Electronic Supplementary Information (ESI-1)

Fabrication of Graphene and Conductive Polymer Nanocomposites Coated Highly Flexible and Washable Woven Thermoelectric Nanogenerator

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Tables

Table.1 Properties of cotton fabric coated with rGO and PEDOT:PSS nanocomposites

Sample Code	Sheet Resistance (Rs)		Tensile Strength (mPa)		Water Contact Angle (WCA)	Air Permeability (cm³/cm²/s)	Fabric Thickness (µm)
Pure Cotton	Before Washing	After Washing	Warp	Weft			
GO Cotton	NiL	Nil	28	20	73.4	165	227.8
rGO Cotton	85	95	32	30	109.2	156	232.6
rGO-PES-1	65	75	39	35	121.3	148	236.4
rGO-PES-2	45	55	47	45	132.5	141	239.5
rGO-PES-3	25	35	63	60	139.4	135	227.8
rGO-PES-4	15	25	65	62	141.6	126	232.6

Note. Average results are taken for each sample for the test results.

Table 1 demonstrates, the Effect of different weight percent of rGO on sheet resistance before and after washing cycles, tensile strength and air permeability.

Tab 2. Effect of rGO content percent and number of padding passes effect on electrical and
thermoelectric properties

Sample No.	Comp	Number of	Sheet	Seebeck	Power Factor	Figure of Merit	
	(rGO %)	Padding	Resistance	Coefficient	(µW/mK²)	ZT Value	
		Cycles	(ΚΩ)	(μV/K)		10 ⁻³	
rGO/PES-1	2.5	10	185±5	2.5	0.12	0.01	
rGO/PES-2	5.0	10	165±5	8.5	0.15	0.02	
rGO/PES-3	10.0	10	145±5	12.0	0.20	0.03	
rGO/PES-4	20.0	10	125±5	15.6	0.25	0.04	

Note. Average results are taken for each sample for the test results.

Table.2 Demonstrates, the effect of rGO content percent and number of padding passes on electric sheet resistance and thermoelectric performance of Nano generator

Sample	C (%)	O (%)	c/o	Ref.
	70.0	24.4		[7.6]
Untreated	72.9	24.1	3.02	[74]
GO	73.7	22.4	3.29	[75]
rGO A.A	82.8	14.2	5.83	[76]
rGO NaBH₄	78.5	20.7	3.79	[77]
rGO H.H	83.6	13.3	6.26	[78]
rGO Na ₂ S ₂ O ₄	85.5	14.0	6.15	[79]
rGO (A.A) 10% coated 1-5 Dips	83.3	16.5	4.86	This work
rGO (A.A) 10% coated 5-10 Dips	85.7	13.4	6.24	This work
rGO (A.A) 10% coated (15-Dips)	87.4	10.6	8.0	This work

Table.3 Comparison for Elemental Analysis of developed rGO coated fabric using proposed method

Table.4 Thermoelectric performance of the developed device as compared to previously reported textilebased wearable and washable devices

Thermoelectri c Materials for TE devices	Type of Substrat e Fabrics	Electric al conduct ivity (S/Cm)	Therma I conduct ivity (W/m·K)	Seeeck Coeffic ient (µV K ⁻¹)	Power Factor (uW./ mK ²)	Figure of merit ZT	Output volts (mV)	Tempera ture gradient (K)	Ref.
PVDF/CNT	PVDF Fibers	14	0.22– 0.36	57.6	90	5.0×10 ⁻⁵	47.5	60.5	[80]
PVP-PEI/CNT- PEDOT:PSS	PVP Yarns	0.25- 5.0	0.32– 0.42	26.7	37.8	3.0×10 ⁻⁴	140	80-120	[81]
Bi2 Te3 Sb2 Te3	Silk Fibers	15	0.35– 0.75	16.5	15.20	5.0×10 ⁻³	10.0	35.5	[82]
PEDOT:PSS	Polyeste r	45	0.36- 0.42	33.5	12.29	5.0×10 ⁻²	24.3	66-120	[83]
WPU/PEDOT:P SS/CNT	Polyeste r	13.86	0.12- 0.20	10.0	1. 41	7.8 × 0⁻⁵	54.4	30- 39	[84]

	Woven								
Bi _{0.5} Sb _{1.5} Te ₃ , Bi ₂ Se ₃	Polyeste r	26.4	0.38- 0.42	28.5	4.4	0.09	55.5	20.4	[85]
	Knitted								
PEDOT:PSS/CN T,	PET Fibers	150.8	0.35– 0.86	25.82	32.4	0.08	143.7	80-116	[86]
PEDOT:PSS	MWCNT /PVP fiber	14.4	0.25– 0.50	15.0	65.5	0.42- 0.50	21.2	75.2	[87]
PEDOT-DMSO	Textile Fabrics	0.5-2.5	0.45- 0.60	156.2	41.2	0.06- 0.09	18.7	39.0	[88]
PEDOT:PSS/CN T/PEI	PU Fibers	45.6	0.45– 0.75	25.6	26.52	0.02- 0.05	6.5- 10.0	33.3	[89]
PEDOT:PSS/CN T	PEI	15.9	0.76– 0.42	65.8	15.22	0.03- 0.07	45.2	36.5	[90]
PEDOT:PSS,- AgNP	Cotton Woven	112.5	0.87– 0.58	45.8	35.22	0.07- 0.08	20.8	56.8	[91]
PEDOT:PSS- rGO/AgNWS	Nylon Fibers	245.5	0.25— 0.45	15.8	31.22	0.05- 0.09	12.5	20.0	[92]
rGO0PEDOT:P SS	Cotton fibers	365	0.15- 0.65	25.8	65	0.04- 0.08	120	16.5	This work

Note: Reference Materials are obtained from cross reference which may variable in results. Authors, may not assure the liability or any miss happen related to results presented in table.4

Electronic Supplementary Information (ESI-S2)

2. CHARACTERIZATIONS

The Scanning electron microscopy (SEM) analysis of the sample was performed by using a Zeiss ultra-scanning electron microscope (SEM) for the surface of the treated and untreated fabrics. Nicolet 5700 Thermofisher, (USA) was used for FTIR/ATR analysis of the GO, rGO, and PEDOT: PSS-rGO nano-composites films and coated fabric. A K-alpha mode on XPS (Thermofisher USA), X-ray diffraction (XRD), and (XPS) used to describe the rGO, GO, and PEDOT: PSS-rGO films and nano-composites. The NEXUS- XPS was recorded using an XPS spectrometer with a monochromatic Al K- α source of 1486.68 eV. The electrical performance of the rGO and PEDOT PSS decorated cotton fabric was measured by using a four-point probe (SZT-2B) system (Suzhou Genisis Electronics Co.Ltd., China). The average test results with a low sheet resistance of each sample were measured six times and recorded for statistical analysis. The electrical performance of the rGO and PEDOT PSS decorated cotton fabric was measured by using a four-point probe (SZT-2B) system (Suzhou Genisis Electronics Co.Ltd., China).

2.1 X-Ray Diffraction



Fig.1 X-Ray Diffraction analysis of Graphite a), Graphene Oxide (GO) b), reduced Graphene oxide (rGO) c) and PEDOT:PSS-rGO nanocomposites d)

2.2 FTIR Spectroscopy



Fig .2 FTIR Spectroscopy analysis of PEDOT:PSS-rGO nanocomposites a), PEDOT:PSS b), reduced Graphene oxide (rGO) c) and Graphene oxide (GO) d)

b) a) Graphite GO D D G G Intensity (a.u) Intensity (a.u) 2D 2D 2G c) d) rGO PEDOT:PSS-rGO D G Intensity (a.u) G Intensity (a.u) 2D 2G 2D 2G 500 1000 1500 2000 2500 3000 3500 4000 0 500 1000 1500 2000 2500 3000 3500 4000 Ó Wavenumber (1/Cm) Wavenumber (1/Cm)

2.3 Raman Spectroscopy

Fig .3 Raman Spectra analysis of Graphite a), Graphene Oxide (GO) b), reduced Graphene oxide (rGO) c) and PEDOT:PSS-rGO nanocomposites d)



3 XPS Spectroscopy

Fig:4 XPS analysis of reduced graphene oxide (rGO) and PEDOT:PSS-rGO showing C1s a) O1s b) N1s c) and S2P Peaks

Electronic Supplementary Information

Measurement of Textile Properties (ESI-S3)

- 1. Textile Properties
- **1.1 Sheet Resistance with Dyeing and Washing Cycles**
- 1.2 and Tensile Strength and Stretching and Bending Cycles



Fig:1 Sheet resistance with number of dyeing cycles and washing cycles a) Tensile strength with number of padding cycles b) Stretching and bending cycles

1.3 Weight pick up and Particle size distribution



Fig: 2 Weight pickup percentage and particle size distribution of GO, rGO and PEDOT:PSS0rGO nanocomposites on woven fabric

2. Thermoelectric Performance



Fig:3 Current Vs output voltage and power relationship a) Thermoelectric performance of the TE device with figure of merit, power factor and seebeck coefficient over temperature gradient of 2.5-36.5 C

- 3. Potential Biomedical Application and Health monitoring
- 3.1 Electrocardiogram (ECG) Response



Fig:1 Output Thermoelectric performance of Textile based Developed TE device, for real time biomedical and health monitoring application performance measurement for ECG electrodes attached to Digital Cardiogram device a) The placement of disposable electrode on commercially available ECG Probes placed on human wrist b) rGO, PEDOT: PSS coated woven Fabric as disposable ECG electrodes attached to volunteer wearing sports bra c) Performance as an Electrical output voltage mV AC signal response obtained and used for ECG performance of the developed device d) Output ECG signal response extended obtained ECG signal response e) for real time biomedical and health monitoring application. **Reproduced with copyright Material from Material & Design, 2020.**

3.2 Pressure Sensor Response

The response of the sensor with the change of sheet resistance to develop the PP sensor was connected with the electrodes to measure response. To record the response of the sensor, we used Instron micro-tester programmed and software Bench View 5.0. Following the method of ASTM D-5034 standard on Universal Testing Machine (UTM) Ortega to analyse the tensile strength of the samples. We used the Instron Model-335 to analyse tensile strength for rGO-coated PP samples whenever we increased the load resultant sheet resistance were also increased, and for the reducing of load, the sheet resistance was decreased. For the change of sheet resistance from 0.2% to 1.5%, the electrical conductivity was 1.5% recorded, as shown in Fig. 4.3a and it was the maximum. At different loading and unloading in the unit of kPa, the sensitivity response of the wearable pressure sensor was 0.050–0.089 kPa, which can detect the small deflection and pressure of human body movement.



3.3 Electrocardiogram and Pulse rate response and 3.4Pressure Sensor response

Fig: 2 ECG Signal response as recorded compared with original ECG Signal a) Pulse rate response detected on human wrist b) Pressure responses obtained with knee a) extended version of pressure response as resistive type sensor b) Pressure responses obtained with knee a) extended version of pressure response as resistive type sensor b) Pressure responses obtained with knee a) extended version of pressure response as resistive type sensor b) Pressure responses obtained with knee a) extended version of pressure response as resistive type sensor b) Reproduced with permission of copyright material of Sensors and actuators, and Materials & Design. 2020

Electronic Supplementary Information (ESI-S5)

Methods

1. Fabrication Pad Dry Cure



Fig:1 Fabrication of Graphene on Textile Substrate using Pad Dry Cure Unit for textile based Thermoelectric Nano generator

2. Designing of Thermoelectric Device



Fig.2 TE design model P- N type legs rGO, rGO/PEDOT: PSS Connected in parallel a) and Series b) arrangement with a closed-Circuit