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A QUICK AND FACILE SOLUTION-PROCESSED METHOD FOR PEDOT:PSS TRANSPARENT CONDUCTIVE THIN FILM

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ABSTRACT: PEDOT:PSS is a conducting organic polymer widely studied for a transparent conductive electrode. The conventional method to fabricate PEDOT:PSS thin film involves a post-treatment process entailing dipping into strong and toxic saturated acid to enhance the film's conductivity. Eliminating the post-treatment process reduces excess strong saturated acid or solvent waste, shortening the fabricating time by half. Therefore, this study presents a quick and facile solution-processed method for fabricating the PEDOT:PSS transparent conductive thin film (without a post-treatment process) while still achieving the requirements for a transparent conductive electrode (TCE). A parametric study was conducted by adding 5 wt% to 80 wt% of benzene sulfonic acid (BA) to PEDOT:PSS during the formulation stage before being dried at elevated temperatures from 80 °C to 200 °C. The optimum sheet resistance and transmittance value could be achieved for a thin film fabricated from PEDOT:PSS added with 40 wt% of BA, and dried at 120 °C. The sheet resistance and transmittance values are 80 Ω/sq and 93.6%, respectively. The generated figure of merit (FOM) value is 70.1, indicating an improvement of almost five times compared to the FOM value of 14.6 generated using the conventional method, requiring a post-treatment process.

ABSTRAK: PEDOT:PSS adalah bahan polimer organik yang mengkonduksi arus dan dikaji secara meluas bagi digunakan sebagai elektrod konduktif telus. Kaedah konvensional untuk menghasilkan filem nipis PEDOT:PSS melibatkan proses pasca rawatan iaitu dengan mencelupkan filem nipis PEDOT:PSS ke dalam asid pekat bertoksik bagi meningkatkan konduksi filem tersebut. Tanpa proses pasca rawatan ini dapat mengurangkan penghasilan sisa lebihan seperti asid pekat bertoksik atau pelarut buangan, memendekkan masa fabrikasi sebanyak separuh. Oleh itu, kajian ini menghasilkan kaedah proses-penyelesaian yang cepat dan mudah bagi fabrikasi filem nipis PEDOT:PSS (tanpa proses pasca rawatan) disamping masih mencapai keperluan sebagai elektrod konduktif telus (TCE). Kajian parametrik telah dijalankan dengan menambah 5 wt% hingga 80 wt% asid sulfonik benzena (BA) ke dalam PEDOT:PSS pada peringkat percampuran kimia sebelum dikeringkan pada kenaikan suhu secara berperingkat dari 80 °C sehingga 200 °C. Nilai optimum bagi rintangan lapisan dan nilai ketelusan bagi filem nipis PEDOT:PSS yang difabrikasi dapat dicapai melalui penambahan sebanyak 40 wt% BA dan dikeringkan pada suhu 120 °C. Rintangan lapisan dan nilai ketelusan telah dicapai sebanyak 80 Ω/sq dan 93.6%, masing-masing. Nilai gambaran merit (FOM) yang terhasil adalah 70.1, menunjukkan peningkatan hampir lima kali ganda berbanding nilai FOM 14.6 yang terhasil menggunakan kaedah konvensional yang memerlukan proses pasca-rawatan.

KEYWORDS: *PEDOT:PSS; thin film; sheet resistance; transmittance; figure of merit*

1. INTRODUCTION

The need for optoelectronic devices in the 21st century has increased daily. The optoelectronic devices developed nowadays, for example; solar cells [1], light-emitting diodes [2], liquid crystal displays [3], touch screens [4], and photodetectors [5], consist of one essential and common component, namely the transparent conductive electrode (TCE) or the transparent conductive thin film. TCE plays a role in electrical contact in the devices, and at the same time, it needs to allow light to pass through it. This type of electrode must provide low sheet resistance and high transmittance level. Indium tin oxide (ITO) has been the most applied TCE in the last decade. However, ITO has several serious drawbacks. For instance, ITO can be depleted as it is a rare material [6]. The cost of fabricating ITO into TCE is rapidly increasing due to shortage and complex production processes [7]. In addition, ITO is also brittle [8] and susceptible to corrosion [9].

Recently, poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) conducting polymer has emerged as a popular and widely studied organic material for replacing conventional ITO. PEDOT:PSS consists of “PEDOT” grains that are hydrophobic and conducting. The “PEDOT” grains are encapsulated by “PSS” shells that are hydrophilic and insulating [10, 11]. PEDOT:PSS has desirable advantages, such as its solution-processed method, which could support low-cost and large-scale fabrication [6]. Other than that, PEDOT:PSS also offers high flexibility [12] and high transparency in the visible light range [6]. Despite all the attractive factors, it cannot be denied that the limitation of PEDOT:PSS organic material remains its low electrical conductivity below 350 S cm^{-1} [10]. Many studies [13,14] focused on removing the insulating “PSS,” commonly through an acid post-treatment, to overcome such deficiency. Generally, the post-treatment process often involves dropping, rinsing, dipping, soaking, or fumigating the fabricated PEDOT:PSS thin film with various acids or solvents (sulphuric acid, H_2SO_4 often gives the best results) [15-17]. Using these various acids and solvents in the post-treatment process to increase the conductivity of the PEDOT:PSS thin film is referred to as the "conventional method" in the present work.

The conventional method discussed above has the drawback of high preparation cost due to the large consumption of volume in strong saturated acids [18]. The harsh acidic post-treatment process would lead to a large amount of strong acid waste after the process and also cause acid residuals to be left on the PEDOT:PSS film surfaces. The residuals could cause corrosion to any attached materials on the PEDOT:PSS surfaces or the underlying substrate. Besides that, the conventional method also prolongs the fabrication processing time because additional steps such as washing, rinsing, and drying must be repeated when each acid treatment is done. Thus, an alternative method is required to improve the conductivity of the PEDOT:PSS thin film by replacing this conventional method. Therefore, this paper proposes a quick and facile solution-processed method (without any post-treatment) to fabricate PEDOT:PSS thin film by adding diluted benzene sulfonic acid (BA). It is expected that a PEDOT:PSS thin film with desirable conductivity and transmittance can be fabricated by introducing an optimised volume of BA into the PEDOT:PSS solution.

2. EXPERIMENTAL APPROACH

2.1 Materials Preparation

For the present study, PEDOT:PSS solution (Heraeus Clevios^{TM} PH1000 with solid content 1.0-1.3 wt% in water and PEDOT:PSS ratio of 1:2.5) was procured from Ossila Ltd. isopropyl alcohol (IPA), benzene sulfonic acid (BA), sulfuric acid (H_2SO_4), ethylene glycol (EG), dimethyl sulfoxide (DMSO) were procured from Sigma-Aldrich. Graphene nanopowder (1.6 nm flakes) was procured from Graphene Supermarket. The as-received PEDOT:PSS solution (PH1000) was diluted by adding IPA to improve the solution's wettability [19] on a quartz substrate. Then, the solution was filtered through a 0.45 µm syringe filter to remove large-sized particles.

2.2 Fabrication of PEDOT:PSS Thin Film

In Fig. 1, process A presents the essential fabricating process involving the chemical formulation of the PEDOT:PSS solution, followed by spin-coating the formulated PEDOT:PSS solution onto the desired substrate before finally drying the PEDOT:PSS thin film. Process B is the post-treatment process, where the dried PEDOT:PSS thin film is dipped into an acid (e.g., sulfuric acid, H_2SO_4). After being immersed for a set time, the sample is rinsed with alcohol or deionised water to wash away the dipping acids before another drying process. The conventional method involves both processes A and B. The present study attempts to fabricate the PEDOT:PSS thin film with only process A and without involving any process B, as given in Fig. 1.

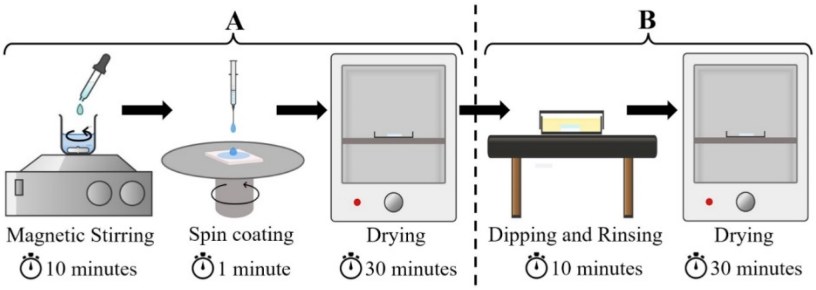

Fig. 1: Conventional method used to fabricate and improve the conductivity of PEDOT:PSS thin film.

Firstly, the IPA-diluted PEDOT:PSS solution was added with BA from 5 wt% to 80 wt%. BA is considered because of its conjugated structure, which can establish π–π interactions between benzene rings, making intermolecular charge transfer easier, thus, enhancing electrical conductivity [20]. Then, the PEDOT:PSS and BA mixture was spin-coated on a pre-cleaned quartz substrate with spin speed and time fixed at 1500 rpm for 50 s, followed by 3000 rpm for a further 10 s. After spin coating, the thin film sample was dried in an oven for 30 mins. The present study also investigated the drying temperature effect (between 80 °C and 200 °C). Note that the drying temperature has been reported to influence the conductivity of fabricated PEDOT:PSS thin film [21]. The drying temperature for PEDOT:PSS thin film is typically not recommended to be over 200 °C because this is the temperature where the chains' degradation and the deterioration of the crystallinity PEDOT:PSS film start to occur [22]. For comparative study, the samples with the post-treatment process were immediately immersed in a saturated H_2SO_4 for 10 mins after being taken out from the oven. Subsequently, the sample was rinsed with IPA and deionised water to remove the excess acid on the surface before being dried in the oven at the same drying temperature and time.

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2.3 Characterisation

The PEDOT:PSS thin film fabricated was characterised for sheet resistance (R_s) and transmittance (T). The relationship between these two properties is represented by the following equation [22, 23]:

$$T = \left(1 + \frac{Z_o}{2R_s}\frac{\sigma_{op}}{\sigma_{dc}}\right)^{-2} \quad (1)$$

where Z_o is the impedance of free space (377 ohm), and σ_{op}, σ_{dc} indicate the optical and dc conductivities, respectively. The term σ_{dc}/σ_{op} refers to the figure of merit (FOM) and acts as an indicator to quantify the quality of thin film sample as TCE. The sheet resistance and transmittance of the PEDOT:PSS thin film were measured using a four-point probe (DMR-1C square resistivity tester, Nanjing Damien Instrument Co., Ltd.) and ultraviolet-visible light spectrometer (model PC2000, Shanghai Wenyi Optoelectronics Technology Co., Ltd.), respectively. The transmittance values were measured at the wavelength of 550 nm, excluding the absorption of the quartz substrate. The thickness of the thin film layer was measured by Atomic force microscopy (AFM, Bruker JPK Instruments) in non-contact mode. Lastly, the water contact angle of the PEDOT:PSS solution on quartz sample was measured using an in house built goniometer, operated following ASTM D7334.

3. RESULTS AND DISCUSSIONS

3.1 IPA-diluted PH1000 for Improving Wettability

The wettability of PEDOT:PSS solution as a function of IPA volumes percentage is shown in Figure 2. It is observed that the solutions' wettability on a quartz substrate increased when the IPA volume percentage increased from 0% to 50%. The contact angle measured for 0%, 12.5%, 25%, and 50% of IPA diluted-PH1000 on quartz substrate were 68.9°, 56°, 43°, and 28°, respectively. A reduced contact angle value indicates a better wetting of solutions on the surface.

The PEDOT:PSS solutions were then spin-coated onto pre-cleaned quartz substrates. Figure 3 shows the effect of the volume percentage of IPA dilution on the uniformity of the spin coating process. Dilution of PH1000 with 25% IPA or more can achieve a good uniform spin coat of PEDOT:PSS thin film. Even though a more uniform spin-coated film can be achieved with better wettability, the resistance of 50% IPA-diluted PH1000 thin film was measured at 745.8 kΩ, which is higher than the one measured for 25% IPA-diluted PH1000 thin film at 645.0 kΩ. Thus, a 25% IPA-diluted PH1000 solution was adopted for the subsequent analysis and is referred to as the pristine PEDOT:PSS.

3.2 Pristine PEDOT:PSS + Benzene Sulfonic Acid (BA)

The R_s and T values for PEDOT:PSS solutions added with different concentrations of BA (from 5 wt% to 80 wt%) are given in Fig. 4(a). The R_s values at 5 wt% and 10 wt% BA concentration are not plotted in Fig. 4(a) because of their excessively high values (over 10,000 Ω/sq). With such high R_s values and T of around 88%, the figure of merit (FOM), calculated using equation (1), is approximately 0.3, which is considered poor (near zero). After adding at least 20 wt% of BA, the R_s value is shown to drastically reduce to 120 Ω/sq with a T of 92.7%. The R_s is the lowest when added with 40 wt% of BA (80 Ω/sq), while T is the highest when added with 40 wt% of BA (93.6%). Figure 4(b) plots the FOM versus BA concentration, indicating that 40 wt% of BA concentration results in the highest FOM value of 70.1.

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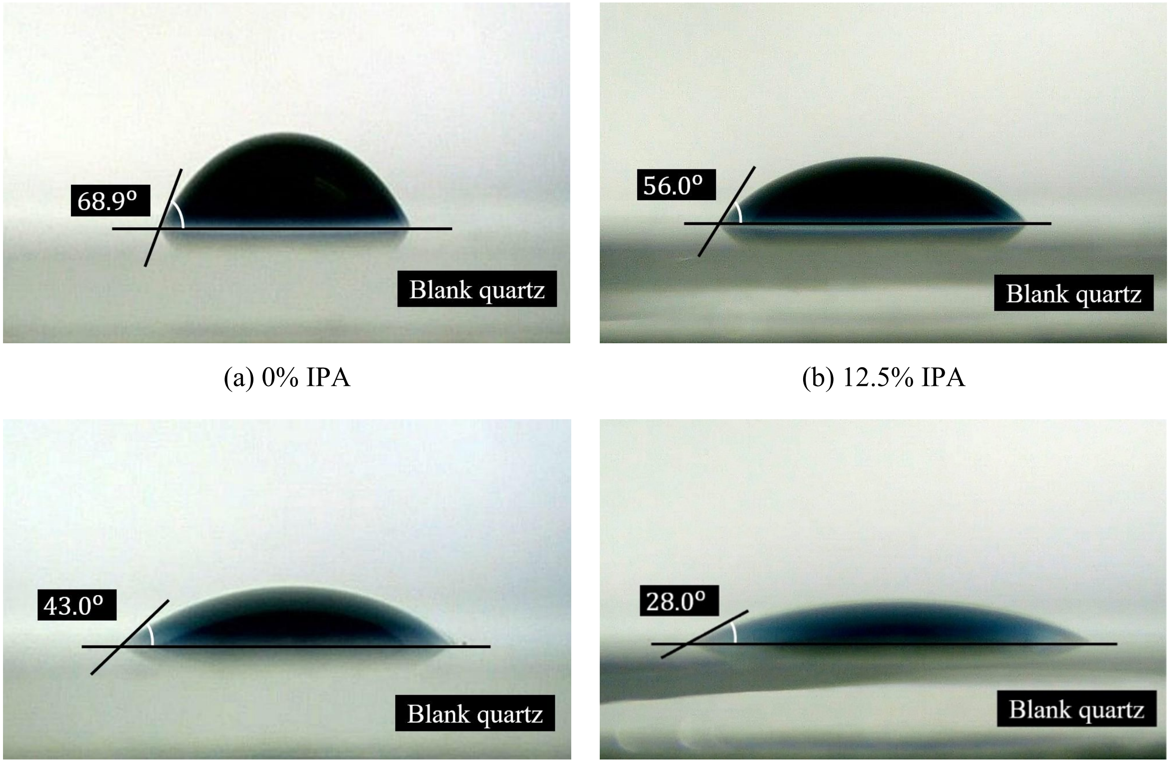
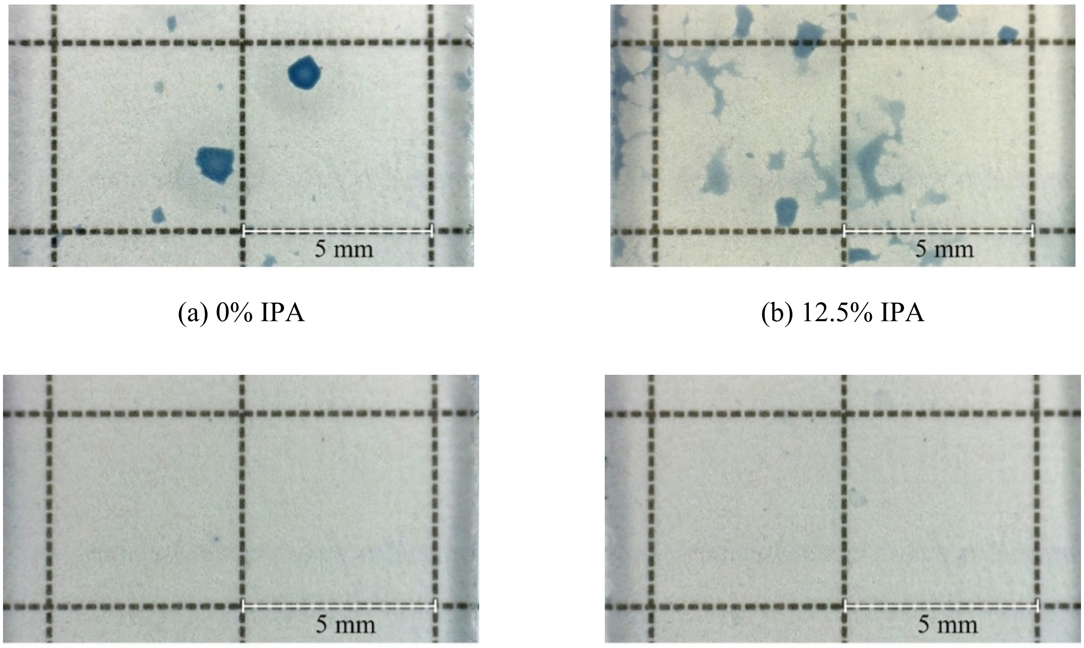


(a) 0% IPA (b) 12.5% IPA

(c) 25% IPA (d) 50% IPA

Fig. 2: Wettability of PEDOT:PSS solutions on quartz substrate with the increase of IPA volume percentage.



(a) 0% IPA (b) 12.5% IPA

(c) 25% IPA (d) 50% IPA

Fig. 3: Effect of volume percentage of IPA dilution towards the uniformity of spin coating.

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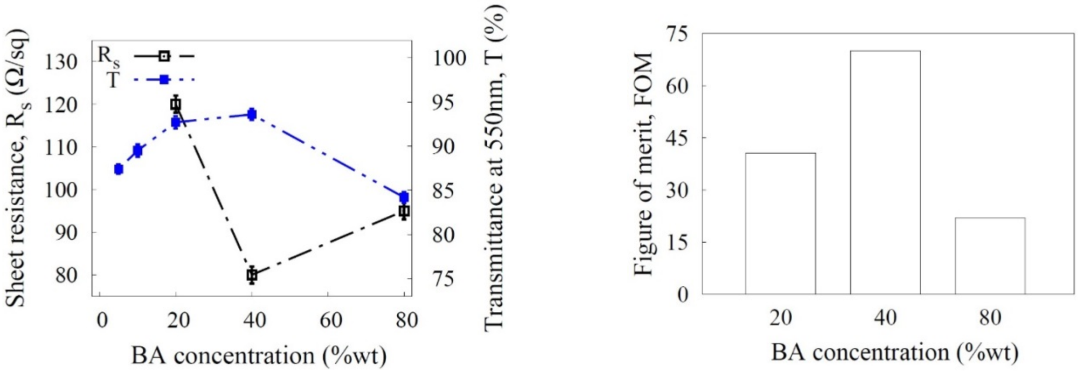
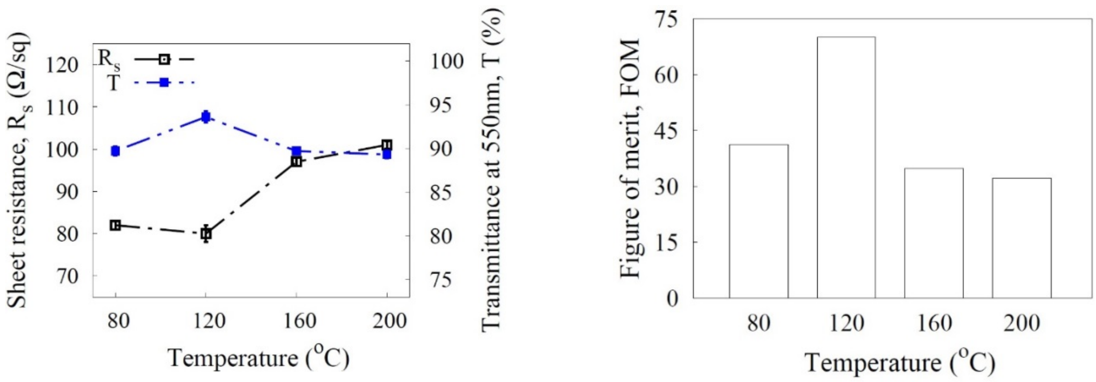


(a) R_s, T (b) FOM

Fig. 4: Effect of benzene sulfonic acid (BA) concentration towards the R_s, T and FOM of PEDOT:PSS transparent conductive thin film dried at 120 °C.

The second fabricating condition tested was the drying temperature. The PEDOT:PSS thin film fabricated with 40 wt% BA was dried under temperatures varying from 80 °C to 200 °C. From Fig. 5(a), it is observed that the lowest R_s and highest T remain at the drying temperature of 120 °C. The PEDOT:PSS thin film drying at temperatures other than 120 °C showed an increase in R_s and a decrease in T. This is related to the water, BA, and IPA (solvents) that exist in the PEDOT:PSS solutions. If such solutions dried at excessively high temperatures (160 °C and 200 °C), the solvents would evaporate before the PEDOT grains could form into a crystal. However, with a much lower drying temperature (80 °C), the solvents will remain between the PEDOT grains molecules and hinder the crystal structure formation [24]. Thus, 120 °C drying temperature is observed as the optimum drying temperature.

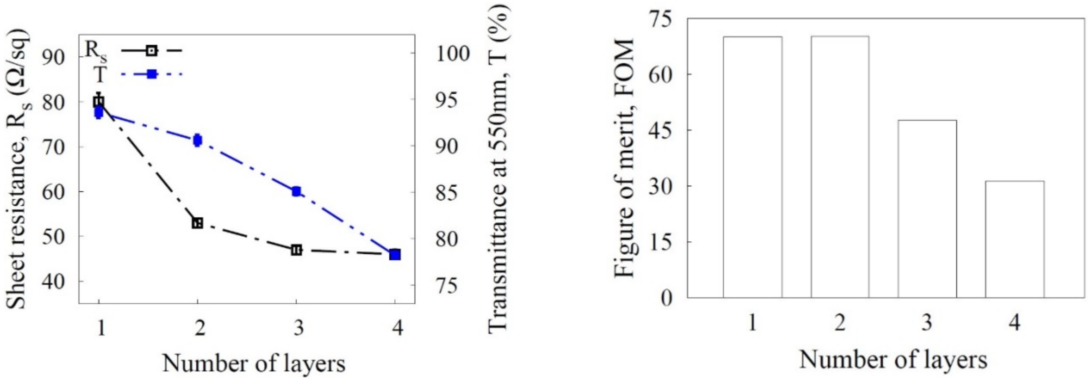


(a) R_s, T (b) FOM

Fig. 5: Effect of drying temperatures towards the R_s, T and FOM of PEDOT:PSS transparent conductive thin film.

In addition, numerous studies [6] have focused on enhancing the electrical conductivity of PEDOT:PSS thin film through layer-by-layer structure coating. Thus, the present study also investigated the effect of the number of coating layers on the FOM value of PEDOT:PSS thin film. Using the optimised fabricating conditions (40 wt% BA and 120 °C drying temperature), the PEDOT:PSS thin film was fabricated (repeated spin coating after each drying process) to achieve one to four layers of film. As shown in Fig. 6(a), although the R_s

and T values of one layer (1 L) and two layers (2 L) are different, it was determined that both cases resulted in similar FOM values of 70.1 and 70.3, as given in Fig. 6(b). However, after more than two layers of spin coating, the third (3 L) and fourth (4 L) layers of spin coating reduced the FOM value to 47.7 and 31.3, respectively.



(a) R_s, T (b) FOM

Fig. 6: Effect of number of coating layers towards the R_s, T and FOM of PEDOT:PSS transparent conductive thin film.

3.3 Comparative Study with Different Solution-processed Methods

Four other solution-processed methods were selected for comparison with the method proposed in the present study (named as method-4). It is to note that methods 1 and 2 were selected as baseline comparisons. The methods are summarised below:

- Method-1: process A (pristine PEDOT:PSS) + process B (dipping saturated H_2SO_4)
- Method-2: process A (pristine PEDOT:PSS) + process B (dipping saturated BA)
- Method-3: process A (pristine PEDOT:PSS added with 40 wt% H_2SO_4)
- Method-4: process A (pristine PEDOT:PSS added with 40 wt% BA)
- Method-5: process A (pristine PEDOT:PSS added with 40 wt% BA) + process B (dipping saturated H_2SO_4)

Figure 7 shows the (a) R_s, (b) T, and (c) FOM values of PEDOT:PSS transparent conductive thin film fabricated with different solution-processed methods. Method-1 and method-5 fabricated PEDOT:PSS thin films resulted in higher R_s of 215 and 176 Ω/sq, respectively, while method-4 resulted in the lowest R_s value of 80 Ω/sq. It is found that PEDOT:PSS thin film undergoing the conventional post-treatment process (process B) tends to generate lower levels of transmittance. For example, method-4 fabricated PEDOT:PSS thin film resulted in a transmittance of 93.6%. However, when subjected to the addition process B (method-5), the T value reduced 8.9% to 84.7%. Similarly, method-1 and method-2, subjected to process B, also resulted in a lower T value than method-4. Hence, through the calculation from equation (1), only the method-4 fabricated PEDOT:PSS thin film gives a FOM value of 70.1, which is over 35.0 (minimum industrial standard of TCE [22, 23]). Note that method-3 is not shown in Fig. 7 because the PEDOT:PSS solution was solidified during the chemical formulation process and could not get a uniform spin coating.

Method-1 (baseline), method-4 (1 L), and method-4 (2 L) PEDOT:PSS thin film samples' thicknesses were measured using atomic force microscopy (AFM). Figure 8 shows the film thickness of samples from method-1, method-4 (1 L), and method-4 (2 L), measured at 43 nm, 85 nm, and 154 nm, respectively. With the thickness (nm) and (Ω/sq) data

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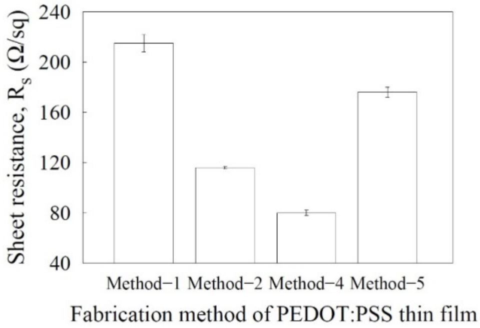
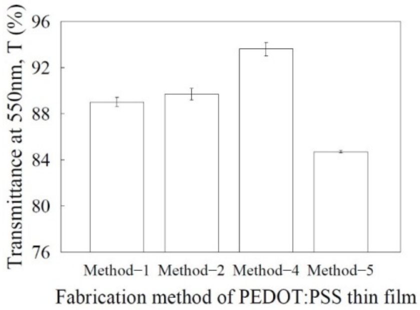
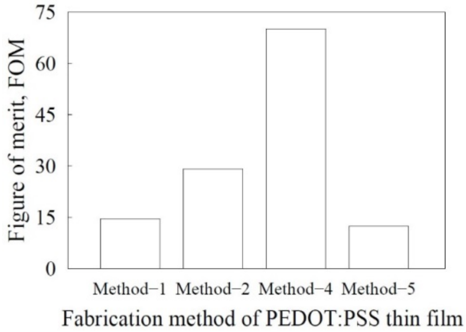
available, the conductivity of the thin film samples can then be determined. Thus, the conductivity of method-1 thin film is approximately 1081.7 Scm^{-1}, method-4 (1 L) thin film is 1470.6 Scm^{-1}, and method-4 (2 L) is 1225.2 Scm^{-1}.



(a) R_s



(b) T



(c) FOM

Fig. 7: Effect of different solution-processed methods towards (a) R_s, (b) T and (c) FOM of PEDOT:PSS transparent conductive thin film.

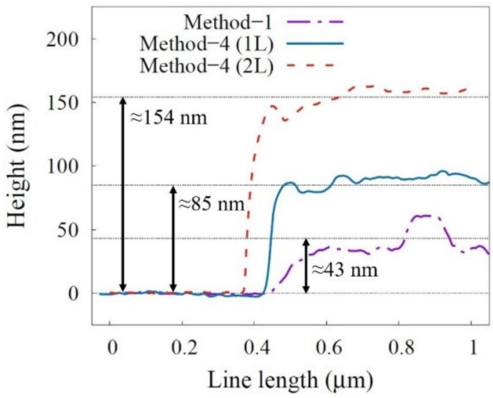


Fig. 8: PEDOT:PSS film thickness from method-1, method-4 (1 L) and method-4 (2 L).

The results obtained from this study were also compared to the recent papers published in the same field. Table 1 compares the properties of PEDOT:PSS transparent conductive film fabricated with or without the post-treatment process. The present study's conventional method (method-1) yielded a FOM value of 14.6, which is comparable to the FOM value of 17.5 obtained by Bießmann et al. [15]. The slightly lower FOM value produced in this study using the same processing procedure as Bießmann et al. [15] can be attributed to the IPA dilution in the PH1000 solution adopted in this research. Wen et al. [18] demonstrated that

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mixing PH1000 with dimethyl sulfoxide (DMSO) solvent and undergoing a post-treatment process with trifluoromethanesulfonic acid (TA) can produce PEDOT:PSS film with a much higher FOM value, which is 116.5. Nevertheless, the result of this study still demonstrates a reasonably high FOM value (70.1) compared to all other studies in the literature in which the fabrication method is completed without any post-treatment process.

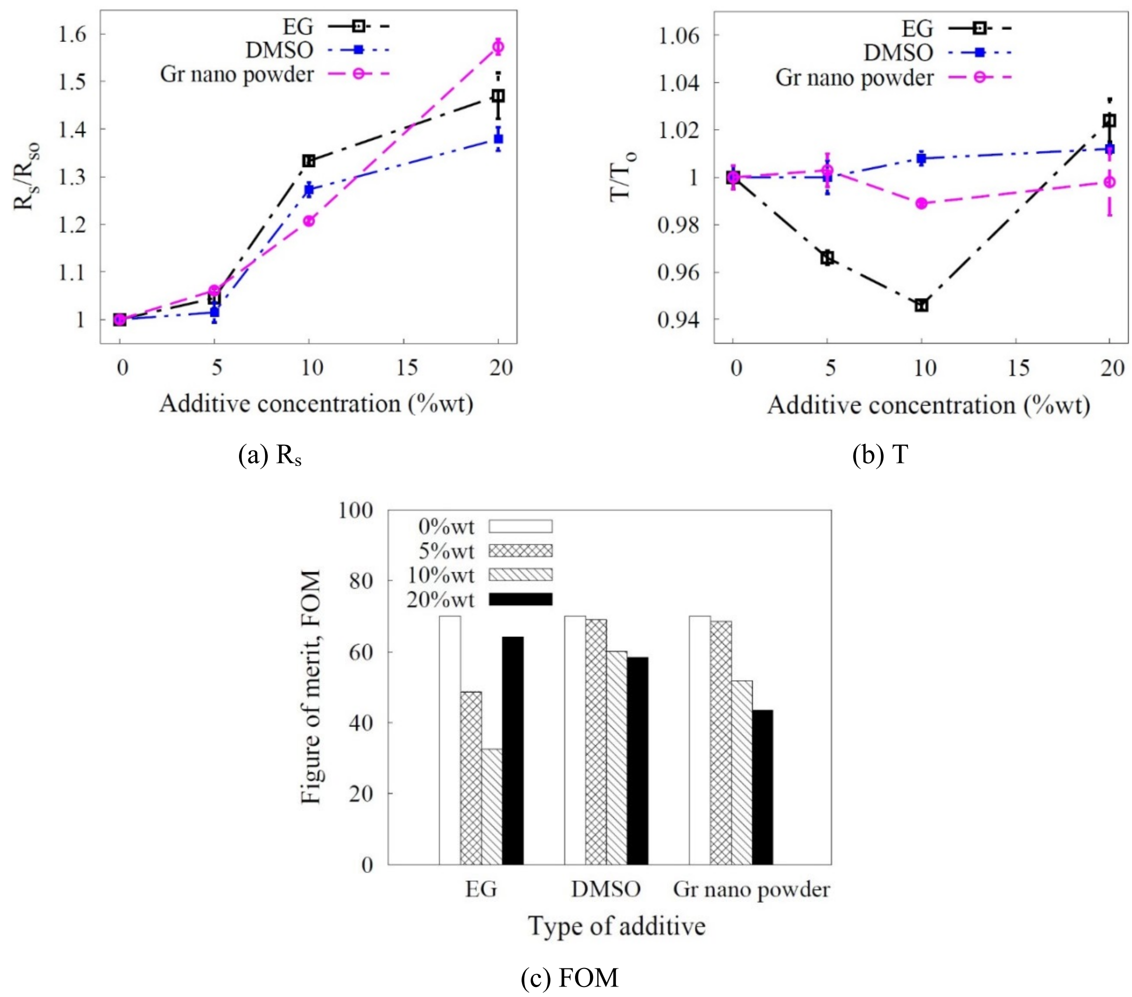
Table 1: A comparison of properties of PEDOT:PSS transparent conductive thin film fabricated.

Type of PEDOT :PSS	Additional mixing materials	W/Wo post-treatment (solvent/acid)	t [nm]	C [S cm^{-1}]	T at 550nm [%]	R_s [Ωsq^{-1}]	FOM	Ref
PH 1000	none	Wo	103.0	1.1	-	88261.3	-	[17]
		H_2SO_4	92.0	1167.0	-	93.1	-	
PH 500	none	Wo	43.2	0.2	-	1500000.0	-	[16]
		H_2SO_4	12.9	596.3	-	1300.0	-	
	MCNT	Wo	150.0	5.1	-	13000.0	-	
		H_2SO_4	150.0	78.4	-	850.0	-	
PH 1000	Zonyl and EG	DMSO	-	-	93.7	124.5	45.6	[25]
		MA	-	-	93.5	111.0	50.0	
		DMSO and MA	-	-	94.9	110.0	64.4	
PH 1000	none	Wo	165.0	7.0	90.0	8658.0	0.4	[15]
		EG	138.0	1128.0	86.0	64.2	37.5	
		HCL	169.0	392.0	84.6	150.9	14.3	
		FA	108.0	1289.0	84.6	71.8	30.1	
		HNO_3	66.0	2099.0	85.7	72.2	32.6	
		H_2SO_4	80.0	2938.0	88.5	42.5	70.3	
		H_2SO_4 (washed)	65.5	1293.0	84.0	118.1	17.5	
PH 1000	SCNT, SDBS and TX100	Wo	-	-	88.0	400.0	7.1	[26]
		Methanol	-	-	89.0	290.0	10.8	
		Methanol and HNO_3	-	-	90.1	100.3	35.1	
PH 1000	DMSO	Wo	-	-	90.5	170.0	21.7	[18]
		TA	-	-	92.0	38.0	116.5	
AI-4083	Sorbitol and Martitol	Wo	173.9	0.4	87.9	150000.0	0	[21]
		FA	129.6	847.9	87.8	91.0	30.8	
PH 1000	IPA	Wo	-	-	88.0	645000.0	0	Present study
		H_2SO_4	43.0	1081.7	89.0	215.0	14.6	
	IPA and BA (1 L)	Wo	85.0	1470.6	93.6	80.0	70.1	
	IPA and BA (2 L)	Wo	154.0	1225.2	90.6	53.0	70.3	

W/Wo = with or without; t = thickness; C = conductivity; T = transmittance; R_s = sheet resistance; FOM = Figure of Merit; EG = ethylene glycol; DMSO = dimethyl sulfoxide; SCNT = single-walled carbon nanotubes; SDBS = sodium dodecyl benzene sulfonate; TX100 = Triton X-100; MCNT = multiwall carbon nanotubes; IPA = isopropyl alcohol; BA = benzene sulfonic acid; MA = methane sulfonic acid; TA = trifluoromethanesulfonic acid; HCl = hydrochloric acid; FA = formic acid; HNO_3 = nitric acid; H_2SO_4 = sulfuric acid

3.4 Effect of Additives

The effect of common additives, namely ethylene glycol (EG), dimethyl sulfoxide (DMSO), and graphene (Gr) nanopowder, on the FOM value for the PEDOT:PSS film produced using method-4 (1L) was further investigated. Figure 9 (a) shows that the R_s value starts to increase with an increase of wt% additives added to the chemical formulation of PEDOT:PSS solution. Figure 9 (b) shows that with an increase in wt% of EG additive, the T value drops at first and rises back. The T value drops 5.4% when added with 10 wt% of EG and rises 2.4% when added with 20 wt% EG. For DMSO additive, adding 5 wt% does not change the T value of the thin film, while adding 10 wt% and 20 wt% of DMSO increases the T value by 8.0% and 12.0%, respectively. For Gr nano powder additive, adding 5 wt% also does not change the T value, but adding 10 wt% decreases the T value of thin film by 11%. Although adding 20 wt% of Gr powder only reduces the T value by 0.2%, it still resulted in an extensive error bar of 14%. Hence, with the FOM calculated as shown in Figure 9 (c), the effect of additive to further enhance the FOM of PEDOT:PSS thin film can be concluded to be not adequate when coupled with method-4. The PEDOT:PSS with 40 wt% of BA concentration remains the best-optimised formulation in this study.



(a) R_s (b) T

(c) FOM

Fig. 9: Effect of adding different concentration of different type additives (EG, DMSO, Gr nano powder) towards the changes of (a) R_s, (b) T and (c) FOM of PEDOT:PSS transparent conductive thin film.

4. CONCLUSION

This study presents a quick and facile solution-processed method for fabricating PEDOT:PSS thin film without a post-treatment process, capable of achieving requirements to be adopted as a transparent conductive electrode (TCE). The sheet resistance, transmittance, and FOM properties are enhanced by optimising the fabricating conditions with 40 wt% of added benzene sulfonic acid BA, drying at 120 °C, and one layer of spin coating. The lowest sheet resistance and highest transmittance achieved were 80 Ω/sq and 93.6%, respectively, producing a FOM value of 70.1. The FOM value improved almost five times compared to the FOM value of 14.6 for the conventional method of fabricating PEDOT:PSS film. The conductivity of PEDOT:PSS thin film fabricated using the proposed method reached 1470.6 Scm^{-1}, improving by 36% compared to the conductivity of 1081.7 Scm^{-1} achieved using the conventional method.

The PEDOT:PSS film produced in this study delivers properties that are either on-par or better than those reported in the literature, which mostly requires a post-treatment with various acids or solvents. Thus, this study concluded that the proposed quick and facile solution-processed method could produce PEDOT:PSS thin films with much-improved sheet resistance, transmittance, and FOM properties. In future studies to validate the potential of the thin film, it will be used as TCE in fabricating photovoltaic devices, allowing for the proof of concept in adopting PEDOT:PSS as an alternative TCE to ITO.

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