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Effect of Gallium Content on Diode Characteristics and Solar Cell Parameters of $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ Thin Film Solar Cells Produced by Three-stage Co-evaporation at Low Temperature

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Anahtar Kelimeler

İnce Film
Güneş Pili
Diyot Özellikleri
Galyum Miktarı

Graphical/Tabular Abstract (Grafik Özet)

In this study, $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells with x values of 0.17, 0.20, 0.23, and 0.26 were successfully produced by three-stage co-evaporation technique at low temperatures. / Bu çalışmada, x değerleri 0,17, 0,20, 0,23 ve 0,26 olan $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pilleri, düşük sıcaklıklarda üç aşamalı eş-buharlaştırma tekniği ile başarıyla üretildi.

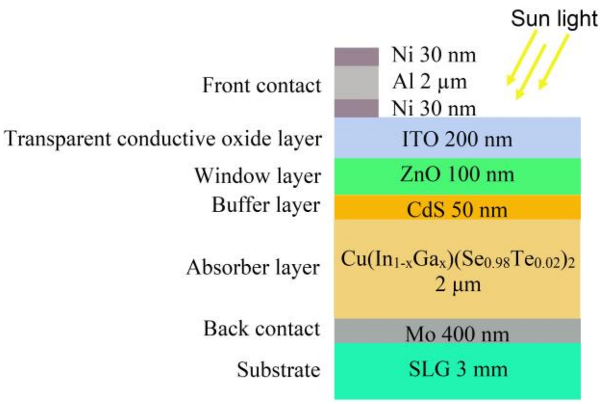

***Figure A**: Schematic illustration of the thin film solar cell / **Şekil A:** İnce film güneş pilinin şematik gösterimi*

Highlights (Önemli noktalar)

- *$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells have been produced by three-stage co-evaporation. / $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pilleri üç aşamalı eş-buharlaştırma ile üretilmiştir.*
- *The change trend of solar cell parameters was similar to the change trend of diode quality. / Güneş pili parametrelerinin değişim eğilimi, diyot kalitesindeki değişim eğilimiyle benzerlik göstermiştir.*
- *The efficiency of the solar cell was improved by optimizing the GGI ratio. / Güneş pilinin verimi GGI oranı optimize edilerek iyileştirilmiştir.*

***Aim (Amaç):** In this study, it was aimed to increase the efficiency of $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cell by optimizing the amount of gallium. / Bu çalışmada, galyum miktarını optimize ederek $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pilinin veriminin artırılması amaçlanmıştır.*

***Originality (Özgünlük):** $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells with different x values have been produced by three-stage co-evaporation technique for the first time. / Farklı x değerlerine sahip $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pilleri üç aşamalı eş-buharlaştırma tekniği ile ilk kez üretilmiştir.*

***Results (Bulgular):** The increase in gallium content decreased the ideality factor and increased the barrier height up to the GGI ratio of 0.23. The sample Ga0.23 was found to have the highest diode quality with the lowest ideality factor value. The change trend of solar cell parameters was similar to the change trend of diode quality. / 0,23 GGI oranına kadar galyum miktarındaki artış idealite faktörünü azaltmış ve bariyer yüksekliğini artırmıştır. Ga0.23 numunesinin en düşük idealite föktörü değeri ile en yüksek diyot kalitesine sahip olduğu bulunmuştur. Güneş pili parametrelerinin değişim eğilimi, diyot kalitesindeki değişim eğilimiyle benzerlik göstermiştir.*

***Conclusion (Sonuç):** The power conversion efficiency of the SLG/Mo/$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$/CdS/ZnO/ITO/Ni-Al-Ni thin film solar cells was increased from 3.7% to 6.3% by increasing the GGI ratio from 0.17 to 0.23. / GGI oranı 0,17'den 0,23'e çıkarılarak SLG/Mo/$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$/CdS/ZnO/ITO/Ni-Al-Ni ince film güneş pillerinin güç dönüşüm verimliliği %3,7'den %6,3'e yükseltilmiştir.*

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Abstract

In this study, $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells with x values of 0.17, 0.20, 0.23, and 0.26 were successfully produced by three-stage co-evaporation technique at low temperatures. The diode characteristics and solar cell parameters of thin film chalcopyrite solar cells with the structure of SLG/Mo/$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$/CdS/ZnO/ITO/Ni-Al-Ni were investigated by current-voltage measurements. The ideality factor, series resistance, and barrier height were obtained by the Cheung-Cheung method using the current-voltage results measured in the dark at room temperature. Open-circuit voltage, short-circuit current density, fill factor, and the power conversion efficiency of the thin film solar cells were derived from the current-voltage measurements realized by a four-point measurement setup under AM1.5G standards at room temperature. It was found that the increase in the gallium content first decreased the ideality factor, however, it increased again after exceeding the x value of 0.23. While the amount of gallium was increasing, fluctuations were observed in the series resistance values. The barrier height first increased with the increasing amount of gallium and decreased after exceeding the x value of 0.23. The solar cell parameters increased by increasing the x value up to 0.23 and decreased after exceeding this point. It was found that the diode parameters have an effect on each other but the most effective diode parameter was the ideality factor. The efficiency of the $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells was increased from 3.7% to 6.3% by increasing the x value from 0.17 to 0.23.

Galyum Miktarının Düşük Sıcaklıkta Üç Aşamalı Eş-Buharlaştırma ile Üretilen $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ İnce Film Güneş Pillerinin Diyot Özellikleri ve Güneş Pili Parametreleri Üzerine Etkisi

Makale Bilgisi

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Anahtar Kelimeler

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Diyot Özellikleri
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Öz

Bu çalışmada, x değerleri 0.17, 0.20, 0.23 ve 0.26 olan $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pilleri, düşük sıcaklıklarda üç aşamalı eş-buharlaştırma tekniği ile başarıyla üretildi. SLG/Mo/$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$/CdS/ZnO/ITO/Ni-Al-Ni yapısına sahip ince film kalkopirit güneş pillerinin diyot özellikleri ve güneş pili parametreleri akım-gerilim ölçümleri ile incelendi. İdealite faktörü, seri direnç ve bariyer yüksekliği, oda sıcaklığında karanlıkta ölçülen akım-gerilim sonuçları kullanılarak Cheung-Cheung yöntemiyle elde edildi. İnce film güneş pillerinin açık devre gerilimi, kısa devre akım yoğunluğu, dolgu faktörü ve güç dönüşüm verimliliği, oda sıcaklığında AM1.5G standartlarında dört noktalı ölçüm düzeneği ile gerçekleştirilen akım-gerilim ölçümlerinden elde edilmiştir. Galyum içeriğindeki artışın önce idealite faktörünü azalttığı, ancak x değeri 0,23'ü geçtikten sonra tekrar arttırdığı tespit edilmiştir. Galyum miktarı artarken seri direnç değerlerinde dalgalanmalar gözlemlenmiştir. Bariyer yüksekliği galyum miktarının artmasıyla önce artmış, x değeri 0,23'ü geçtikten sonra azalmıştır. Güneş pili parametreleri x değerinin 0,23'e kadar artmasıyla artmış, bu noktanın aşılmasıyla azalmıştır. Diyot parametrelerinin birbirini etkilediği ancak en etkili diyot parametresinin idealite faktörü olduğu tespit edilmiştir. $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ ince film güneş pillerinin verimliliği, x değeri 0,17'den 0,23'e artırılarak %3,7'den %6,3'e çıkarılmıştır.

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1. INTRODUCTION (GİRİŞ)

The copper indium gallium selenide (CIGS) compound is a member of the chalcogenide group with its chalcopyrite structure and contains at least one chalcogen (S, Se, Te) in its composition [1,2]. There are also applications such as $Cu(In,Ga)(Se,S)_2$ and $Cu(In,Ga)(Se,Te)_2$, where these chalcogens are in the structure together [3-7]. CIGS thin films are very suitable for thin film solar cell applications with features such as tunable bandgap, stability, and high absorption coefficient [8-10].

Research on thin films and solar cells containing selenium and sulfur as chalcogens has increased considerably. On the other hand, although thin film research containing tellurium as chalcogen has reached a certain number, studies on chalcopyrite solar cells containing tellurium are very few [11-13].

The main motivation for adding tellurium to CIGS solar cells is to reduce the bandgap value and use the CIGS solar cell as the bottom cell of the tandem solar cells [14,15]. However, by adding a low amount of tellurium to the CIGS structure, it is also possible to control the properties of gallium in the structure without reducing the bandgap value [13]. Since the addition of gallium increases the bandgap value, the amount of gallium is very low in studies where the bandgap value is tried to be reduced [16]. On the other hand, in the study where tellurium was added to control the properties of gallium, the amount of gallium was quite high [13].

The issue of how gallium and tellurium affect each other in the presence of moderate amounts of gallium in a CIGS solar cell with tellurium addition has not yet been investigated. In this study, the effect of the gallium amount of the $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ solar cell on the diode parameters by changing the x values to 0.17, 0.20, 0.23, and 0.26 and how the photovoltaic parameters are affected accordingly was examined in a systematical approach.

After the huge potential in electricity production with solar cells was realized, there has been a significant increase in the production of all types of solar cells [17]. Since high temperatures that the substrate material can withstand are generally used for high-efficiency thin film production, the production cost is high. The total thermal budget can be reduced by developing the production of high-efficiency solar cells at lower temperatures. In this study, the production of thin film solar cells at low temperatures was investigated to reduce the production cost.

2. MATERIALS AND METHODS (MATERYAL VE METOD)

$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film absorbers with x values of 0.17, 0.20, 0.23, and 0.26 were deposited on molybdenum coated soda lime glass (SLG) substrates by three-stage co-evaporation method using a multi-source physical vapor deposition chamber under $2x10^{-7}$ mBar vacuum atmosphere. NaF post-deposition treatment was applied after absorber deposition without breaking the vacuum of the chamber.

The final thickness of the absorbers was about 2 µm. The substrate temperature was set to 480℃ to reduce the thermal budget. CdS was coated on the absorbers by chemical bath deposition. ZnO and indium tin oxide (ITO) layers were deposited by RF magnetron sputtering. The thicknesses of CdS, ZnO, and ITO were around 50, 100, and 200 nm, respectively. The front contact, consisting of Ni-Al-Ni layers, was coated by e-beam evaporation. The thickness of the aluminum layer was 2 µm and that of the nickel layers was 30 nm.

The diode characteristics and solar cell parameters of thin film chalcopyrite solar cells with $SLG/Mo/Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2/CdS/ZnO/ITO$ /Ni-Al-Ni structure were analyzed by current-voltage (I-V) measurements. The ideality factor (n), series resistance (R_S), and barrier height (Φ_B) were obtained by the Cheung-Cheung method using the I-V results measured in the dark at room temperature.

Open-circuit voltage (Voc), short-circuit current density (Jsc), fill factor (FF), and the power conversion efficiency (PCE) of the solar cells were derived from the I-V results measured by a four-point measurement setup under AM1.5G standards with 1000 W/m^2 illumination at room temperature. The chemical composition of absorber layers was obtained by energy dispersive spectroscopy (EDS) under 15 kV voltage. The samples with x values of 0.17, 0.20, 0.23, and 0.26 were named Ga0.17, Ga0.20, Ga0.23, and Ga0.26, respectively.

3. RESULTS (BULGULAR)

The chemical composition of the absorber layers is shown in Table 1. Ga/(Ga+In) (GGI) and Cu/(Ga+In) (CGI) ratios, which are very important for solar cell parameters, can be calculated from Table 1. The GGI ratio is also equal to the x value of the samples.

Table 1. The chemical composition of the absorber layers (Absorbe edici katmanların kimyasal bileşimi)

Sample	Cu (at. %)	In (at. %)	Ga (at. %)	Se (at. %)	Te (at. %)
Ga0.17	21.82	21.04	4.38	51.73	1.03
Ga0.20	21.75	20.19	5.19	51.89	0.98
Ga0.23	21.91	19.76	5.95	51.37	1.01
Ga0.26	21.80	19.04	6.76	51.44	0.96

According to the calculations made using the data given in Table 1, the CGI ratios of the samples were approximately 0.85. This ratio is slightly lower than the CGI ratio in similar studies where high solar cell efficiency was achieved [18]. As can be seen from Table 1, the increase in the GGI ratios of the absorbers was adjusted by the increase of gallium and the decrease of indium. This adjustment was achieved by changing the temperatures of the gallium and indium sources of the multi-source physical vapor deposition chamber. When the Te/(Te+Se) (TTS) ratio was calculated according to the values in Table 1, it was seen that this ratio was approximately 0.02 for all absorbers. It was observed that the TTS ratio was slightly higher compared to solar cell studies in which selenium and tellurium were used together as chalcogens [13].

The ideality factor (n), series resistance (R_S), and barrier height (Φ_B) were obtained by the Cheung-Cheung method [19] which can be expressed as follows:

$$\frac{dV}{d(lnI)} = \frac{nkT}{q} + IR_S \quad (1)$$

$$H(I) = V - n\left(\frac{kT}{q}\right) ln\left(\frac{I}{AA^*T^2}\right) \quad (2)$$

$$H(I) = IR_S + n\Phi_B \quad (3)$$

In equation (1), V, I, n, k, T, q, and R_S represent the voltage, current, ideality factor, Boltzmann's constant, temperature in Kelvin, electron charge, and series resistance, respectively. In equation (2), A^* represents the Richardson constant which equals 84 A/cm^2K^2 for p-type CIGS [20]. In equation (3), Φ_B represents the barrier height. The ideality factor and series resistance can be derived from the dV/dlnI versus I graphs shown in Figure 1.

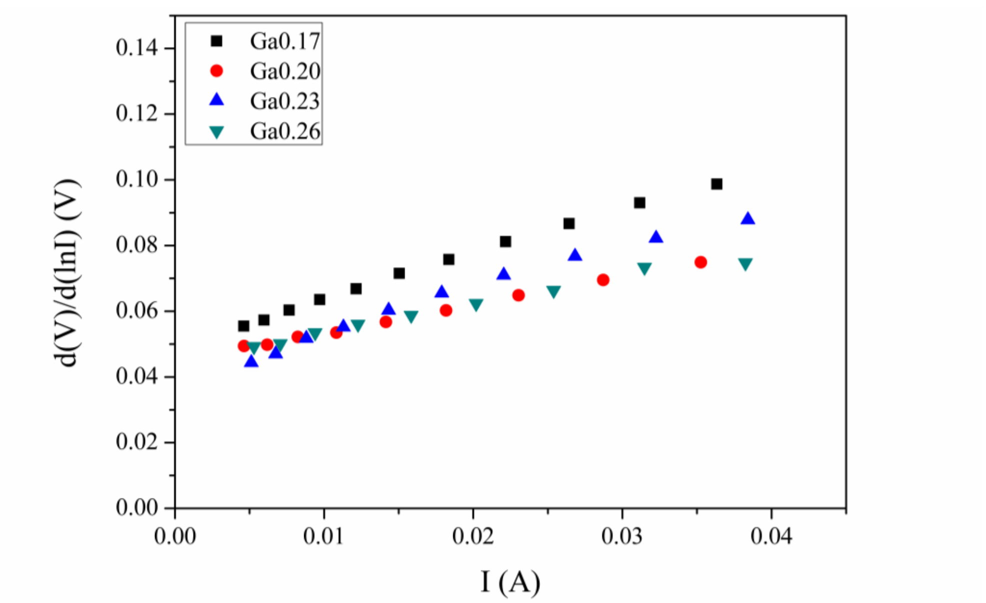

Figure 1. dV/dlnI versus I graphs (dV/dlnI ve I grafikleri)

According to equation (1), the slope of the best fits of the dV/dlnI versus I graphs equals R_S and the ideality factor can be calculated by dividing the y-axis intercept of the best fits by kT/q. R_S values of the samples Ga0.17, Ga0.20, Ga0.23, and Ga0.26 were found to be 1.38, 0.85, 1.32, and 0.82 kΩ, respectively. On the other hand, the ideality factor values of the samples were calculated according to the equation (1) and were found to be 1.90, 1.74, 1.51, and 1.74, respectively. The barrier height can be derived from the H(I) versus I graphs shown in Figure 2.

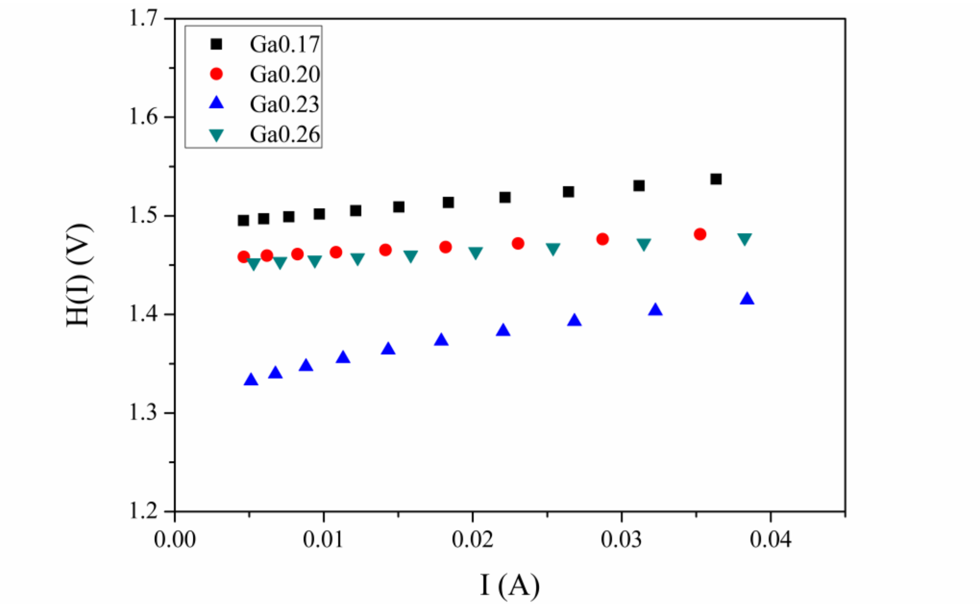

Figure 2. H(I) versus I graphs (H(I) ve I grafikleri)

According to equation (3), the barrier height can be calculated by dividing the y-axis intercept of the best fits of the H(I) versus I graphs by the ideality factor values obtained from equation (1). Barrier height values of the samples Ga0.17, Ga0.20, Ga0.23, and Ga0.26 were found to be 0.78, 0.83, 0.96, and 0.76 eV, respectively. The ideality factor (n), series resistance (R_S), and barrier height (Φ_B) values of the samples are shown in Table 2.

Table 2. The diode parameters of the solar cells (Güneş pillerinin diyot parametreleri)

Sample	Ideality factor (n)	Series resistance (R_S) (kΩ)	Barrier height (Φ_B) (eV)
Ga0.17	1.90	1.38	0.78
Ga0.20	1.74	0.85	0.83
Ga0.23	1.51	1.32	0.96
Ga0.26	1.74	0.82	0.76

The ideality factor shows the deviation of a diode from the thermionic emission model and should be 1 for an ideal diode [21]. As seen in Table 2, the sample with the highest diode quality is the Ga0.23 sample which deviates the least from ideal diode behavior. The increase in the amount of gallium first decreased the ideality factor, but it increased again after the GGI ratio of 0.23. The reason for high ideality factor values may be the current mechanism of the solar cell, the barrier height inhomogeneity, and the recombination–generation mechanism [22,23]. As the amount of gallium increased, fluctuations were observed in the series resistance values. On the other hand, the barrier height first increased with the increasing amount of gallium and decreased after exceeding the GGI ratio of 0.23. Contrary to the linear relationship in the literature [24], there is an inverse relationship in the results. This may be due to the absorber/CdS interface inhomogeneities which can be affected by the gallium amount. Another reason may be the fact that the addition of tellurium may have triggered the formation of an additional barrier in the back contact region by causing the gallium in the structure to accumulate on the back contact side [13]. In this way, as the amount of gallium in the structure increases, the barrier height may also increase. The decrease of the barrier height after the GGI ratio exceeded 0.23 may be due to the decrease in the series resistance [25]. Solar cell parameters of the samples were derived from the current density-voltage graphs which are shown in Figure 3.

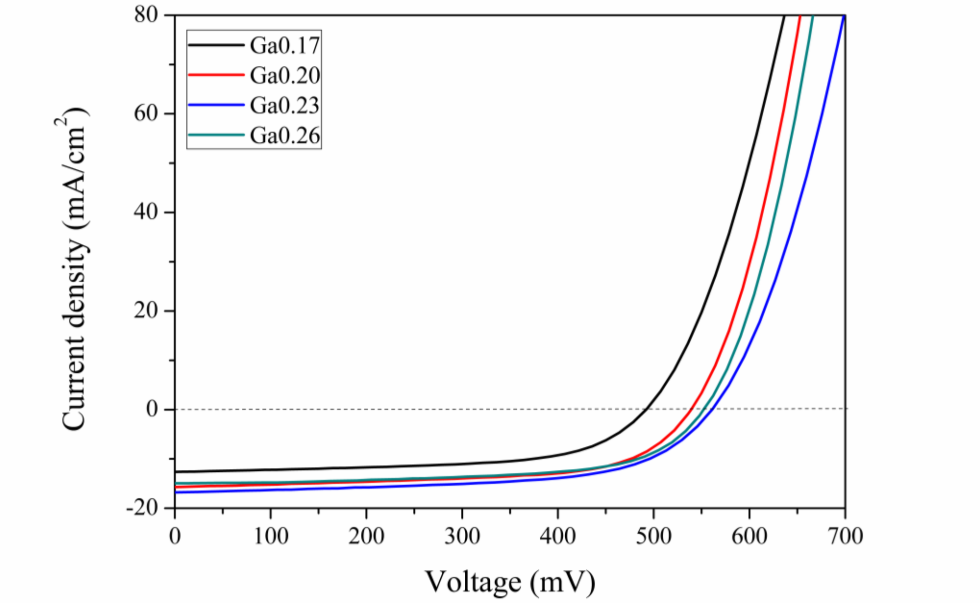

Figure 3. Current density-voltage graphs (Akım yoğunluğu-voltaj grafikleri)

As can be seen in Figure 3, the current at zero voltage, the voltage at zero current, and the area of the part of the graph below zero current are equal to the Jsc, Voc, and FF, respectively. Then the PCE can be calculated by using these parameters. It is clear from Figure 3 that, the Jsc value increased by increasing the gallium amount up to the GGI ratio of 0.23 and then decreased by increasing the gallium amount further. Moreover, the same trend is also seen in Voc and FF values. According to the literature, as the slope in the high voltage region of the J-V graph decreases, the series resistance increases [26]. This situation was found to be in good agreement with the data obtained by the Cheung-Cheung method. Ga0.17 and Ga0.23 samples had higher series resistance values than those of Ga0.20 and Ga0.26 samples. The trend in the change of the solar cell parameters Voc, Jsc, FF, and PCE with the increasing gallium content is shown in Figure 4 for a better understanding.

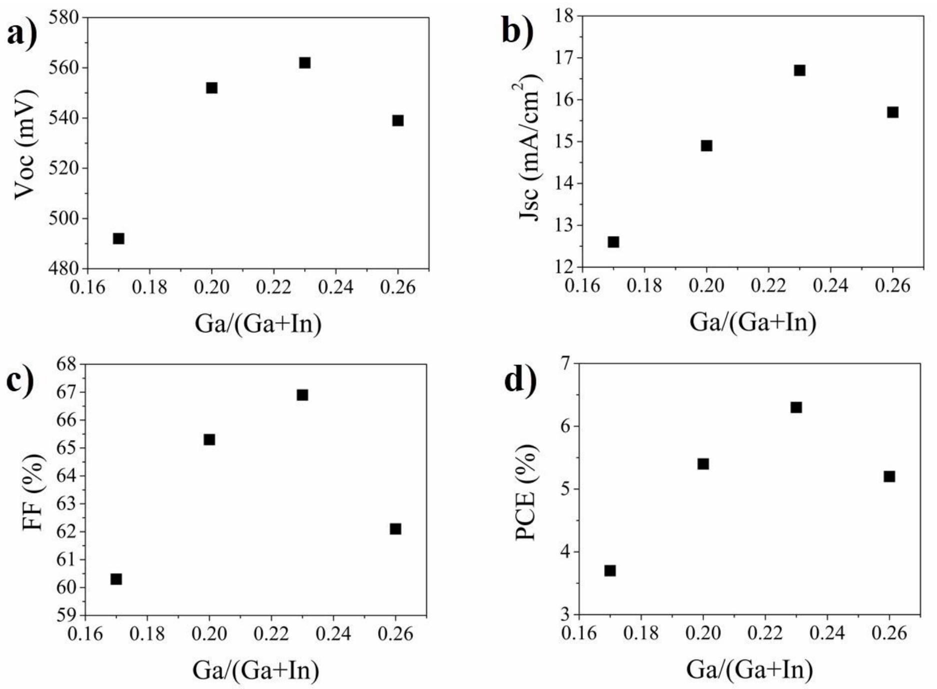

Figure 4. The trend in the change of the solar cell parameters a) Voc, b) Jsc, c) FF, and d) PCE (Güneş pili parametrelerinin değişim eğilimi a) Voc, b) Jsc, c) FF ve d) PCE)

As can be clearly seen in Figure 4, all the solar cell parameters increased by increasing the GGI ratio up to 0.23 and decreased after this point. The reason for the decrease in solar parameters in the sample with the highest GGI ratio may be due to deeper defects acting as recombination centers generated by the high gallium content [27]. Another reason may be the fact that the increase in gallium content may enhance the amount of ordered vacancy compounds (OVCs), which can affect the recombination mechanism of the solar cells [28]. The solar cell parameters of the samples are also shown in Table 3.

Table 3. The solar cell parameters of the samples (Numunelerin güneş pili parametreleri)

Sample	Voc (mV)	Jsc (mA/cm^2)	FF (%)	PCE (%)
Ga0.17	492	12.6	60.3	3.7
Ga0.20	552	14.9	65.3	5.4
Ga0.23	562	16.7	66.9	6.3
Ga0.26	539	15.7	62.1	5.2

When the data in Table 2 and Table 3 are evaluated together, it is understood that the most effective diode parameter on solar parameters is the ideality factor. Although other diode parameters had an effect on the ideality factor, it was observed that they did not have a significant effect on the solar cell parameters. It can be seen from Table 3 that the efficiency of the $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ solar cells was increased from 3.7% to 6.3% by increasing the GGI ratio from 0.17 to 0.23.

4. CONCLUSIONS (SONUÇLAR)

$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ thin film solar cells with x values of 0.17, 0.20, 0.23, and 0.26 were successfully produced at low temperatures. The effect of the gallium amount of the $Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$ solar cell on the diode parameters and the photovoltaic parameters was investigated. The ideality factor, series resistance, and barrier height were obtained by the Cheung-Cheung method. The increase in the gallium content first decreased the ideality factor, however, it increased again after exceeding the GGI ratio of 0.23. The sample Ga0.23 was found to have the highest diode quality with the lowest ideality factor value. As the amount of gallium increased, fluctuations were observed in the series resistance values. On the other hand, the barrier height first increased with the increasing amount of gallium and decreased after exceeding the GGI ratio of 0.23. Solar cell parameters of the samples were derived from the current density-voltage graphs. Series resistance values calculated by the Cheung-Cheung method were found to be in good agreement with the current density-voltage graphs. The change trend of solar cell parameters was similar to the change trend of diode quality. All the solar cell parameters increased by increasing the GGI ratio up to 0.23 and decreased after exceeding this point. It was found that the diode parameters have an effect on each other but the most effective diode parameter was the ideality factor. The power conversion efficiency of the SLG/Mo/$Cu(In_{1-x}Ga_x)(Se_{0.98}Te_{0.02})_2$/CdS/ZnO/ITO/Ni-Al-Ni thin film solar cells was increased from 3.7% to 6.3% by increasing the GGI ratio from 0.17 to 0.23.

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DECLARATION OF ETHICAL STANDARDS (ETİK STANDARTLARIN BEYANI)

The authors of this article declare that the materials and methods they use in their work do not require ethical committee approval and/or legal-specific permission.

Bu makalenin yazarları çalışmalarında kullandıkları materyal ve yöntemlerin etik kurul izni ve/veya yasal-özel bir izin gerektirmediğini beyan ederler.

AUTHORS’ CONTRIBUTIONS (YAZARLARIN KATKILARI)

Semih AĞCA**:** Performing the experiments, analyzing the results, and writing the manuscript.

Deneylerin yapılması, sonuçların analiz edilmesi ve makalenin yazılması.

***Güven ÇANKAYA*:** Bringing the idea, supervising the study, and final check of the manuscript.

Fikrin geliştirilmesi, çalışmanın danışmanlığı ve makalenin son kontrolü.

CONFLICT OF INTEREST (ÇIKAR ÇATIŞMASI)

There is no conflict of interest in this study.

Bu çalışmada herhangi bir çıkar çatışması yoktur.

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