

Production and Characterization of Al/NiO:ZnO/p-Si/Al Composite Photodiodes for Solar Energy Tracking Systems

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(Geliş/Received: 10/02/2022;

Kabul/Accepted: 26/02/2022)

Abstract: In present study, NiO:ZnO thin films in molar ratios of 1:0, 0:1, 3:1, 1:1 and 1:3 were formed on p-Si layers with Aluminium (Al) bottom contact. The dynamic sol-gel spin coating method was used as coating method. The Al top contacts were deposited on thin films and Al/NiO:ZnO/p-Si/Al photodiodes were fabricated. The structural and morphological properties of the photodiodes were determined by X-ray diffraction (XRD), emission scanning electron microscopy (FE-SEM), and energy dispersive X-ray spectroscopy (EDX). The photoresponse and electrical properties of the produced photodiodes were investigated by current-voltage (I-V) and capacitance-voltage (C-V) measurements. The Al/NiO:ZnO/p-Si/Al photodiodes were successfully fabricated. It was determined that the thin films were composed of nanostructures. All photodiodes are sensitive to light. It was seen that the photosensitivity of composite photodiodes was higher than the pure photodiodes and photosensitivity decreased as the ZnO ratio increased. It was determined that the most sensitive photodiode to light was the composite photodiode with a NiO:ZnO ratio of 3:1, and the highest photosensitivity was measured as 3.12×10^3 at 100 mW/cm^2 light intensity in this photodiode. In all photodiodes, the capacitance values decreased as the frequency increased. The results show that by changing the NiO:ZnO ratio, the photoresponse and electrical parameters of the photodiodes can be controlled and the produced photodiodes can be used as a photosensor in solar tracking systems and optoelectronic applications.

Key words: Semiconductor metal oxide, Sol-gel, Composite photodiode, Nanomaterial, Solar irradiation.

Güneş Enerjisi Takip Sistemleri için Al/NiO:ZnO/p-Si/Al Kompozit Fotodiyotların Üretimi ve Karakterizasyonu

Öz: Mevcut çalışmada, 1:0, 0:1, 3:1, 1:1 ve 1:3 molar oranlarında NiO:ZnO ince filmler Alüminyum (Al) alt kontaklı p-Si tabakalar üzerinde oluşturuldu. Kaplama yöntemi olarak dinamik döndürerek kaplama metodu kullanıldı. Al üst kontaklar ince filmler üzerinde biriktirildi ve Al/NiO:ZnO/p-Si/Al fotodiyotlar üretilmiş oldu. Fotodiyotların yapısal ve morfolojik özellikleri X-ışını kırınımı (XRD), emisyon taramalı elektron mikroskobu (FE-SEM) ve enerji dağıtıcı X-ışını spektroskopisi (EDX) ile belirlendi. Üretilen fotodiyotların fototepki ve elektriksel özellikleri akım-voltaj (I-V) ve kapasite-gerilim (C-V) ölçümleri ile araştırıldı. Al/NiO:ZnO/p-Si/Al fotodiyotlar başarılı bir şekilde üretildi. İnce filmlerin nano yapılarından meydana geldiği tespit edildi. Tüm fotodiyotlar ışığa karşı duyarlıdır. Kompozit fotodiyotların ışığa karşı duyarlılıklarının saf fotodiyotlara göre daha yüksek olduğu ve ZnO oranı arttıkça azaldığı belirlendi. Işığa karşı en duyarlı olan fotodiyotun NiO:ZnO oranı 3:1 olan kompozit fotodiyot olduğu tespit edildi ve en yüksek fotosensitivite bu fotodiyotta 100 mW/cm^2 ışık şiddetinde 3.12×10^3 olarak ölçüldü. Tüm fotodiyotlarda frekans arttıkça kapasite değerleri düşmüştür. Sonuçlar, NiO:ZnO oranının değiştirilerek fotodiyotların fototepki ve elektriksel parametrelerinin kontrol edilebileceğini ve üretilen fotodiyotların güneş takip sistemlerinde ve optoelektronik uygulamalarda fotosensör olarak kullanılabileceğini göstermektedir.

Anahtar kelimeler: Yarı iletken metal oksit, Sol-jel, Kompozit fotodiyot, Nanomalzeme, Güneş ışınımı.

1. Introduction

In recent years, advanced and functional materials are needed in photovoltaic applications [1]. Solar tracking systems are one of the most important systems used in recent years to increase the gathering efficiency of solar energy [2, 3]. In these systems, photodiodes are generally preferred sensors for direct detection of light [4, 5]. The researches on metal oxide semiconductors are increasing day by day in many fields. This is because metal oxide semiconductors such as ZnO, TiO₂, SnO₂, NiO and CdO have unique physical and chemical properties. Therefore,

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these oxides have become very interesting materials in electronic and optoelectronic applications [6-8]. Among these metal oxide semiconductors, ZnO (n-type) is very interesting because of its properties such as, has a direct and wide (~3.3 eV) energy band gap, can be directed optical and electrical properties by doping, high exciton binding energy, relatively low toxicity and relatively low processing temperature. Due to these properties, it finds use in many electrical and optoelectronic applications [9, 10]. NiO is another promising p-type metal oxide semiconductor in electronic and optoelectronic devices due to its excellent optoelectronic properties, wide band gap (3.6-4.0 eV), low cost, high chemical and thermal stability [11-14]. The properties of photodiodes can be modified and improved by doping with a different element. One of the methods used to change and improve the properties of photodiodes is to obtain a new composite structure by mixing various semiconductor metal oxides in different ratios, in addition to doping [15-18]. The devices consisting of (p-n) NiO:ZnO heterojunction thin films are one of the promising alternatives for optoelectronic and photovoltaic applications [19].

ZnO and NiO thin films can be produced by methods such as (Direct Current) DC and RF (Radio frequency) sputtering, sol-gel, chemical vapour deposition (CVD), pulsed laser deposition and spray pyrolysis [20]. Among these methods, the sol-gel spin coating method has advantages such as allowing a large area to be coating, simplicity of the process, controllable thickness, low temperature and low cost [12, 19, 20].

Park et al., have performed gradual ZnO and NiO composite coatings on ITO glass using the sol-gel spin coating method. They performed the structural characterization of the thin films they produced and investigated the photodiode properties [19]. Again, Tyagi et al., first coated ZnO thin film with RF magnetron sputtering technique on ITO glass, and then formed a composite structure by coating NiO on ZnO with the same method. They investigated the structural, electrical and photoresponse properties of the photodiode they produced [21]. Demirezen et al., obtained ZnO:NiO thin film on the p-Si layer by spin coating method by mixing 2%, 10% and 20% NiO with ZnO and PCBM was coated on this layer. Thus, composite Al(PCBM/NiO ZnO)/p-Si photodiodes were produced. They investigated the electrical and photo-sensing properties of the photodiodes they produced [22]. As seen in the literature, there is not much work on the coating of NiO:ZnO on p-Si by dynamic spin coating method and investigating its electrical and photoresponse properties. Therefore, in this study, NiO:ZnO/p-Si composite photodiodes were produced to obtain photodiodes with different properties and their photodiode properties were investigated.

In this study, NiO:ZnO/p-Si composite photodiodes were fabricated by dynamic sol-gel spin coating. The structural and morphological properties of the thin films coatings were characterized by XRD, FE-SEM and EDX. The photoresponse and electrical properties were investigated by I-V and C-V analyses.

2. Experimental Details

In this study, NiO:ZnO thin films were formed on p-Si layers by dynamic sol-gel spin coating method. The (100) orientation, 600 μm thickness and 1-10 cm^2 resistance p-Si layers were first cleaned in acetone in an ultrasonic bath for 5 minutes. After this process, it was rinsed in an ultrasonic bath in deionized water at the same amount of time. The same procedures were repeated with ethanol and distilled water, and finally, p-Si plates were immersed in HF:H₂O solution mixed at a ratio of 1:10 for 30 seconds and then rinsed in deionized water in an ultrasonic bath for 5 minutes. The opaque sides of the plates dried with nitrogen gas were coated with 150 nm thick Al (99.99% purity) by Nanovak brand vacuum thermal evaporation device. Then, the plates were annealed under nitrogen gas in an oven at 570 °C for 5 minutes. To produce pure NiO thin films, 0.5 M Carlo Erba brand Nickel (II) acetate tetrahydrate ($\text{Ni}(\text{OCOCH}_3)_2 \cdot 4\text{H}_2\text{O}$) was poured into 10 ml of Chemsolute brand 2-methoxyethanol ($\text{CH}_3\text{OCH}_2\text{CH}_2\text{OH}$) and mixed with a magnetic stirrer. After all the powders were dissolved, Chemsolute brand monoethanolamine ($\text{NH}_2\text{CH}_2\text{CH}_2\text{OH}$) was added to this mixture in the same molar ratio while the mixing process was continuing, and the sol-gel was obtained by mixing together at 80 °C for 2 h. After resting for one day, this sol-gel was coated on p-Si plates cut into certain sizes at 3000 rpm for 30 seconds. The coating process was performed on the FYTRONIX spin coating device in dynamic mode. The coated samples were dried at 150 °C for 10 minutes, and after this process, they were allowed to cool at room temperature for 10 minutes. By repeating the same processes, one more layer of coating was formed. The produced thin films were annealed at 450 °C for 1 h. The coated surfaces were covered with 99.99% pure Al using a Nanovak brand vacuum thermal evaporator with a physical mask, and thus top contacts were obtained on thin films. The same procedures were repeated to produce pure ZnO photodiodes, with the only difference being that Sigma Aldrich brand Zinc acetate dihydrate ($\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$) was used instead of $\text{Ni}(\text{OCOCH}_3)_2 \cdot 4\text{H}_2\text{O}$. In composite NiO:ZnO photodiodes, on the other hand, $\text{Ni}(\text{OCOCH}_3)_2 \cdot 4\text{H}_2\text{O} / (\text{CH}_3\text{COO})_2\text{Zn} \cdot 2\text{H}_2\text{O}$ was used in molar ratios of 3:1, 1:1 and 1:3 with a total of 0.5 M. XRD analyses of thin films were performed in PANalytical Empyrean brand XRD device at 45

kV/40 mA, CuK α ($\lambda = 1.5406 \text{ \AA}$) radiation and $2\theta = 20 - 80^\circ$. Surface properties and compositions of thin films were determined with Zeiss Crossbeam 540 brand FE-SEM device and EDX. The photoresponse and electrical characterizations were performed using KEITHLEY 4200 semiconductor analysis system and FYTRONIX brand solar simulator.

3. Results and Discussion

3.1. Structural and Morphological Properties of NiO:ZnO/p-Si Thin Films

The XRD analysis results of the produced NiO:ZnO thin films are shown in Figure 1. As can be seen, intense peaks occurred at 37.26° , 43.30° , 62.92° , 75.48° and 79.44° 2θ degrees in pure NiO. These peaks are consistent with JCPDS card no. 47-1049 [23-25] and correspond to planes (111), (200), (220), (311) and (222), respectively. The peaks in (100), (002), (101), (102), (110), (103), (200), (112), (201), (004) and (202) planes in pure ZnO observed at 31.85° , 34.50° , 36.32° , 47.63° , 56.67° , 62.94° , 66.46° , 68.01° , 69.14° , 72.63° and 77.07° 2θ degrees, respectively. The peaks are in agreement with the JCPDS card no. 36-1451 and literature studies and show that the produced ZnO films are in hexagonal wurtzite structure [26-29]. The (111) and (200) planes seen in high density in pure NiO are attributed to the crystalline nature of NiO [23-25] and different peaks in pure ZnO indicate that ZnO is polycrystalline [30]. On the other hand, characteristic peaks of both oxides were found in composite thin films. Although the intensities and positions of the peaks change, they are in agreement with the XRD patterns of pure NiO and pure ZnO. The intensities of characteristic peaks of NiO and ZnO increased as their ratio in the composite increased. No impurities or peaks of a different phase were found in pure and composite thin films. This indicates that the NiO:ZnO films were successfully formed and high purity.

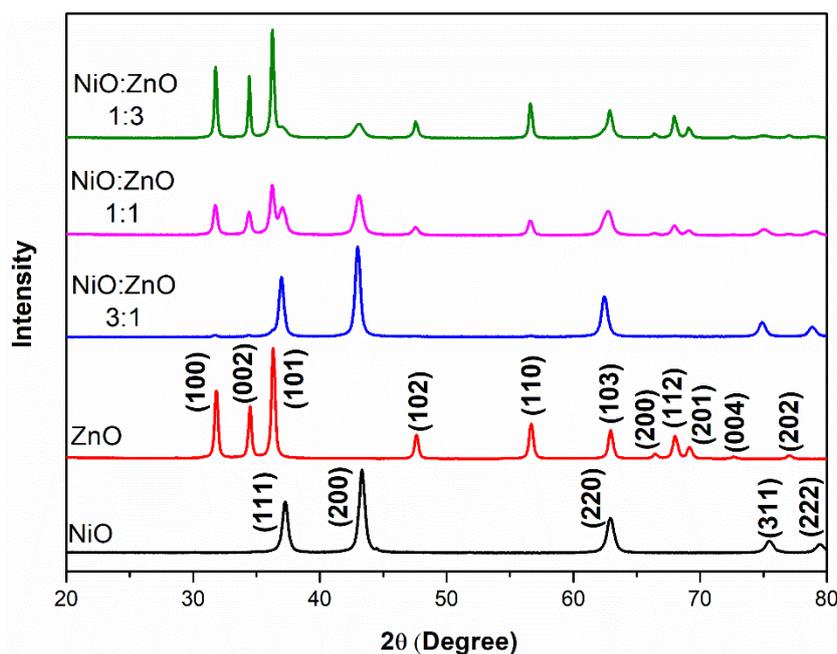


Figure 1. The XRD patterns of NiO:ZnO thin films.

Figure 2 shows FE-SEM images and EDX analysis results of NiO:ZnO thin films. All thin films were successfully coated on p-Si in a crack-free and homogeneously. Thin films are composed of nanostructures. These structures are in the form of aggregated fine-grained nanoparticles in NiO. In ZnO, on the other hand, it is composed of fiber-like structures in addition to the same structures [31]. The composite thin films are composed of aggregated fine-grained nanoparticles that are somewhat larger than pure NiO and ZnO. When the EDX analysis results were examined, it was seen that they were in agreement with the XRD analysis results and the doping ratio.

These results show that NiO:ZnO thin films were successfully formed on p-Si by dynamic sol-gel spin coating method.

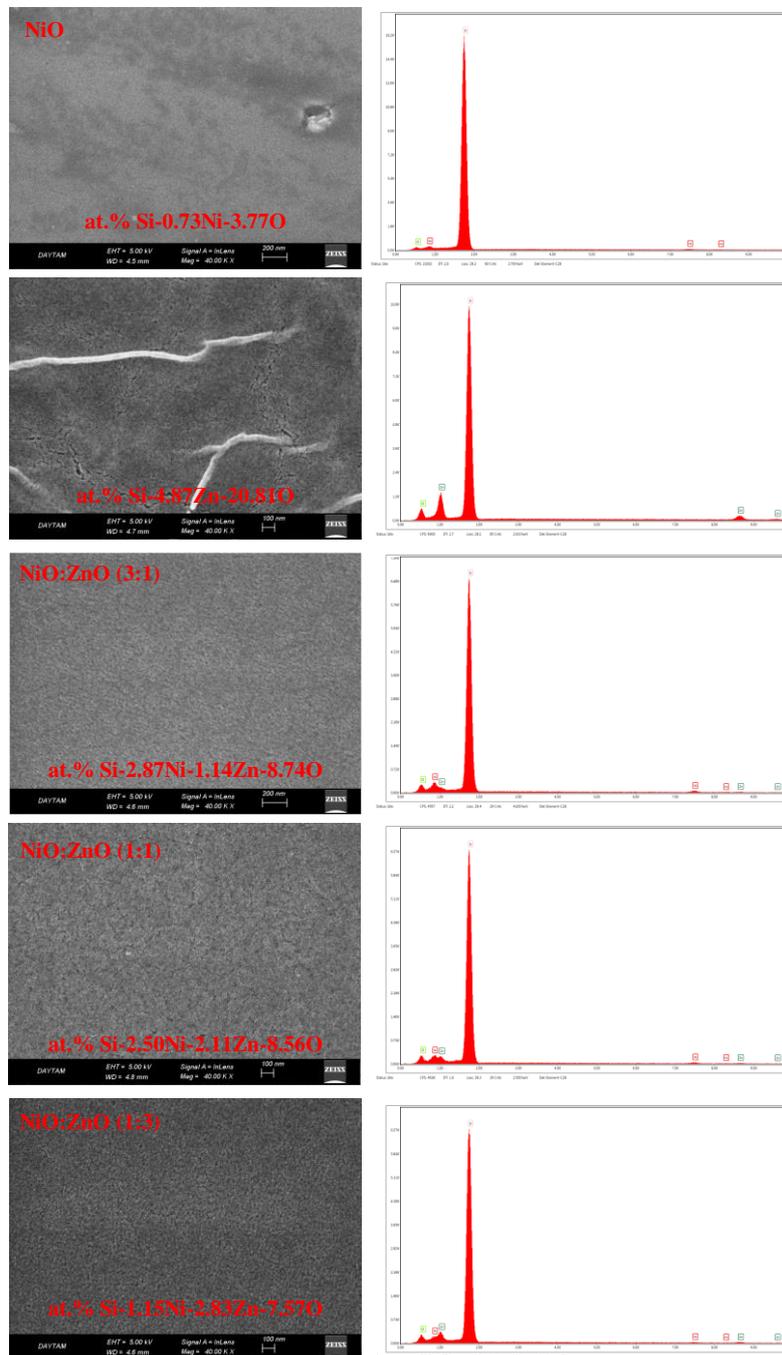


Figure 2. The FE-SEM images and EDX analysis results of NiO:ZnO thin films.

3.2. I-V Characteristics of Prepared Photodiodes

The I-V characteristics of the produced NiO:ZnO/p-Si photodiodes in the dark and at light intensities varying between 20 and 100 mW/cm² are shown in Figure 3. All samples showed rectifying properties. Except for the photodiode with a NiO:ZnO ratio of 3:1, it was observed that the current in the forward region was not affected

much by the light intensity in other photodiodes. In this sample, on the other hand, variations in current values were observed in the range of approximately 2.5-3.5 V. In the reverse region, the current was affected by the light intensity and as the light intensity increased, the current increased. This shows that the produced devices exhibit photodiode behaviour and it is thought that this is due to the spread of the electron-hole pair over a wider area in the reverse bias region [17, 27]. The results indicate that photodiodes exhibit photoconductive behaviour and the photoconductive behaviour can be controlled by changing the NiO:ZnO ratio. The photosensitivity values of the photodiodes were found at -5V using the Equation 1 given below [5].

$$PS = \frac{I_{ph} - I_{dark}}{I_{dark}} \quad (1)$$

In this equation, I_{ph} represents the photocurrent and I_{dark} represents the current in the dark. The photosensitivity values of the produced photodiodes are shown in Figure 4. The photosensitivity of photodiodes under illumination was higher than in the dark state and increased with increasing illumination intensity. The composite photodiodes are more sensitive to light than pure ones and decrease as the ZnO ratio increased. The photodiode with the highest sensitivity to light is the NiO:ZnO ratio of 3:1 and the highest photosensitivity is 3.12×10^3 at 100 mW/cm^2 light intensity in this device. The results show that photoresponse can be controlled by changing the NiO:ZnO ratio and the produced photodiodes can be used as photodiodes in solar tracking systems [1]. The photoresponsivity (P_R) of the produced photodiodes was determined using Equation 2 given below [12].

$$P_R = \frac{I_{ph} - I_{dark}}{PA} \quad (2)$$

where P is the light intensity and A is the surface area of the photodiode. The P_R values of the devices under 100 mW/cm^2 light intensity are $4.83 \times 10^{-4} \text{ A/W}$, $1.49 \times 10^{-3} \text{ A/W}$, $2.46 \times 10^{-4} \text{ A/W}$, $1.54 \times 10^{-4} \text{ A/W}$ and $2.12 \times 10^{-4} \text{ A/W}$ for 1:0, 0:1, 3:1, 1:1 and 1:3 respectively according to the NiO:ZnO ratio. All photodiodes are sensitive to light. The P_R values of photodiodes can be varied by changing the NiO:ZnO ratio.

The I-t characterizations of photodiodes have an important place in understanding their photoconductive behaviour. The I-t graphs of the produced NiO:ZnO/p-Si photodiodes at light intensities varying between 20 and 100 mW/cm^2 and at -5V are shown in Figure 5. As can be seen, the photocurrents increased as the light intensity increased. This proves that the photodiodes show photoconductive behaviour [27, 32]. When the devices were illuminated, the photocurrent suddenly increased to a certain level and fell back to its previous level after the lighting was turned off. When the illumination is first turned on, there is an increase in free charge carriers and in this case photogenerated electrons contribute to the photocurrent. When the illumination is turned off, the charge carriers are trapped in the lower levels, thus reducing the photocurrent [17, 27, 32]. The highest photoconductivity value was obtained in pure ZnO photodiode and the photoconductivity of composite photodiodes is lower. The results show that the photoconductivity properties of the photodiodes can be controlled by the NiO:ZnO ratio.

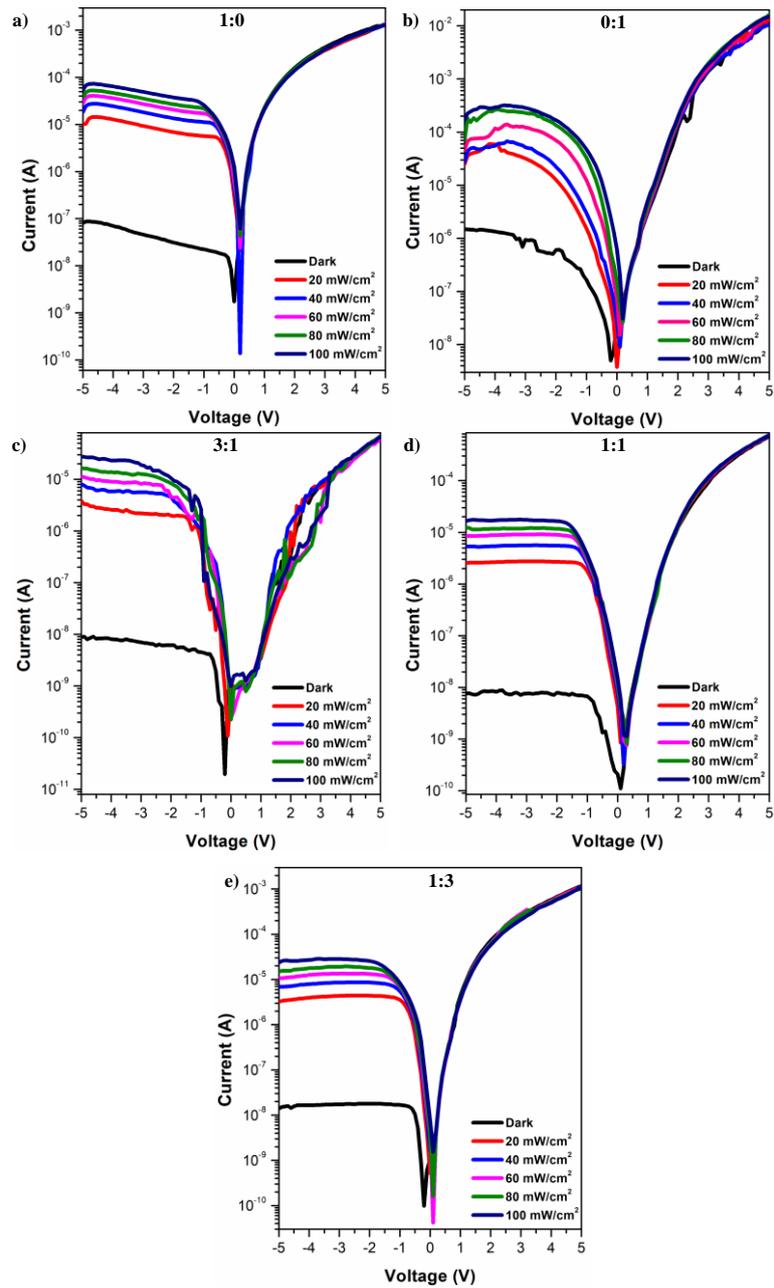


Figure 3. I-V graphs of NiO:ZnO/p-Si photodiodes under dark and various illuminations.

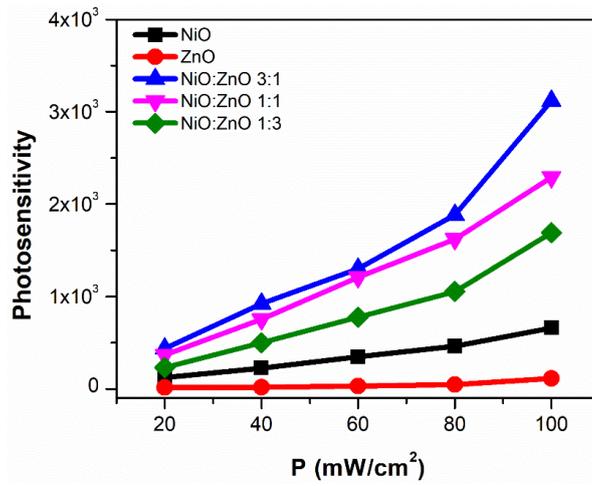


Figure 4. The photosensitivity values of NiO:ZnO/p-Si photodiodes.

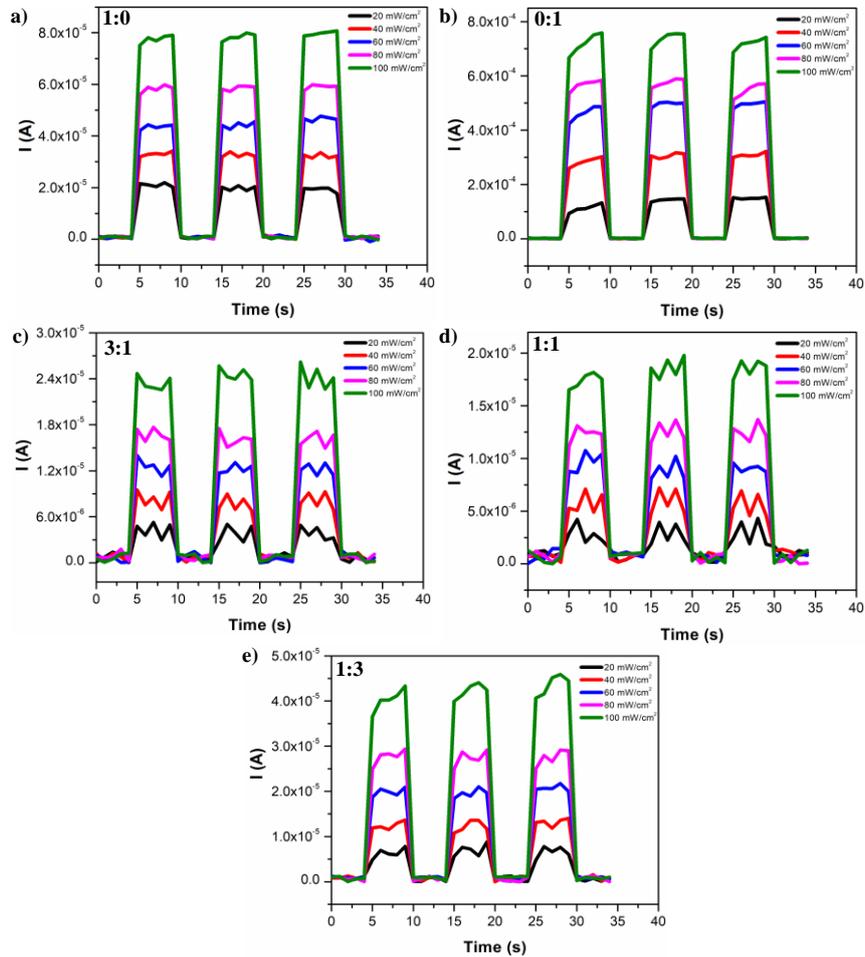


Figure 5. I-t graphs of NiO:ZnO/p-Si photodiodes.

3.3. C-V Characteristics of Prepared Photodiodes

The C-V characteristics of photodiodes are used to better characterize their electrical behaviour. The C-V characteristics of the produced photodiodes at frequencies ranging from 10 kHz to 1MHz are shown in Figure 6. As can be seen, the capacitance decreased as the frequency increased in all photodiodes. In the NiO photodiode, a peak occurred in the reverse bias region at a frequency of 10 kHz. At frequencies higher than this, the peaks occurred in the forward bias. It is thought that the peaks in the C-V graphs occur due to the presence of interfacial charge carriers and series resistance. In ZnO and composite photodiodes, while the photodiode capacitance changes with frequency in the reverse bias region, the capacitance in the forward bias region generally does not change with frequency. It is thought that the change in the negative region is due to the change of the depletion region of the device depending on the applied voltage and the response of the interface states in different frequency and electric fields [1, 33].

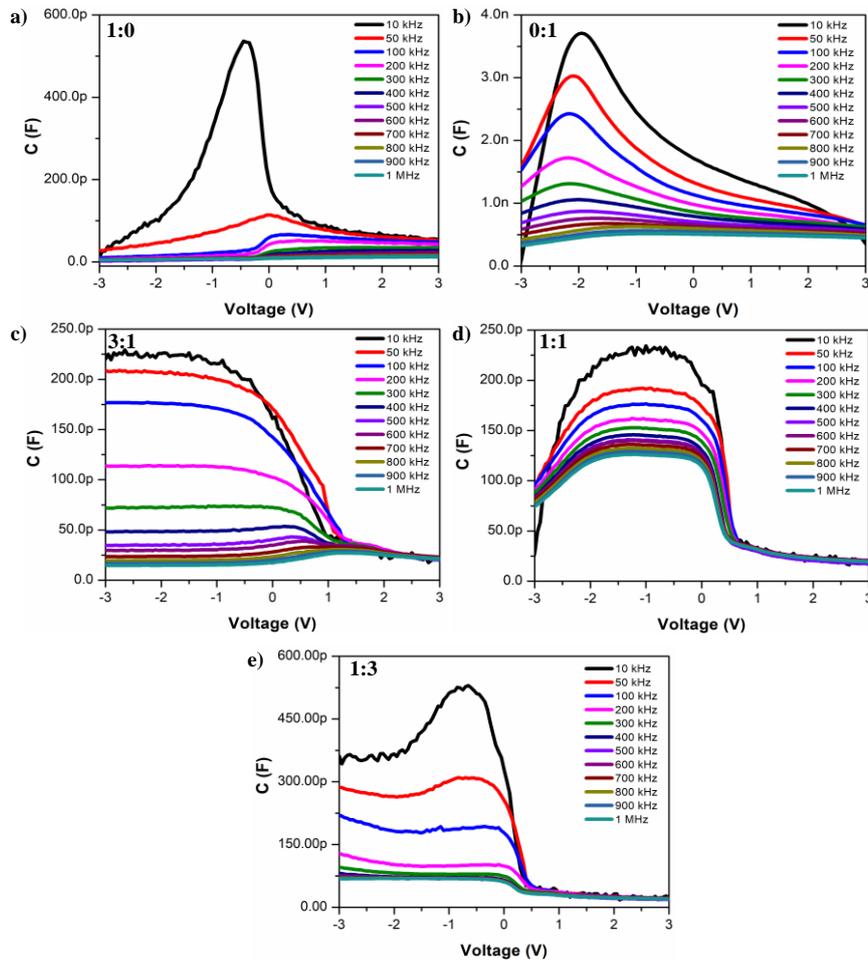


Figure 6. C-V characteristics of NiO:ZnO/p-Si photodiodes.

Series resistance (R_S)-V curves are shown in Figure 7. The R_S values decreased as the frequency increased in the accumulation and depletion region and was especially high at low frequency. In the non-ideal behaviour of diodes and deviations have an important place, and therefore R_S has a great importance. The peak values of the R_S -V curves also decreased as the frequency increased, and the peak positions changed depending on the Voltage and the NiO:ZnO ratio. It is thought that this situation is caused by the interface conditions that follow the AC signal more easily at low frequencies and the relaxation time of the charges [22].

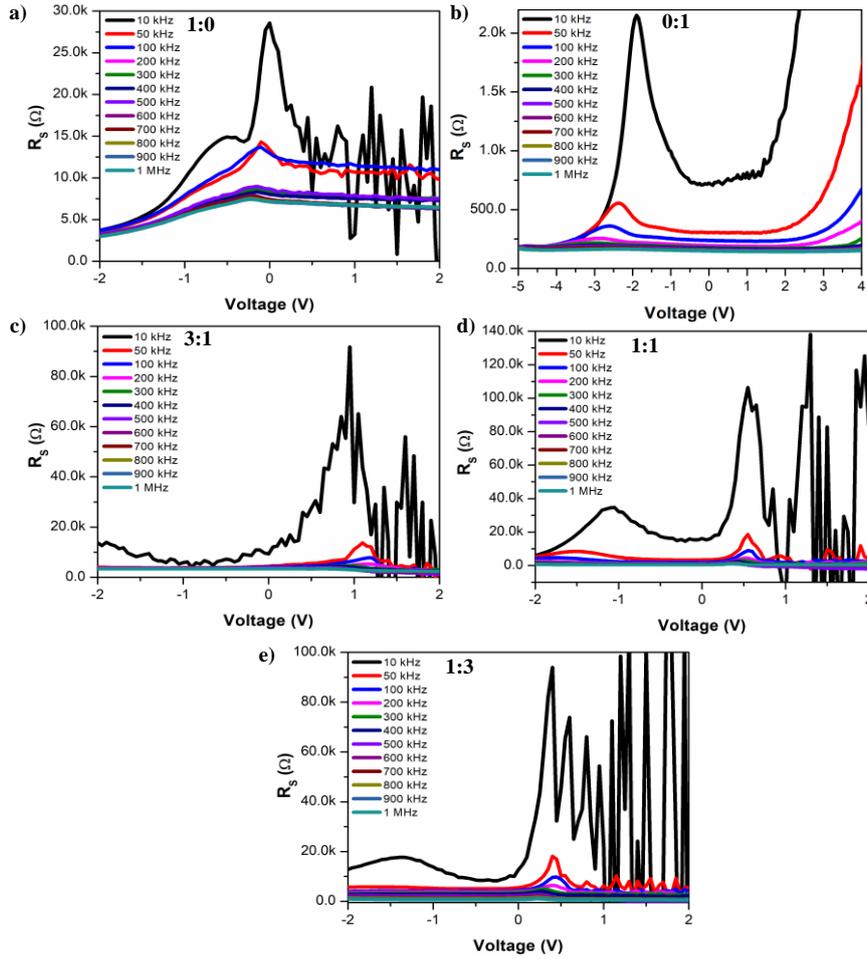


Figure 7. R_s -V characteristics of NiO:ZnO/p-Si photodiodes.

4. Conclusions

The NiO:ZnO/p-Si composite photodiodes were successfully fabricated using the dynamic sol-gel spin coating method. Characteristic peaks of NiO and ZnO were found in the composite thin films and it was observed that their intensity increased as their ratio in the composite increased. It was determined that NiO:ZnO thin films consisted of aggregated fine-grained nanoparticles. It was concluded that NiO:ZnO/p-Si composite Photodiodes are light sensitive. It has been seen that the photoresponse, photocurrent and capacitance properties of photodiodes can be controlled by light. The highest photosensitivity value was measured as 3.12×10^3 in the photodiode with a NiO:ZnO ratio of 3:1. It was seen that NiO:ZnO/p-Si composite photodiodes showed rectifying property and it was concluded that they exhibited photoconductive behaviour. It was observed that the voltage and frequency affected the electrical properties of the photodiodes. It was concluded that photodiodes with different properties can be obtained according to the NiO:ZnO ratio. The results show that, NiO:ZnO/p-Si composite photodiodes can find use in solar energy tracking systems and optoelectronic applications.

Acknowledgment

The authors thank the Firat University Research Fund (FUBAP-TEKF.21.11) for their financial contribution to this research and the author Ezgi GURGENC would like to thank Council of Higher Education (CoHE) for its scholarship support with the 100/2000 Ph.D. scholarship. For instance, support of the authors is recommended to be given as follows: E.G and A.D. owner of the idea, E.G did the experiment, E.G, A.D and F.A discussed the results, E.G, A.D and F.A wrote the article.

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