



## Investigation of Optoelectronic Properties of Organic Semiconductor Tetracyanoquinodimethane Based Heterostructures

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**Abstract:** Recently, interfacial layer such as metal oxide, insulator and polymer have been used by scientists between the metal and semiconductor to increase the stability of the metal-semiconductor heterojunctions. These materials have been varied according to their usage aims. In this study, graphene nanoribbons (GNR) and 7,7,8,8 Tetracyanoquinodimethane (TCNQ, C<sub>12</sub>H<sub>4</sub>N<sub>4</sub>) layer has been used as interfacial layer between the metal and semiconductor for photodiode applications. The TCNQ layer collects and extracts more electrons in the interface of the device and is used as electron acceptor material for organic solar cells. Herein, we fabricated Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al heterojunctions by physical vapor deposition technique. *I-V* measurements has been employed under dark and various light illumination conditions to show dielectric properties of the fabricated heterojunctions. From current-voltage characteristics, we calculated the electronic parameters such as ideality factor, barrier heights, series resistances and rise times. It can be concluded from overall results that TCNQ and TCNQ:GNR layers had a major impact on quality and can be considered as quite proper materials for optoelectronic applications.

**Keywords:** Al/p-Si/TCNQ:GNR/Al, Heterostructure, Optoelectronic, Photodiode

## Organik Yarıiletken Tetrasiyanoquinodimetan Tabanlı Heteroyapıların Optoelektronik Özelliklerinin Araştırılması

**Öz:** Son yıllarda bilim insanları metal-yarıiletken heteroeklemlerinin dayanıklılığını arttırmak amacıyla metal ile yarıiletken arasında metal oksit, yalıtkan veya da polimer tabakalar eklemektedirler. Bu malzemeler amaca göre değişiklik göstermektedir. Bu çalışma kapsamında, fotodiyot uygulamaları için metal ve yarı iletken arasında ara yüzey olarak grafen nanoribbon (GNR) ve 7,7,8,8 Tetrasiyanoquinodimetan-(Tetracyanoquinodimethane TCNQ, C<sub>12</sub>H<sub>4</sub>N<sub>4</sub>) katmanı kullanılmıştır. TCNQ katmanı, cihazın arayüzünde daha fazla elektron toplar ve çıkarır ve organik güneş pillerinde elektron alıcı malzeme olarak kullanılır. Daha sonra fiziksel buhar biriktirme yöntemiyle Al/p-Si/Al, Al/p-Si/TCNQ/Al ve Al/p-Si/TCNQ:GNR/Al heteroeklemleri elde edilmiştir. Elektriksel karakterizasyon kapsamında Akım-voltaj ölçümleri hem karanlık ortamda hemde farklı aydınlatma değerlerinde gerçekleştirilmiştir. Akım-voltaj karakteristiklerinden, idealite faktörü, bariyer yüksekliği, seri direnç ve yükselme zamanı gibi elektronik parametreler hesaplanmıştır. Sonuç olarak, TCNQ ve TCNQ:GNR katmanlarının kalite üzerinde büyük bir etkisi olduğu ve optoelektronik uygulamalar için oldukça uygun malzemeler olarak kabul edilebilir.

**Anahtar Kelimeler:** Al/p-Si/TCNQ:GNR/Al, Heteroeklem, Optoelektronik, Fotodiyot

## 1. Introduction

Metal-Semiconductor (MS) contacts are called Schottky diodes after its inventor W. Schottky (Wager et al., 2008). Schottky barrier diodes (SBDs) are metal-semiconductor (MS) contacts that are extensively used in semiconductor manufacturing (Berk et al., 2021), temperature-sensing (Zeghdar et al., 2020) and solar cells applications (Ramadan & Martín-Palma, 2020). On the other hand, metal-semiconductor heterojunctions have a wide range of usages in solar cells, rectifiers, capacitors, photodetectors and transistors fabrications (Munikrishana Reddy et al., 2013).

SBDs have shown high electric field and breakdown voltages (Rouger & Maréchal, 2019). In addition, Schottky diodes have very high switching speed and temperature stability compared to p-n junction diode (Kyoung et al., 2016). Because the operation of Schottky diodes is based on carriers (Anthopoulos et al., 2006). If the interface layer thickness ( $d_i$ ) between metal and semiconductor increases above a few hundred Angstroms (Å), then these structures are termed as metal/insulator-oxide-polymer/semiconductor (MIS, MOS, MPS) diodes rather than Schottky diodes (Gökçen et al., 2012; Yücedag et al., 2014). In recent years, polymer interface materials have been used to increase the properties of

these devices (Srivastava & Chakrabarti, 2015; Tozlu & Mutlu, 2016).

There has been rising amount of researches to enhance the electrical properties of diodes by inserting organics (Erdal et al., 2021; Eroğlu et al., 2020; Meftah et al., 2020), metal oxides (Yenel et al., 2021) and nanocrystals materials (Kocyiğit et al., 2021; Koçyiğit et al., 2021; Yıldırım et al., 2020). Recently, graphene (Gr) has attracted numerous attention due to its properties in optoelectronic devices. Moreover, graphene nanoribbons (GNR) seems to be advantageous than graphene due its band gap, and applicable in diodes and transistors manufacture (Sato, 2017).

Ye et al. (2011) have reported the first synthesis of GNR doped heterojunction using as light-emitting diodes (LEDs). The obtained device can emit light with 380 nm, 523 nm and 705 nm wavelengths. Furthermore, Shamsir et al. (2021) have also modified p-n junction diode based on GNR. In another research, Erdal et al. (2019) have used GNR and MWCNT layers on *p*-Si successfully which can be applied in various industries potentially. Double gate GNR diode composed by Kargar and Lee (2009), have shown  $\sim 2 \times 10^7$  rectification ratio at 0.2 V bias voltage. Rahmani et al. (2013) have explored application of bilayer GNR doped SBDs. Tataroğlu et al. (2021) have fabricated graphene-PVP/Au/*n*-Si diode via spin-coating. Gr-PVP addition to the

obtained device have shown incredible performance which can be applied instead of the insulator layer. In another study, Wang et al. (2020) have coated graphene oxide (GO) on graphene-silicon heterojunction. GO in obtained photodetector device have exhibited great effects by suppressing the dark current and increasing the photocurrent 2.73 times and responsivity to 0.65 A/W under 633 nm illumination. Moreover, Orhan et al. (2020) have synthesized CuO–Graphene/*p*-Si device by spin-coating and investigated the impact of gamma irradiation on it. In another research by Karadaş et al. (2021) 1%, 3%, 5%, 7%, and 10% graphene have been doped in PVA then coated on *n*-Si. Among them, 7% Gr doped-PVA interlayer have demonstrated best rectifying rate, with low series resistance.

Many Schottky diodes are prepared and characterized using metals, inorganic semiconductors and organic conductive polymers. The electrical and photoelectric properties of polymeric and non-polymeric organic compounds are widely investigated. It was found that the heterostructures containing non-polymeric and polymeric organic thin films have considerable rectifying properties. Al/PVC–TCNQ:ZnO/*p*-Si device synthesized by Erdal et al. (2021) demonstrated a linear photoconductivity, and photoresponsivity of the obtained device has been improved by ZnO doping significantly. In another study,

Taşçıoğlu et al. (2017) have fabricated Au/P3HT:PCBM:F4-TCNQ/*n*-Si Schottky barrier diode (SBDs) via spin-coating and investigated electrical and dielectric properties between 10 kHz–2 MHz. Çimen et al. (2018) have investigated the concentration of TCNQ on Au/P3HT:F4-TCNQ/*n*-Si on SBDs which improves the quality of devices. Moreover, Mun et al. (2020) have successfully fabricated F4-TCNQ doped stretchable semiconductor showing high mobility and stability.

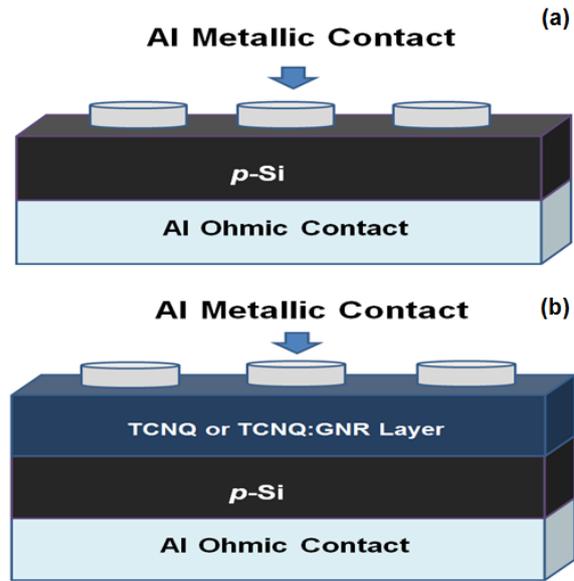
Within the scope of this study, Al/*p*-Si/Al, Al/*p*-Si/TCNQ/Al and Al/*p*-Si/TCNQ:GNR/Al heterojunctions have been fabricated successfully. The effects of TCNQ and TCNQ:GNR layers to the interlayer of the metal-semiconductor heterojunction structure have been investigated. The devices have been characterized by taking temperature-dependent *I-V* measurements for determination of dielectric constants. The aim of the project is basically to obtain the TCNQ:GNR heterojunction structure and to determine the photosensitivity properties depending on the illumination intensity.

## 2. Material and Method

The *p*-type Si wafer used as substrate was cut to 1.5 cm<sup>2</sup> pieces, then they were cleaned by H<sub>2</sub>O<sub>2</sub>:H<sub>2</sub>O:HCl, H<sub>2</sub>O<sub>2</sub>:H<sub>2</sub>O:NH<sub>4</sub>OH, isopropanol and acetone solutions in an ultrasonic bath and dried by

nitrogen subsequently. Moreover, in order to remove impurities, Si pieces were steeped into HF:H<sub>2</sub>O (1:1) solution. The wafer owned perfect crystalline orientation and  $7.5 \times 10^{16} \text{ cm}^{-3}$  carrier concentration.

Ohmic contact was created on the back surface of the Si pieces by evaporating pure Al (99.999%) at  $8 \times 10^{-6}$  torr pressure in PVD. Si substrates with 150 nm thickness of Al achieved in PVD was dealt with 450 °C temperature for 3 minutes. TCNQ and TCNQ:GNR solutions were coated on Si wafer via spin-coating. Spin-coating has been performed in 1500 rpm for 45 s using Fytronix SC-500 spin coater. After coating, doped and undoped Si substrates were put into PVD for Al evaporation via hole array mask. The hole radius was determined as  $7.85 \times 10^{-3} \text{ cm}^2$ . The hole radius was determined as  $7.85 \times 10^{-3} \text{ cm}^2$  and 150 nm of thickness. This evaporation was created in same pressure. Obtained heterostructures were analyzed via Fytronix FY-7000 Electronic Device characterization system for the  $I-V$  and  $I-T$  measurements. The measurement temperatures were changed between 50–400 K by 50 K steps. Schematic diagrams of fabricated heterojunctions are given in Figure 1.



**Figure 1.** Schematic diagrams of (a) Al/*p*-Si/Al and (b) Al/*p*-Si/TCNQ/Al or Al/*p*-Si/TCNQ:GNR/Al heterostructures.

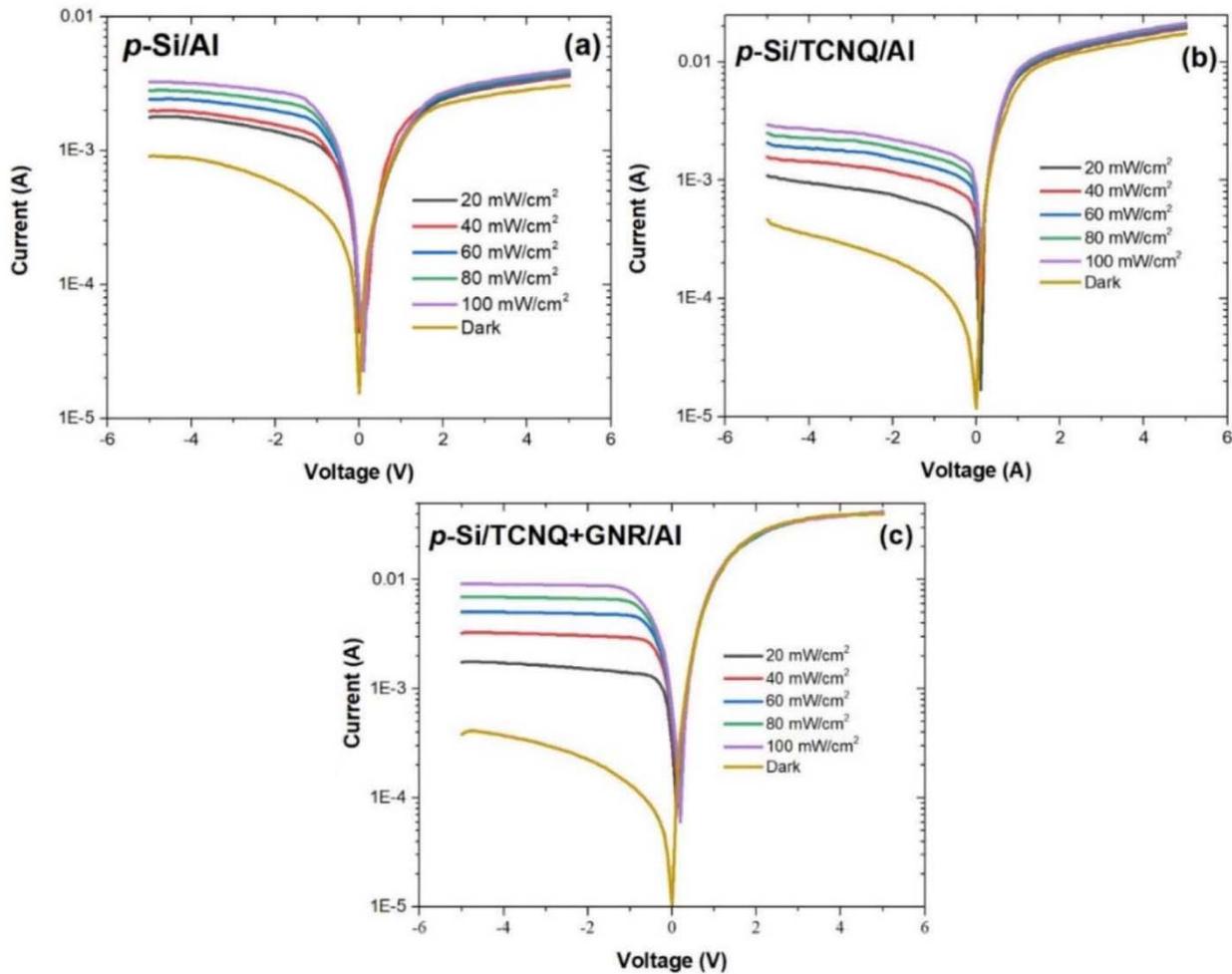
### 3. Result and Discussion

#### 3.1. Electrical characteristics of the fabricated photodiodes

$I-V$  characteristics of the Al/*p*-Si/Al, Al/*p*-Si/TCNQ/Al and Al/*p*-Si/TCNQ:GNR/Al heterostructures have been shown in Fig. 2 under various light power intensities. Al/*p*-Si/TCNQ/Al and Al/*p*-Si/TCNQ:GNR/Al devices exhibited increasing current profile in the forward bias region with increasing light power intensity. Furthermore, Al/*p*-Si/TCNQ/Al and Al/*p*-Si/TCNQ:GNR/Al heterostructures can be considered as two-lead phototransistors. In as much as, they have been exhibiting a phototransistor behavior and increasing base current with increasing light power intensity and staying constant with increasing voltage in the forward bias region. The phototransistors usually can be grown on the SiO<sub>2</sub>/Si surface

as FET structure or PNP structure as BJT on a one substrate and they are illuminated for switching (Kim et al., 2017; Kostov et al., 2013; Shao et al., 2019; Xie et al., 2020). In this study, we used normal Schottky type photodetector with *n*-type TCNQ and TCNQ:GNR interlayers. The *n*-type TCNQ and TCNQ:GNR layers can absorb the light and emit electrons to the medium

significantly. While the photodiodes or photodetectors work in the reverse biases, the phototransistors run in the forward bias region. We have both photodiode behavior in reverse biases and phototransistor behavior in the forward bias region. The threshold values of the fabricated device were calculated from these graphs and discussed broadly.



**Figure 2.**  $\ln I$ - $V$  plots of the (a)  $\text{Al}/p\text{-Si}/\text{Al}$ , (b)  $\text{Al}/p\text{-Si}/\text{TCNQ}/\text{Al}$  and (c)  $\text{Al}/p\text{-Si}/\text{TCNQ}:\text{GNR}/\text{Al}$  heterostructures.

$\ln I$ - $V$  plots are used generally to calculate ideality factor, barrier height and series resistance of the metal semiconductor devices by thermionic emission theory, Norde and Cheung methods (Kocyigit et al.,

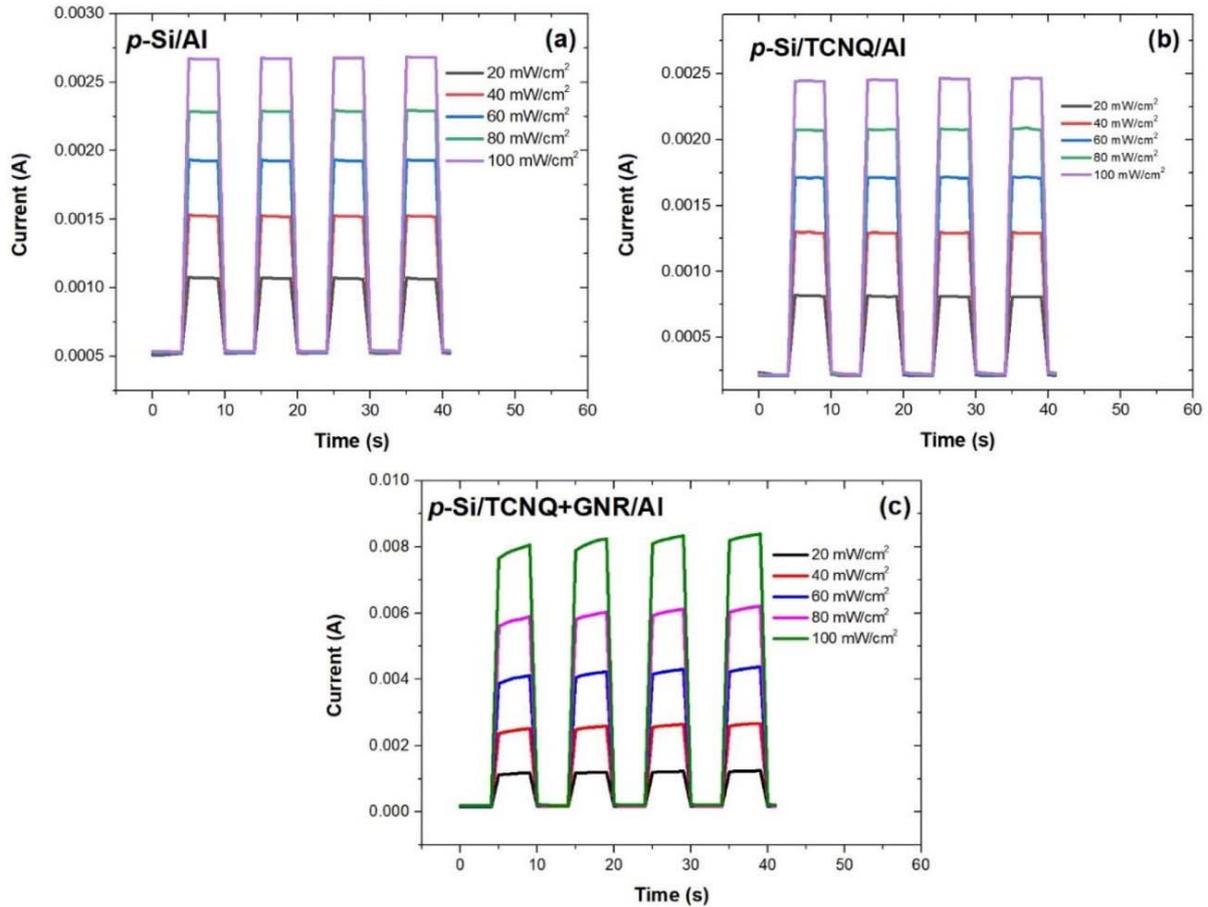
2019). According to thermionic emission theory, while the slope of the second region at the forward bias  $\ln I$ - $V$  plot provides to calculate the ideality factor, y-intercept of the plot helps calculate barrier height value

(Tataroğlu et al., 2021). Fig. 2(a), 2(b) and 2(c) show the  $\ln I-V$  plots of the Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al heterostructures under dark and 100 mW/cm<sup>2</sup> light power intensity, respectively. The changes of the current in the reverse bias regions can clearly be seen in given graphs. Obtained devices exhibited increasing photocurrent profile in the reverse bias as well as forward bias regions. This result demonstrates the photodiode behavior of the fabricated devices with phototransistor behaviors (Özmen et al., 2019). The ideality factor, saturation current and barrier height values were calculated and listed in Table 1 according to thermionic emission theory for dark condition. The ideality factor values of the Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al heterostructures were determined as 2.56, 2.07 and 2.20, and the barrier height values were calculated as 3.76 eV, 1.99 eV and 2.45 eV, respectively. The high ideality factor values at the Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al devices can be attributed to barrier

inhomogeneity and interfacial TCNQ and TCNQ:GNR layers as well as series resistance rather than more than unity (Cifci et al., 2018; Kacus et al., 2020). Current transient ( $I-t$ ) measurements of the Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al devices have been shown in Fig. 3(a), 3(b) and 3(c), respectively. The currents of the fabricated devices suddenly increased when the light is applied for all various light power intensity values, and decreased immediately when it is off. This result accentuated that the fabricated devices have good responsivity to the light illumination (İlhan et al., 2021). Moreover, the current increased almost linearly on the all devices with increasing light power intensity and maximum current was obtained for 100 mW/cm<sup>2</sup> values. The rise times of the devices were obtained as 45.08 ms, 46.12 ms and 46.55 ms for the Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al devices, respectively. These results can be indicated to good response time according to literature (Hu et al., 2021).

**Table 1.** Various diode parameters for the Al/p-Si/Al, Al/p-Si/TCNQ/Al and Al/p-Si/TCNQ:GNR/Al heterostructures.

Interface Type	$n$ (I-V)	$n$ Cheung	$\Phi_b$ (I-V) (eV)	$\Phi_b$ Cheung (eV)	$R_s$ Cheung ( $\Omega$ (H(I)))	$R_s$ Cheung ( $\Omega$ (dln(I)))
Undoped	2.56	3.76	0.68	0.56	180	203
TCNQ	2.07	1.99	0.82	0.77	2950	1345
TCNQ:GNR	2.20	2.45	0.75	0.61	2260	1012



**Figure 3.**  $I-t$  graphs of the (a) Al/ $p$ -Si/Al, (b) Al/ $p$ -Si/TCNQ/Al and (c) Al/ $p$ -Si/TCNQ:GNR/Al heterostructures.

Current transient ( $I-t$ ) measurements of the Al/ $p$ -Si/Al, Al/ $p$ -Si/TCNQ/Al and Al/ $p$ -Si/TCNQ:GNR/Al devices have been shown in Fig. 3(a), 3(b) and 3(c), respectively. The currents of the fabricated devices suddenly increased when the light is applied for all various light power intensity values, and decreased immediately when it is off. This result accentuated that the fabricated devices have good responsivity to the light illumination (İlhan et al., 2021). Moreover, the current increased almost linearly on the all devices with increasing light power intensity and maximum current was obtained for 100 mW/cm<sup>2</sup> values. The

rise times of the devices were obtained as 45.08 ms, 46.12 ms and 46.55 ms for the Al/ $p$ -Si/Al, Al/ $p$ -Si/TCNQ/Al and Al/ $p$ -Si/TCNQ:GNR/Al devices, respectively. These results can be indicated to good response time according to literature (Hu et al., 2021).

#### 4. Conclusion

TCNQ and TCNQ:GNR layers on  $p$ -Si substrate have been fabricated successfully. The electrical characterization of the devices was performed by  $I-V$  measurements under dark and various light power intensities. From current-voltage

characteristics, the electronic parameters such as ideality factors, barrier heights, series resistances and rise times were calculated. The fabricated devices exhibited considerable responsivity and specific detectivity. It can be concluded from overall results that TCNQ and TCNQ:GNR layers had a major impact on quality and can be

considered as quite proper materials for optoelectronic applications.

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