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Research Article

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Enhance the efficiency of [CeO_2(NP)/Cu(NP)] PEC cell photoanode for hydrogen production by laser illumination

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Abstract

This research introduces a novel approach to enhancing solar-to-hydrogen (STH) efficiency by applying laser illumination with the assistance of a UV source to illuminate the photoanode in photoelectrochemical (PEC) cells, representing a significant departure from conventional methods relying solely on solar light. The photoanode was fabricated using a (CeO_2 and Cu) nanoparticles thin film through the drop-casting method, the materials were chosen for their favorable optical and electronic properties, which are key in improving the hydrogen production rate. What sets this study apart is the innovative use of laser illumination, which allows for more precise and concentrated light absorption that match the materials' absorption peaks compared to the broad spectrum of traditional sunlight. The structural properties of the materials were examined using X-Ray diffraction (XRD), (FE-SEM) to get the cross-sectional image and the optical properties were characterized by the UV-VIS-NIR spectrophotometer. Photoelectrochemical measurements were carried out by linear sweep voltammetry in the dark and under illumination condition AM 1.5 G of 100 mW/cm2.Our findings reveal that under laser and UV illumination, the PEC cell substantially improves photocurrent density and efficiency by 12.3%, outperforming traditional solar light irradiation with an efficiency of 5.5%. This work sets a new direction for PEC cell optimization by demonstrating the effectiveness of laser-assisted light in boosting hydrogen production, representing a significant step forward in renewable energy research.

Keywords: Water splitting, PEC, Thin film, hydrogen production.

1. Introduction

The increasing global demand for renewable energy solutions has intensified research into alternative methods for sustainable energy production. One promising avenue is hydrogen generation through photoelectrochemical (PEC) water splitting, a process that converts solar energy into chemical energy by splitting water into hydrogen and oxygen. Hydrogen, as a clean and highly energy-dense fuel, plays a critical role in the emerging hydrogen economy, with its potential to replace fossil fuels in various sectors. [1,2]. However, a major challenge in PEC systems lies in enhancing the solar-to-hydrogen (STH) conversion efficiency, which remains limited by several factors including material performance, light absorption, and charge carrier dynamics[3]. These systems typically involve the use of a photoelectrochemical (PEC) cell, where a semiconductor photoanode absorbs sunlight, generating electron-hole pairs that drive the water-splitting reaction [4]. The efficiency of this process is often limited by the photoanode's ability to absorb light, separate charge carriers, and catalyze the water oxidation reaction. In fact, the rate at which light is absorbed by the photoanode material is thought to be the key to enhanced hydrogen generation. It is estimated that the UV portion of sunlight on Earth makes up just 4%. Consequently, to achieve maximum efficiency, the photoresponse of the working electrode's (photoanode) absorption must reach the visible light area. Realizing the full potential of solar energy conversion technologies can be attributed to the importance of creating effective photoanode materials for PEC cells, such as thin film composites and inventive designs [5].

Historically, solar light has been the primary source of illumination in PEC systems, with pioneering works dating back to the early 1970s when Fujishima and Honda first demonstrated water splitting using a TiO_2 photoanode under ultraviolet light. Since then, efforts to improve efficiency have focused on optimizing the materials and structures of photoanodes, including the development of thin films, nanostructured surfaces, and doped semiconductors [6]. CeO_2 (ceria), for instance, has emerged as a highly attractive material due to its high oxygen vacancy concentration, superior charge storage capability, dielectric property, and stability in water oxidation reactions. Coupled with copper (Cu) nanoparticles, the CeO_2-Cu combination offers improved conductivity and enhanced charge separation, making it an effective photoanode material for PEC applications [7,8]. On the other hand, Cu nanoparticles (NP) have excellent plasmonic properties including the surface plasmon resonance effect that spans the UV–visible region of the optical spectrum [9,10].

In recent years, there has been a growing interest in exploring alternative light sources to further enhance the efficiency of PEC cells [11]. Laser illumination, which provides highly focused, coherent, and monochromatic light, represents a novel approach to boosting photoactivity. Unlike conventional solar light, which covers a broad and less concentrated spectrum, laser light can be precisely tuned to the absorption characteristics of the photoanode material, leading to more efficient photoexcitation and charge generation. This precise control over the light source opens new possibilities for overcoming the limitations posed by traditional sunlight, such as poor energy concentration.

This study seeks to explore the innovative application of laser illumination in PEC water splitting, using a [CeO_2(NP)/Cu(NP)] thin film photoanode fabricated by the drop-casting method. By comparing the performance of this system under both laser and traditional solar light, we aim to assess the extent to which laser illumination can enhance STH efficiency. This work not only builds on the historical advancements in PEC technology but also introduces a novel illumination strategy that could redefine the approach to maximizing hydrogen production in solar-driven systems.

2. Experimental

Fabrication of the (P) photoanode: The P photoanode is a two-layer thin film of CeO_2 (NP) and Cu (NP) drop-casted on a glass substrate coated with fluorine tin oxide (FTO). The CeO_2 is a 99.9% pure powdered nanoparticle that was bought from (HONGWUNEWMATERIAL). A 0.2 g quantity of cerium dioxide (CeO_2) nanoparticles was dispersed in 25 mL of deionized (DI) water to form the suspension. The mixture was magnetically stirred for 1 hour at room temperature to ensure proper dispersion of CeO_2 particles. A micropipette was used to drop-cast 10 drops of the CeO_2 suspension onto the clean (1 × 1) cm^2 FTO substrate at the conductive face to form the first layer. Then, the film was allowed to dry on a hot plate. The 99.99% pure Cu nanoparticles were supplied by (Nanjing Nano Technology Co., ltd.).

The second layer was formed by dispersing 0.2 g of copper (NP) in 25 mL of deionized (DI) water. The suspension was stirred continuously for one hour to achieve a uniform mixture. Following the Cu (NP) suspension preparation, 10 drops were drop-cast onto the pre-formed CeO_2 layer and dried on a hot plate.

3. Characterizations

3.1 X-Ray Diffraction Measurement (XRD)

XRD characterization was used for materials identification to confirm their structural properties. The X-ray diffraction patterns of CeO2 (NP) and Cu (NP) were obtained using (Aeris – Malvern Panalytical's X-ray diffractometer). The scanning was over the required range for 2ϴ values (15 – 75) o with ± 0.02° 2θ peak position accuracy. Peaks of crystalline phases were compared with those of standard compounds of the materials with previous research.

3.2 Optical Analysis

In order to investigate the optical properties of the designed photoanode, the UV-VIS-NIR-1800 (SHIMADZU, Japan) spectrophotometer was used. This instrument was computerized with a CRT screen and keyboard to operate the input value. The absorption spectra were characterized with a wavelength range of (190–1100) nm.

3.2 *Photoelectrochemical Characteristics*

To study the photoelectrochemical performances of the P photoanode as an efficient photonode, it was tested by an electrochemical workstation (ER466, EDAQ company Australia, potentiostat). Ag/AgCl serves as the reference electrode, Pt serves as the counter electrode, and the designed photoanode (P) serves as the working electrode in the three-electrode system. Regular testing at room temperature used a 125 mL quartz cell with 2g KOH in DI water with PH 9, which served as the electrolyte. The linear sweep voltammetry curve was recorded with a scan starting at (0-1) V versus (Ag/AgCl).
J-V curves were obtained at a rate of 100 mW/s and at a frequency of 20 kHz at the corresponding program. The light-emitting diode (LED) Source (42 W, Zethors H7) illuminated the photoanode under an AM 1.5 G condition of (100) mW/cm^2.

4. Results and discussion

The structural information of CeO_2 (NP) and Cu (NP) was characterized by powder X-ray diffraction (XRD) patterns. Figure 1A displays the pattern of the CeO_2 nanoparticles. It revealed all of the major peaks of CeO_2 which coincided with lattice planes (111), (200), (220), (311), (222), and (400) and were positioned at 2θ = 28.5 °, 33.0 °, 47.5 °, 56.3 °, 59.1 ° and 69.4 °. These findings are in line with previous studies on CeO_2 nanoparticles [12,13]. An XRD diffractogram of copper nanoparticles is shown in Fig. 1 B. Typical lines indexed as (111), (200), and (220) that were found at diffraction angles of 43.3 °, 50.4 °, and 74.1 °, respectively, correspond to the copper nanoparticles diffraction peaks. That matches the references [14,15]. Using Field emission scanning electron microscopy FE-SEM, cross-sectional images of the fabricated photoanode have been acquired and displayed in Fig. 2, which shows a recorded thickness ranging between 9.0 and 12.3 µm for the thin film photoanode, including the two layers of CeO_2 (NP) and Cu(NP) with approximately equal thickness for each layer. The absorption enhancement was evaluated through an examination of the absorption spectra of the photoanodes P. As observed from the P's Fig. 3, the bilayer [CeO_2(NP)/Cu(NP)] thin film revealed an optimal light absorption, demonstrated by the three peaks in the visible range (446nm, 646 nm and 709) in addition to the peak of (294 nm) in the UV region. It is important to mention that, among the above peaks of P, the (446) nm is the intensive one. It's clearly that the peak at the UV region, (294) nm related to CeO_2 (NP) layer. While the rest of the peaks are belong to the Cu nanoparticles, which are form the second layer of the P photoanode. This could be in line with reported researches for the absorption peak of Cu (NP) which is around 570 nm [16,17].

The presence of more than one peak in the visible range of absorption spectrum related to the incorporation of a dielectric semiconductor CeO_2 with a plasmonic Cu nanostructure, related to higher efficiency. Because one of the pathways that surface plasmon resonance (SPR) might boost semiconductor photocatalytic activity is by increasing light absorption through metallic enhancement [18,19].

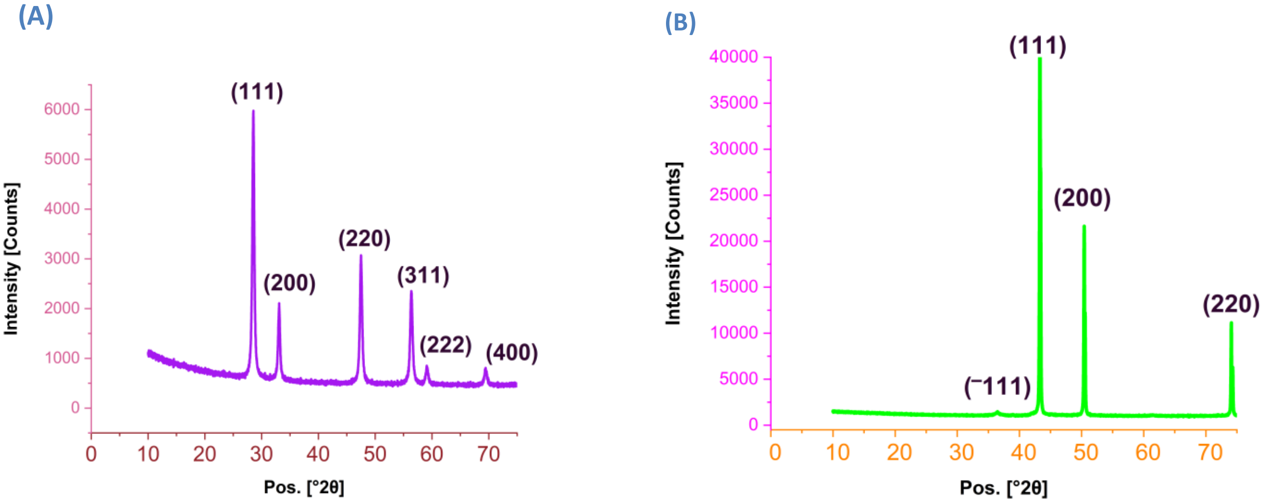

Fig. 1: XRD pattern (A) of CeO_2 NP; (B) of Cu NP.

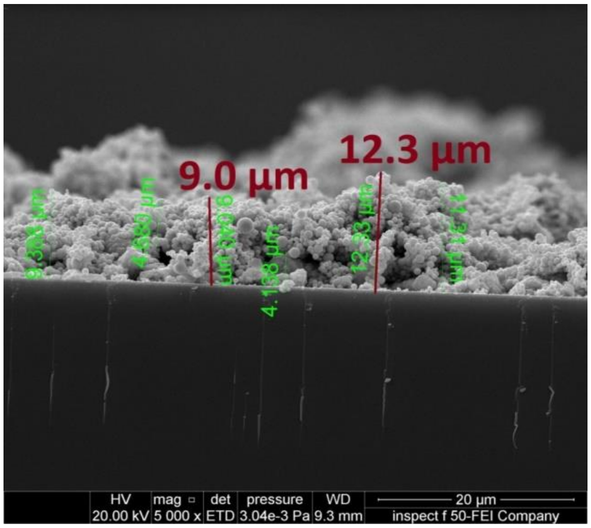

Fig. 2: Cross-sectional FE-SEM image of the (P) photoanode [FTO/CeO_2 (NP) /Cu (NP)].

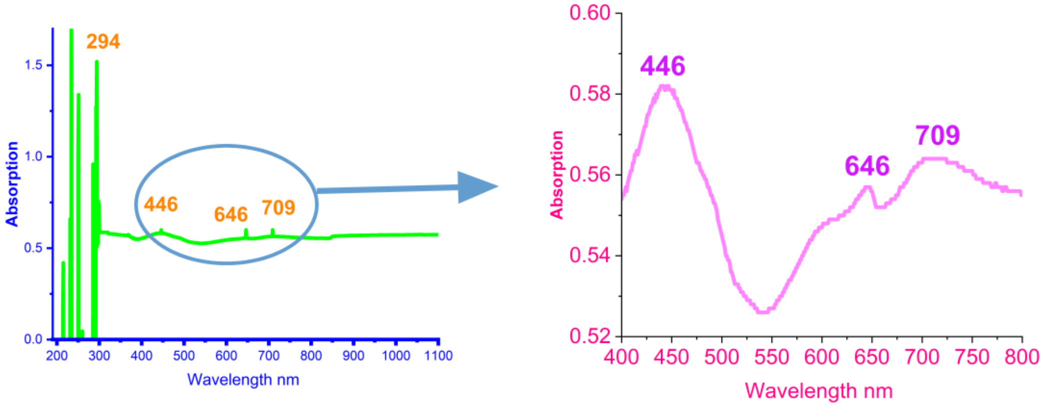

Fig. 3: Absorption spectrum of the (P) photoanode [FTO/CeO_2 (NP) /Cu (NP)]. indicates its absorption peaks at both UV and VIS regions.

Based on these findings, it can be said that the materials chosen for the fabricated photoanode allowed for maximal absorption to be regulated beyond the UV region and cover the visible region of the solar spectrum. The investigation of Dark current measurement, which reflects the baseline current in the absence of light, was the first step in determining the photoelectrochemical performance of the P photoanode. To confirm that incident light independently is the source of the observed photocurrent, to ensure it is not influenced by other factors, and to aim for accurate assessments of PEC performance. As a working electrode, the fabricated photoanode (P) was tested with a scan starting at (0-1) V versus Ag/AgCl in darkness. The dark current density of the P working electrode increased from ($3.2* 10^{-3}$ to $5.0*10^{-3}$) mA. cm^{-2} when the Cu (NP) were added as a second layer in the thin film fabricated. An increase in conductivity is connected to this rise. The low current observed in the P suggests that there aren't numerous defects in the electrode fabrication, which would otherwise cause recombination losses and decreased photoactivity. Due to the differentiation between photocurrent (produced by light-induced processes) and non-photoinduced currents, high dark current may indicate insufficient electrode quality or defective manufacturing. The low dark current in our experiment led to a higher effective efficiency, and stability, which could have an impact on the PEC cell's overall performance.

It was decided that one should utilize the comparison principle in order to achieve the best and most accurate evaluation of the photoelectric response of the the designed photoanode. By examining the LED illumination influence on the P photoanode [FTO/CeO_2(NP)/Cu(NP)] and investigating its behavior, it was found that P exhibited a current density of 23.0 mA.cm^{-2} at 0.99 V vs. Ag/AgCl as displayed in Fig. (4) The positive photogenerated current value indicated that the P photoanode was an n-type semiconductor [20,21]. It is noteworthy to emphasis that the LED source (42 W, Zethors H7) was used to simulate the full solar spectrum [22], meaning that the results obtained accurately showed how this photoanode behave under solar spectrum characteristics.

It was assumed that a new strategy should be followed to improve hydrogen production, where an illumination procedure was adopted, including illumination of the (P) photoanode separately by laser sources with wavelengths of 473 nm, 532 nm, and 632 nm(DPSS laser model MBL-FN-473nm-200mW-15050466, Diode laser 532nm -150mW- Shanghai Dream Laser Technology and Diode laser 632nm). This procedure's principal objective is to examine how these lasers' wavelengths affect the photoanode's current reading by utilizing the idea that the absorption peak of an electrode or substance can be matched to the wavelength of incident light to achieve maximal absorption. The laser wavelengths were chosen closer to each of the P absorption peaks in the visible range. The laser with the best result was then chosen in order to be used as the primary source in the following stage. The results recorded in Fig. (5) illustrated that the 473 nm laser yielded the highest current density of (37.2) mA.cm^{-2} at 0.99 V vs. Ag/AgCl, while the 632 nm laser recorded the lowest current density of (29.1) mA.cm^{-2} at 0.99 V vs. Ag/AgCl. Conversely, the 532 nm laser revealed a value of (33.1) mA.cm^{-2}, which fell in between the 473 and 632 nm lasers.

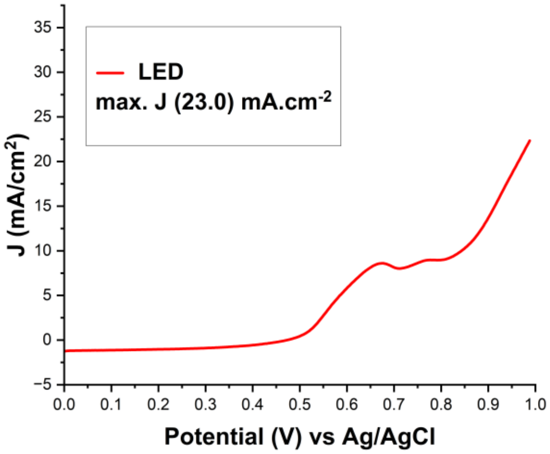

Fig. 4: I-V curve of the P photoanode [FTO/CeO_2 (NP) /Cu (NP)] tested by linear sweep voltammetry under AM 1.5G Illumination of LED source.

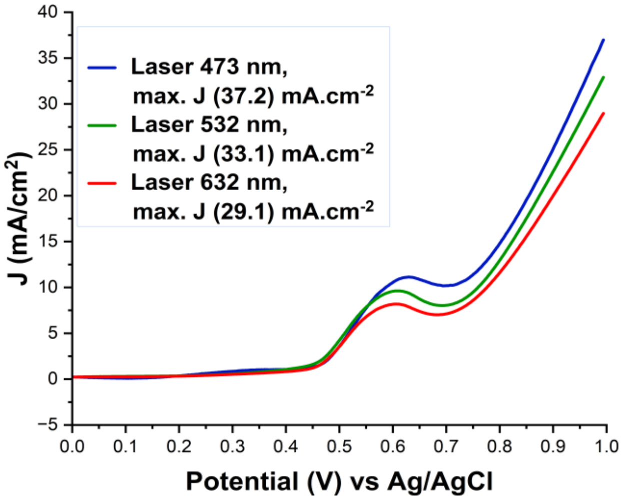

Fig. 5: I-V curve of the P photoanode [FTO/CeO_2 (NP) /Cu (NP)] tested by linear sweep voltammetry under AM 1.5G Illumination of 473 nm, 532 nm and 632 nm lasers sources (separately).

These findings align with the absorption peaks which identified in Fig. (3) as the maximum peak reported at 446 nm, a higher photocurrent value associated with laser 473 nm was observed. Consequently, due to its highest effect, the 473 nm laser was selected and used in the following step of the new procedure which was carried out by simultaneously exposing the P photoanode to a UV source of 6 W which has a wavelength of around 290 nm and a laser beam with a wavelength of 473 nm under AM 1.5 G illumination condition. The photocurrent density of the P photoanode increased significantly to (51.1) mA.cm-2 at 0.99 V vs. Ag/AgCl, which emphasizing the benefits of the photoanode modification and procedure that were employed. Many factors contributed to this rise in current density, including the high conductivity of Cu

(NP), the materials' nanostructure, which enhanced the operative surface area, and the strong SPR effect, which was primarily responsible for the high measured current. The surface plasmon resonance (SPR) effect produced by Cu nanoparticles (NPS) on the dielectric semiconductor CeO_2 extends the light adsorption edge, encourages charge separation, and improves hydrogen generation [8,21,23-25]. This result demonstrates the significant improvement obtained when compared to that of the influence of LED on the P photoanode. This comparison is clear in Fig.(6), the high increment from (23.0) mA.cm^{-2} to (51.1) mA.cm^{-2} at 0.99 V vs. Ag/AgCl shows that employing a laser 473 nm + UV source might boost current by roughly (28.1) mA.cm^{-2}. A closer look at the same figure reveals that while the photoanode P is illuminated by different sources, its behaviour is similar but not identical. While as can be seen in Fig. (5) where the illumination source is comparable since it is a laser with a different wavelength, the behavior is completely identical with a rise in the photoelectric response. All the results of photocurrent densities for the P fabricated photoanode are illustrated in Table 1.

Table 1: illustrates the photocurrent density at 0.99 V vs. Ag/AgCl of the P photoanode with the illuminated sources and their represented readings.

Result Symbol	Illumination source	Thin film components on a (1*1) cm^2 FTO substrate	Photocurrent density at 0.99 V vs. Ag/AgCl J (mA.cm^{-2})
R1	LED	CeO_2(NP)/Cu(NP)	23.0
R2	Laser 473 nm	CeO_2(NP)/Cu(NP)	37.2
R3	Laser 532 nm	CeO2(NP)/Cu(NP)	33.1
R4	Laser 632 nm	CeO_2(NP)/Cu(NP)	29.1
R5	Laser 473 nm + UV source	CeO_2(NP)/Cu(NP)	**51.1**

The Solar-to-Hydrogen efficiency (η) of all the resulting current densities is presented in Table 2. And in a 3-electrode PEC configuration, when an external bias is applied between the working (WE) and the counter electrodes (CE), conversion efficiency is usually evaluated using the following equation [26,27]:

$$\eta = [I\,(1.23 - E\,\text{bias}) / J\,\text{light}] \times 100\% \qquad (1)$$

Where, (I) is photocurrent density at the given potential observed from the experiment (mA.cm^{-2}), E bias is the used potential, 1.23 is the criterion of potential of water splitting, and J light refers to the intensity of the irradiating light at 100 mW/cm^2 for AM 1.5 G standard.

Table 2: The results of the calculated Solar-to-Hydrogen efficiencies for the P photoanode illuminated by different sources.

Result Symbol	Illumination source	Thin film components on a (1*1) cm^2 FTO substrate	Solar-to-Hydrogen efficiency (η)
R1	LED	CeO_2(NP)/Cu(NP)	5.5 %
R2	Laser 473 nm	CeO_2(NP)/Cu(NP)	8.9 %
R3	Laser 532 nm	CeO2(NP)/Cu(NP)	7.9 %
R4	Laser 632 nm	CeO_2(NP)/Cu(NP)	7.0 %
R5	Laser 473 nm + UV source	CeO_2(NP)/Cu(NP)	12.3 %

To visually compare values of all result calculated efficiencies, Fig. (6) provides information on the percentage efficiency of the photoconversion process. The Solar-to-Hydrogen efficiency of the P fabricated photoanode [CeO_2(NP)/Cu(NP)] which was illuminated by employing (laser 473 nm + UV source) is much higher than other resulted efficiencies , which implies that P have a strong ability for hydrogen production when the new procedure employed in comparison to traditional illumination represented by the LED source as it doubled increasing from (5.5%) to (12.3%). Moreover, the response at lower efficiencies is also considerably good results at the same time. This leads in overall good performance of the PEC cell

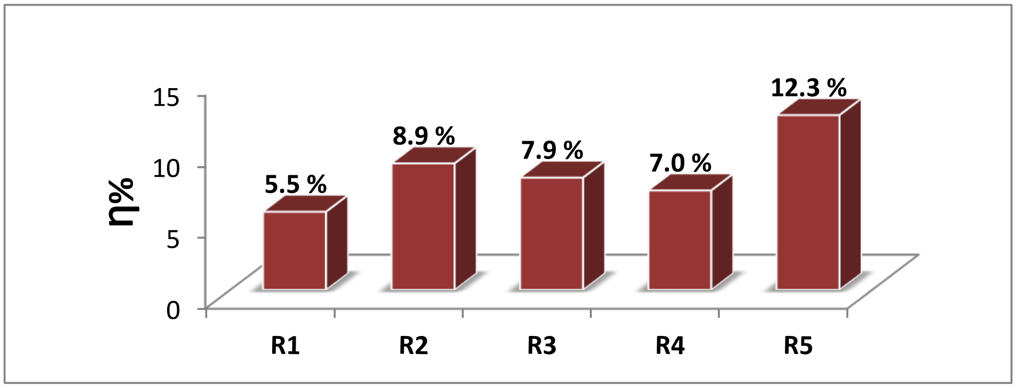

Fig. 6: Diagram illustrating the calculated efficiency for each outcome as: R1 of LED source, R2 of Laser 473 nm, R3 of Laser 532 nm, R4 of Laser 632 nm, and R5 of Laser 473 nm + UV source.

5. Conclusions

An efficient and low-cost photoanode for PEC cells was designed and fabricated. Its absorption was successfully controlled and enhanced to cover the UV and visible regions. Fine materials were selected and deposited on an FTO substrate by drop-casting technique. The first photoanode layer was formed by cerium oxide nanoparticles, which were then cast with copper nanoparticles to form the second layer. Advanced procedures were used to illuminate this photoanode by using laser and UV sources, resulting in higher photoconversion efficiency. The strategy was examined by comparisons with a conventional light source (LED) by linear scanning voltage with AM1.5 G of (100) mW/cm^2. The photoanode illuminated by the LED source was shown to record a current density of (23.0) $mA.cm^{-2}$ at 0.99 V vs Ag/AgCl. While an impressive result recorded to the implementation of our new procedure. As the photoanode is first illuminated by three laser sources separately with wavelengths matching the photoanode absorption Peaks in the visible region. The higher current density was recorded for the 473 nm laser source of (37.2) $mA.cm^{-2}$ at 0.99 V vs. Ag/AgCl. The second step of the new procedure of illumination the P photoanode [CeO_2/Cu(NP)] by 473 nm laser + UV source which enhanced the current density up to (51.1) $mA.cm^{-2}$ at 0.99 V vs. Ag/AgCl. This result show the validity of the strategy that was adopted and the hypothesis that was taken into considered when carrying out this investigation, and demonstrated the high performance of the PEC cell configuration with these electrodes designed to produce hydrogen via the water splitting process, which can be expressed in a practical and clear way through the high recorded Solar-to- Hydrogen efficiency of up to 12.3 %.

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تحسين كفاءة قطب الخلية الفوتوكهروكيميائية $[CeO_2(NP)/Cu(NP)]$ لانتاج الهيدروجين بواسطة اضاءة الليزر

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الخلاصة

يقدم هذا البحث نهجًا جديدًا لتعزيز كفاءة تحويل الطاقة الشمسية إلى هيدروجين (STH) من خلال تطبيق الإضاءة بالليزر بمساعدة مصدر الأشعة فوق البنفسجية لإضاءة الأنود الضوئي في الخلايا الكهروضوئية (PEC)، مما يمثل انعطافًا كبيرًا عن الطرق التقليدية التي تعتمد فقط على ضوء الشمس. تم تصنيع الأنود الضوئي باستخدام فيلم رقيق من الجسيمات النانوية لاوكسيد السيريوم والنحاس (CeO_2 و Cu) من خلال طريقة الصب بالتنقيط، وقد تم اختيار المواد لخصائصها البصرية والإلكترونية المواتية، والتي تعد مفتاحًا لتحسين معدل إنتاج الهيدروجين. ما يميز هذه الدراسة هو الاستخدام المبتكر لإضاءة الليزر، مما يسمح بامتصاص الضوء بشكل أكثر دقة وتركيزًا يتوافق مع ذروات امتصاص المواد مقارنة بالطيف الواسع لأشعة الشمس التقليدية. تم فحص الخصائص البنيوية للمواد باستخدام حيود الأشعة السينية (XRD) و (FE-SEM) للحصول على صورة مقطعية وتم وصف الخصائص البصرية بواسطة مطياف UV-VIS-NIR. تم إجراء القياسات الكهروضوئية الكيميائية باستخدام الفولتية الخطية في الظلام وفي ظل ظروف الإضاءة AM 1.5 G من 100 mW / cm2. تكشف نتائجنا أنه تحت إضاءة الليزر والأشعة فوق البنفسجية، تعمل خلية PEC على تحسين كثافة التيار الضوئي وكفاءتها بشكل كبير بنسبة 12.3٪، متفوقة على إشعاع الضوء الشمسي التقليدي بكفاءة 5.5٪. يحدد هذا العمل اتجاهًا جديدًا لتحسين خلية PEC من خلال إظهار فعالية الضوء بمساعدة الليزر في تعزيز إنتاج الهيدروجين، مما يمثل خطوة مهمة إلى الأمام في أبحاث الطاقة المتجددة..