concentrations of 1 M, 3 M, 6 M and 8 M. These curves have a symmetrical shape and were obtained at a sweep rate of 25 mV/s. It is noteworthy that the current density increases with increasing LiCl concentration from 1 M to 6 M, followed by a subsequent decrease. The observed current density values from the CV curves are 0.05 mA/cm<sup>2</sup> (1 M LiCl), 0.07 mA/cm<sup>2</sup> (3 M LiCl), 0.13 mA/cm<sup>2</sup> (6 M LiCl) and 0.10 mA/cm<sup>2</sup> (8 M LiCl). At low electrolyte concentrations (1 M and 3 M), the current density is significantly reduced due to a lack of ions participating in the redox reactions. Conversely, the increased availability of ions at a higher concentration (6 M) leads to a higher current density [6]. However, if the electrolyte concentration increases further, the current density decreases. This phenomenon is due to the starvation of the electrolyte, in which almost all ions are adsorbed at the large-area interface. This increases the internal resistance effect, which affects the overall state of charge. This behavior, observed by Zheng and Conway [7, 8], explains the suboptimal performance at higher concentrations. Therefore, all subsequent electrochemical studies were carried out with a 6 M LiCl electrolyte.



**Fig. S7** (a) CV and (b) GCD curves of Te@PPy-V<sub>2</sub>O<sub>5</sub> electrode at scan rate of 50 mV/s and current density of 2 mA.cm<sup>-2</sup> for 1M, 3 M, 6 M and 8 M LiCl electrolyte concentrations.



**Fig. S8** Specific capacitance of Te@PPy-V<sub>2</sub>O<sub>5</sub> electrode as a function of LiCl electrolyte concentration (a) based on CV data and (b) based on GCD curves.



Fig. S9 CV curves of best electrode at different scan rates in light condition



Fig. S10 CV curves of best electrode with different working voltage window at 10 mV/s in light condition



Fig. S11 Specific capacitance of different electrode based on GCD curves.



**Fig. S12** Specific capacitance of Te@PPy-V<sub>2</sub>O<sub>5</sub> electrode *Vs.* potential based on GCD curves (a) in dark (b) under light condition.



Fig. S13 The specific capacitance of best electrode at various current densities that optical charging

Sample	Flat band potential (Efb/ V)	Donor density (ND/cm <sup>3</sup> )
5.0 min PE	0.21	$3.03  imes 10^{29}$
5.0 min E	0.20	$1.88 \times 10^{29}$
10 min PE	0.1	$1.85\times10^{31}$
10 min E	0.12	$4.24\times10^{30}$
15 min PE	0.13	$3.7 \times 10^{30}$
15 min E	0.14	$3.37\times10^{30}$
20 min PE	0.17	$5.94\times10^{29}$
20 min E	0.18	$4.64 \times 10^{29}$

**Table S3** The flat band potential and carrier concentration values obtained from the linear fits performed for different samples



Fig. S14 (a) CV of Te@PPy-V<sub>2</sub>O<sub>5</sub> sample at a scan rate of 3.0-10 mV/s. (b) Relationship between the logarithm anodic peak current and logarithm scan rates. (c) Square root of scan rate ( $v^{0.5}$ ) vs I/ $v^{0.5}$  graph at a potential of 0.1 V for k<sub>1</sub> and k<sub>2</sub> determination (d-g) Capacitive-controlled and diffusion-controlled contribution process at a scan rate of 3, 5, 8,10 mV/s. (h) Percentage distribution of capacitance at different scan rates at dark condition.



Fig. S15 (a) CV of Te@PPy-V<sub>2</sub>O<sub>5</sub> sample at a scan rate of 3.0-10 mV/s. (b) Relationship between the logarithm anodic peak current and logarithm scan rates. (c) Square root of scan rate ( $v^{0.5}$ ) vs I/ $v^{0.5}$  graph at a potential of -0.1 V for k<sub>1</sub> and k<sub>2</sub> determination (d-g) Capacitive-controlled and diffusion-controlled contribution process at a scan rate of 3, 5, 8,10 mV/s. (h) Percentage distribution of capacitance at different scan rates at light condition.



Fig. S16 GCD curves of FSSPC at current density of 2.8 mA/cm<sup>2</sup> in different potential ranges in dark condition.



Fig. S17 The specific capacitance of fabricated FSSPC at various current densities that optical charging.



Fig. S18 A schematic illustration of the photo-charge and discharge mechanism in self-powered photosupercapacitors.



Fig. S19 Ragone plot showing the variation of energy density with the power density of FSSPC and other photo supercapacitors in dark and under light conditions.



Fig. S20 Specific capacitance of FSSPC at different temperature based on CV curves.



Fig. S21 Specific capacitance of FSSPC at different temperature based on GCD curves.



**Fig. S22** Charge-discharge cycling test and cycling stability performance of FSSPC at 3.2 mA/cm<sup>2</sup> for 12000 charge/discharge cycles at -10 °C and 0 °C. (Insets) First and Last three charge–discharge cycles.



**Fig. S23** Charge-discharge cycling test and cycling stability performance of FSSPC at 3.2 mA/cm<sup>2</sup> for 12000 charge/discharge cycles at 50 °C. (Insets) First and Last three charge-discharge cycles.



Fig. S24 Schematic wearable application of FSSPCS as the series to light 15 red LED.

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