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Article

Impact of light intensity and electrolyte volume on performance of photo-electrochemical (PEC) solar cell

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SUMMARY

The world's energy demand is continuously increasing. Non-renewable energy sources, such as petroleum and coal, are extremely limited. On the other hand, conversion efficiency of renewable energy sources into useful forms is insufficient. In our study, we pursued methods to make more efficient solar cells – in particular, photo-electrochemical (PEC) solar cells. We selected the Molybdenum diselenide ($MoSe_2$) based PEC solar cell because of its high resistance to corrosion. First, we explored the relationship between light intensity and efficiency of PEC solar cells. We expected that corrosion of the PEC solar cell would increase with increase in intensity of incident light. Thus, we hypothesized that the efficiency of the solar cell would decrease with increased intensity of incident light. Further, we investigated the relationship between solar cell efficiency and electrolyte volume. We hypothesized that efficiency of PEC solar cell would increase as the electrolyte volume increased. We analyzed several parameters—such as light intensity, fill factor, efficiency, etc.—to draw conclusions. First, we found that percentage efficiency of PEC solar cell was proportional to (light intensity)$^{-0.9}$. Further analysis showed that PEC solar cell performance was positively influenced by increasing the electrolyte volume. Data revealed that this increase in solar cell performance was more consequential at low intensities of light (around 10 mW/cm^2). Our study could contribute to improving the quality of PEC solar cells and might help in the development of carbon-free hydrogen economy.

INTRODUCTION

There has been a recent spike in demands for solar energy due to several disadvantages of non-renewable energy sources (such as petroleum and coal), which has attracted researchers' attention to develop more efficient solar cells (1). Apart from being limited in amount, non-renewable resources create a large amount of pollution (2). Furthermore, renewable energy sources are becoming more cost-efficient in comparison to fossil-fuels (2). With increasing prices of oil and petroleum, the world is turning to solar energy as their main energy source (2, 3). The total energy production by solar power was 254.67 TWh in 2015, which rose to 844.39 TWh in 2020 (2). With such drastic shift of global energy distribution towards solar energy and other renewables, the need for more efficient solar cells has risen. Consumers prefer solar energy over other sources due to its incredible availability. Estimates say that the sun provides enough energy in 80 minutes to supply the world's energy needs for 1 year (4). Another benefit of solar energy is that it is a clean energy source; it does not emit large amounts of carbon dioxide (CO_2) and hence does not contribute to the increase in global warming by the green-house effect (2).

Photo-electrochemical (PEC) solar cells are solid-liquid junction solar cells that convert light energy into electrical energy. There has been rapid progress in the field of PEC solar cells since they were first created as a laboratory version of the traditional solid-solid junction solar cells (5). When photons strike the semiconductor ($MoSe_2$ in this case) of PEC solar cell, some of the electrons get excited. These electrons are then conducted along the circuit created by electrolyte solution, working electrode, counter electrode, and external circuit producing an electric current **(Figure 1)**. In solid-solid junction solar cells, on the other hand, there is no electrolyte; rather, the opposite polarity junctions called positive-negative junction are in contact with each other. Researchers are currently working toward using PEC solar cells as an efficient source of hydrogen fuel (6, 7). The electrons produced during photo-excitation (excitation of electrons when photons of light fall on semiconductor) get involved in following reactions (7):

$$H_2O(l) \xrightarrow{light} 2H^+(aq) + \frac{1}{2}O_2(g) + 2e^- \quad \{1\}$$

$$2H^+(aq) + 2e^- \rightarrow H_2(g) \quad \{2\}$$

The hydrogen gas produced can be used as hydrogen fuel. The hydrogen fuel is potentially useful since it does not release CO_2 on combustion; rather, it produces H_2O (water), which is significantly less harmful to the environment (8). However, there are several challenges which must be overcome before PEC solar cell can be used as a producer of hydrogen fuel. The current efficiency of PEC solar cells is not sufficient to be industrially useful for carbon-free hydrogen economy (9). Thus, we questioned if the efficiency of PEC solar cells could be somehow improved.

Our study aimed to improve the efficiency of PEC solar cells. We created a PEC solar cell as a simple model system to evaluate the relationship of efficiency with the intensity of the incident light and volume of electrolyte. We

fabricated the working electrode using $MoSe_2$. We used the reference electrode to measure the characteristic voltage **(Figure 1)**. The electrolyte consists of a combination of 0.035M I_2 + 0.5 M NaI + 0.5M Na_2SO_4 solution, which serves as an electrical connection between different parts of the circuit. We investigated the performance of PEC solar cells using $MoSe_2$ crystal as a semiconducting electrode. $MoSe_2$ is a member of group-VI transition metal dichalcogenides (TMDCs), a class of materials with structure MX_2, where M is a group VI transition metal and X is a chalcogen (9). $MoSe_2$ has been widely investigated because of its interesting electrical and optical properties, such as d → d transitions (the transfer of one electron from the lower energy d-orbital to another higher energy d-orbital when excited by a photon) (9). Whenever a high energy photon is incident on $MoSe_2$, energy is dissipated by d → d transitions (10, 11). This dissipation of energy ensures that normal covalent bonding of $MoSe_2$ crystal remains intact (10, 11). Thus, $MoSe_2$ is highly resistant to corrosion, which makes it an ideal material for studying the performance of solar cells.

The percentage efficiency of a solar cell is a theoretical relationship between maximum power output, intensity of incident light, and the area of the crystal. The performance of a solar cell is better when its percentage efficiency is higher. Maximum theoretical power output and fill factor are some other mathematical relationships which are directly correlated to the performance of solar cell. In general, higher fill factor represents better performance of solar cell.

Apart from these mathematical considerations, we needed to know the types of defects in solids that might be used as electrodes. There are two kinds of defects of interest: macroscopic and microscopic defects. Macroscopic defects range from terraces, steps and edges, while the microscopic defects include atomic vacancies, grain boundaries, or anything where the uniform crystal structure is missing or modified (12). These microscopic defects are usually affected when incident beam of light strikes the surface of the electrodes. Taking this into consideration, we hypothesized that solar cell efficiency would be inversely related to the intensity of incident light. Further, we hypothesized that as the volume of electrolyte increases, the resistance of internal circuit will decrease due to an increase in conducting free ions. We analyzed several parameters such as light intensity, fill factor, efficiency, photovoltage (V_{ph}), and photocurrent (I_{ph}) to draw conclusions. Our results supported both of our hypotheses. The first experiment showed that percentage efficiency of PEC solar cell was significantly proportional to (light intensity)$^{-09}$. The second experiment revealed that performance of solar cell was positively influenced by increasing the volume of electrolyte, which was more consequential at low intensities of light (around 10 mW/cm^2).

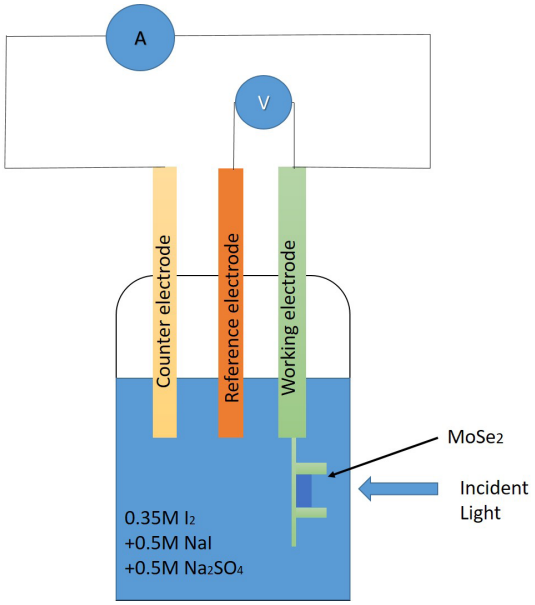

Figure 1. Experimental setup of the PEC solar cell circuit, showing the positions of ammeter (A) and voltmeter (V), used to determine measure I_{ph} and V_{ph} respectively.

RESULTS

First, to determine relation between intensity of incident light and efficiency of solar cell for the first experiment, we irradiated polychromatic light (produced by an incandescent lamp) of different intensities on a PEC solar cell **(Figure 1)**. The emission spectrum (obtained by spectrophotometry) of the incandescent lamp peaked near infrared (wavelength ranged from 600nm to 700nm approximately). We took the reading of the Iph and Vph using digital multimeters **(Figure 1)**. We obtained the Iph vs. Vph characteristic curve of the PEC solar cell by plotting different readings of voltmeter and ammeter when the incident light ray was at 10 mW/cm^2 of intensity and the volume of the electrolyte was 250 mL **(Figure 2)**. This characteristic curve matched with theoretical aspects of solar cells (13).

We obtained the margin of error in measurement of I_{ph} and V_{ph} using the auto-range function of the corresponding multimeter. This margin of error was used to plot error bars of Iph vs. Vph characteristic curve of the PEC solar cell. The auto-range function accounted for instrumental error of the digital

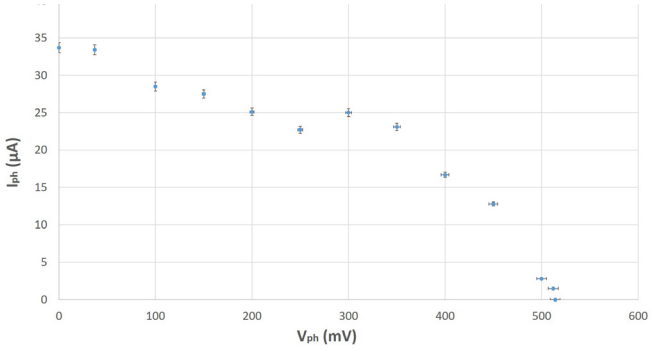

Figure 2. I_{ph} vs. V_{ph} characteristics of PEC solar cell (n=2). Horizontal and vertical error bars present percentage error in measurement of V_{ph} and I_{ph} respectively calculated by auto-range function of the multimeters used to record each measurement.

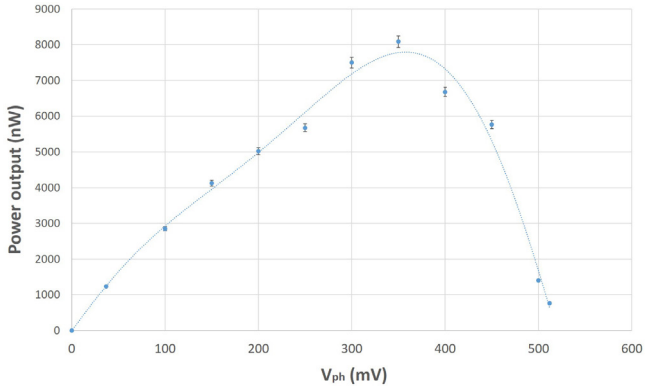

Figure 3. Power output vs. V_{ph} graph used to determine maximum output power of PEC solar cell at a particular value of intensity of incident light (n=2). Vertical error bars present percentage error in the measurement of power output.

multimeters, which were used as ammeter and voltmeter in our study **(Figure 1)**. Using this function, the value of margin of error for calculation of Iph came out to be 2%, while that for calculation of Vph came out to be 1% **(Figure 2)**.

We then predicted the maximum power delivered by using the values of I_{ph} and V_{ph} from our experiment. We found the power delivered at a particular I_{ph} using the relationship between power delivered, I_{ph}, and V_{ph}. Power maxima was obtained when V_{ph} was equal to 350 ± 5 mV **(Figure 3)**. Using the I_{ph} at this value of V_{ph} we obtained P_{max}≈8000 nW (where P_{max} is maximum output power at a particular value of intensity of light and volume of electrolyte).

The experiment was repeated with multiple values of intensity of light for 250 mL electrolyte. Each experiment was performed twice (n=2). We calculated the variation of efficiency of the solar cell on changing the intensity of light from 10 to 120 mW/cm^2 and found that the efficiency of solar conversion decreased with increased intensity of light, which is in accordance with our initial hypothesis **(Figure 4)**.

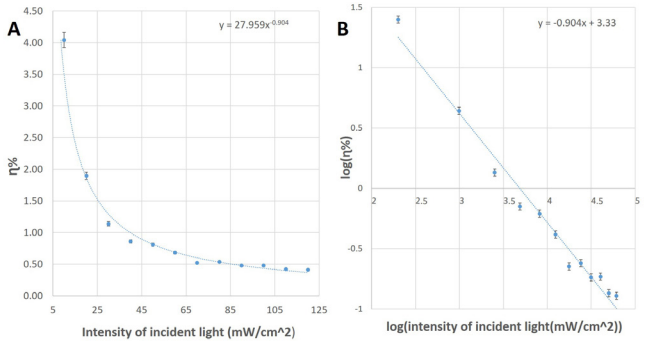

Figure 4. Percentage efficiency vs. intensity (n=2). Error bars present error in measurement of parameters calculated using auto-range function of multimeters. **(A)** Relation between efficiency of $MoSe_2$ based PEC solar cell and intensity of the incident light for the range of intensity 10mw/cm^2 – 120mW/cm^2. **(B)** Efficiency vs. intensity graph drawn on the logarithmic scale. Efficiency of PEC solar cell is significantly proportional to inverse 0.9th power of intensity (One-Way ANOVA, p=0.04).

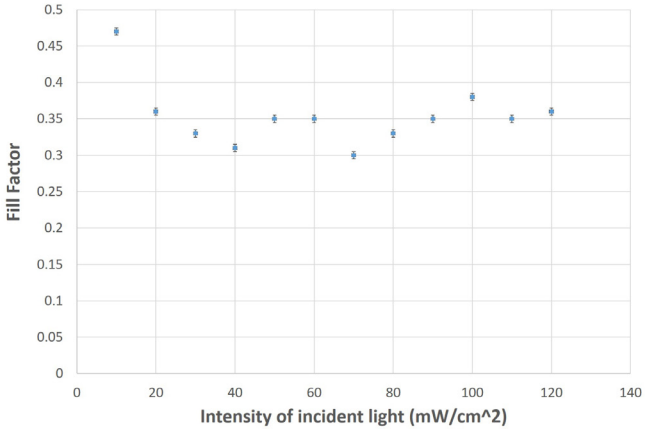

Figure 5. Variation in Fill Factor of PEC solar cell with intensity of incident light (n=2). Error bars present error in measurement of fill factor calculated by auto-range function of multimeters.

To further assess the performance of the solar cell by changing light intensities and volume of electrolyte, we plotted the variation in fill factor and intensity of light **(Figure 5)**. The fill factor variation was not systematic; rather, it was random and could not be explained by the factors we measured for. Hence, we concluded that the actual performance of solar cell should be calculated using the variation in its efficiency, which is physically explainable, and theoretical fill factor should be avoided until further experiments are performed to explain the random variation.

The open circuit voltage (V_{oc}) did not show a general increasing or decreasing trend with intensity of light but was random **(Figure 6a)**, which might be explained by complex factors such as macroscopic and microscopic defects of semiconductors and simpler experimental factors such as intensity of light control. On the other hand, short circuit current (I_{sc}) showed a general positive correlation with intensity of light perhaps because of the increase in number of photons incident with increasing intensity of incident light **(Figure 6b)**.

For the second experiment, the analysis used in the first experiment was repeated using different volumes of electrolyte, including 100 mL, 150 mL, 200 mL, and 300 mL, keeping the concentration of electrolyte constant. We found that, in accordance to our initial hypothesis, as the volume

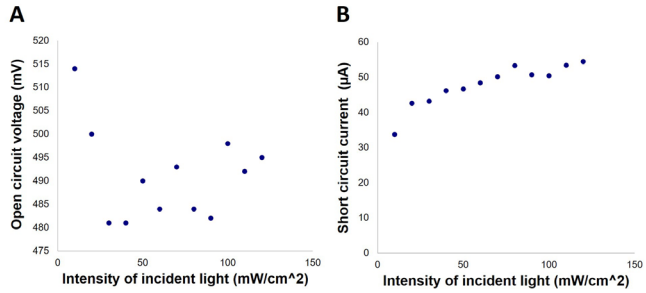

Figure 6. Circuit parameters vs. intensity of incident light (n=2). **(A)** Change in open circuit voltage with change in intensity. **(B)** Change in short circuit current with change in intensity.

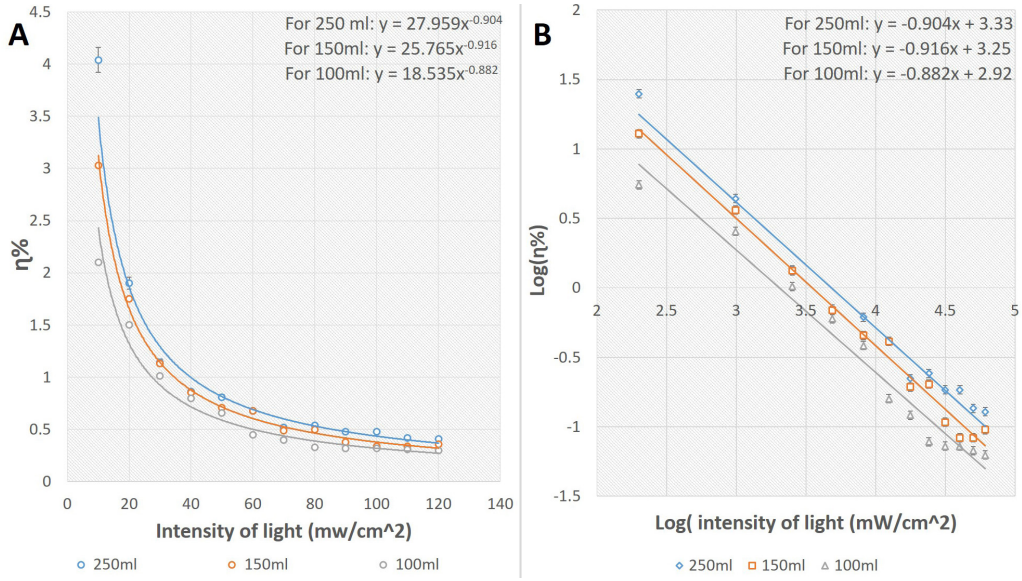

Figure 7. Efficiency vs. intensity curves used to compare significance of increase in efficiency of PEC solar cell with increase in volume of electrolyte (n=2). Error bars present error in measurement of parameters calculated using auto-range function of multimeters. **(A)** The curves represent efficiency vs. intensity when 250 mL (blue), 150 mL (orange), and 100 mL (grey) of electrolyte was used. **(B)** Efficiency vs. intensity curve on the logarithmic scale for 250 mL, 150 mL, and 100 mL, respectively. Efficiency of PEC solar cell is significantly proportional to inverse 0.9th power of intensity.

of electrolyte increased there was an increase in efficiency of PEC cell on an average **(Figure 7)**. However, it was inconsequential at higher values of intensity of incident light (intensity > 20 mW/cm^2). Therefore, we concluded that solar cells with a higher volume of electrolyte performed more efficiently than those with a lower volume of electrolyte for low intensities of light (around 10 mW/cm^2), but at higher intensities, the volume of electrolyte played no major role.

To predict the efficiency of PEC solar cell at intensity of light other than those included in our experiment, we modelled the approximate variation in percentage efficiency (η%) of solar cell with respect to change in intensity of light, as shown in Equation 1:

$$\eta\%_{250ml} \approx 28 \times [Intensity\ of\ incident\ light(^{mW}/_{cm^2})]^{-0.9} \quad (1)$$

To check whether the inverse 0.9th power of intensity variation of efficiency worked for other volumes as well, we plotted intensity vs. efficiency for 150 mL, and we got the same results. Equations 2 and 3 represent the value of efficiency for 150 mL and 100 mL of electrolyte, respectively.

$$\eta\%_{150ml} \approx 26 \times [Intensity\ of\ incident\ light(^{mW}/_{cm^2})]^{-0.9} \quad (2)$$

$$\eta\%_{100ml} \approx 19 \times [Intensity\ of\ incident\ light(^{mW}/_{cm^2})]^{-0.9} \quad (3)$$

Thus, we concluded that within the intensity range 10 mW/cm^2 to 120 mW/cm^2, percentage efficiency was significantly proportional to *Light Intensity*$^{-0.9}$ (One-Way ANOVA p=0.04, **Figure 7**).

DISCUSSION

The aim of our work was to find the optimal conditions for PEC solar cells to have maximum efficiency. In the first experiment, we found that efficiency was reduced as light intensity increased. We theorize that the reason for the decrease in efficiency of the PEC solar cell could be an increase in the number of macroscopic and microscopic defects in the semiconducting device, which might affect the quality of electrodes. The microscopic defects tend to increase as we provide external energy to them. This increase is a consequence of the fact that atomic vibrations increase due to the photons excited from outside, which provides the necessary activation energy needed for the atoms to escape the physical force responsible for holding the atoms in place in any crystal (14). In other words, when the intensity of the incident light rays increases, the number of microscopic defects might increase leading to reduction in efficiency of conversion process. This theory could be tested in future experiments by observing and calculating the defect density of the electrode. Combined with the I_{sc}-intensity curve, these results prompted interesting inferences **(Figure 6b)**. If more radiation from the sun falls on the PEC solar cell, then though the total output will increase due to increase in I_{ph} **(Figure 6b)**, the overall efficiency of conversion will be significantly reduced due to an increment in defects **(Figure 4)**.

The increase in efficiency in the second part of our experiment could be attributed to the fact that the number of ions increased with an increase in volume of electrolyte. This increased number of ions would increase total amount of I_{ph} for same illumination, thus increasing the efficiency of the solar cell. Our experiment showed that the increase

in efficiency was not consequential at higher intensities (intensity ≥ 20 mW/cm^2) **(Figure 6b)**. One reason that the increase in efficiency is not consequential at higher intensities (intensity ≥ 20 mW/cm^2) could be that higher intensities have a higher amount of radiation, which in turn means higher I_{ph} **(Figure 6b)**. Due to this, the number of ions in the electrolyte solution would already be significantly high in number; hence, a greater number of ions obtained by increase in volume of electrolyte will not cause any significant effect on efficiency. However, further studies are required to find exact reason for this observation.

Once source of error which could have affected our readings was the residual light from environment. The light from environment could have affected the reading of photometer and might have reduced the accuracy of our readings. So, we accounted for this error by taking the null reading (reading of photometer when source of light of our experiment was switched off). Still, the environmental conditions can fluctuate and these could be a source of random error in our experiment. Therefore, our experiment was conducted in a dark room to minimize external fluctuations. The error which still remained would be statistically insignificant and hence cause no threat to our data analysis.

Error in measurements can also be attributed to a number of factors such as poor quality of exposed surfaces, the series resistance of PEC solar cells, and the defect density in the semiconductor. We ignored the error that arose due to these factors for the sake of simplicity, but future experiments could be made more statistically sound by taking into account those factors. Throughout our experiment, we had not included the small fluctuations in the concentrations due to external factors such as slow reactions of internal electrolytes due to passage of electricity. Though these fluctuations did not affect the overall integrity of our experiment because of electrolytes' kinetic stability, there may have been systematic errors in the calculations due to change in concentration of electrolytes. The electric current induces splitting of cationic and anionic part of electrolytes, which might induce further chemical reactions, and, thus, change the concentration of electrolytes (15). Thus, future experiments should use conditions where electrolytes are chemically inert to electricity and electrodes.

Moreover, our experiment was conducted with incandescent light which has its emission spectrum near infrared. For practical purposes, future experiments should be conducted with actual solar light. In our experiment, we had to vary the intensity of incident light, which would not be possible with a natural light source. Furthermore, though we used the standardized methods to prepare the $MoSe_2$ crystals, the purity of the crystals was not examined in the experiment, which may affect the replication of our experiment. This limitation could be addressed in the future experiments by running standard tests to measure crystal purity.

Studies on the relationship between the electrolyte concentration and solar cell efficiency have found that efficiency is maximized when the concentration of I_2 is around 0.03-0.034M (16, 17). Together with these results, we can identify the optimal material, electrolyte concentration, and electrolyte volume for fabrication of PEC solar cells. Although the behavior of PEC solar cell and other solar cells are very similar, we need to repeat the experiment with other solar cells having solid-solid junctions to determine whether the conclusions obtained are applicable to them and are industrially useful. After such tests are made, our results can have profound impact on increasing the efficiency of solar cells. The fuel produced by reactions {1} and {2} might be industrially viable since the only products are oxygen (O_2) and hydrogen (H_2), which are beneficial to environment. The study contributes to developing a carbon-free hydrogen economy (6), as it may improve the efficiency of solar cells, which are used to carry on reactions {1} and {2}.

MATERIALS AND METHODS

The experimental setup was designed so that only the intensity of incident light and volume of electrolyte were variable, and all other factors—including spectral distribution of incident light, concentration of electrolyte, etc.—were maintained to a constant value. Experimental set up consisted of reference electrode, counter electrode, working electrode, electrolyte, voltmeter, and ammeter **(Figure 1)**. A combination of 0.035M I_2 + 0.5M NaI + 0.5M Na_2SO_4 solution was used as the electrolyte. This electrolyte concentration was chosen in accordance with the optimal concentration needed to maximize efficiency of PEC solar cell (13, 17). For experimental purposes, five different values of electrolyte volume (100, 150, 200, 250, and 300 mL) were taken. Copper was chosen as the counter electrode. The reading of the I_{ph} and V_{ph} were taken using digital multimeters **(Figure 1)**. One of the multimeters was kept in μA range and used as the ammeter, while the other one was kept in mV range and used as the voltmeter. The voltmeter was attached between the reference electrode and the working electrode to measure the V_{ph} at a specific volume of electrolyte and intensity of incident light **(Figure 1)**. The margin of error, used to plot the I_{ph} vs. V_{ph} characteristic curve of the PEC solar cell, was obtained using the auto-range function of multimeters.

The experimental setup **(Figure 1)** was illuminated with a polychromatic light using an incandescent lamp. The emission spectrum (obtained by spectrophotometry) of the incandescent lamp included wavelengths from 600 nm to 700 nm, approximately. Intensities of light for which data was taken were 10 to 120 mW/cm^2 with increasing steps of 10 mW/cm^2. The intensity was varied by changing the distance between the incandescent lamp and the working electrode, and not by brightening or attenuating the bulb. This was, in part, to ensure that the emission spectrum of the lamp remained the same throughout the experiment so that it did not interfere with other variable parameters of our experiment. The intensity of the incident light rays was measured using a digital photometer in order to obtain required accuracy in data. The readings of the photometer (measured in candela)

were converted to mW/cm^2 for the sake of graph plotting and data analysis.

$MoSe_2$ was chosen as the working electrode because of several reasons. First, its bandgap (1.55 eV) falls in the maxima of the solar radiation, which is important to get better efficiency (18). It might be noted here that the optimal wavelength range of $MoSe_2$-based electrodes is from 500 nm to 900 nm (18). Furthermore, $MoSe_2$ has high corrosion resistance (due to d → d transitions) which helps in maintaining accuracy of results throughout the experiment (10, 11).

The $MoSe_2$ crystals were grown using the standard direct vapor transfer (DVT) technique in the two-zone metallic furnace (19). DVT was used instead of the chemical vapor transfer technique because DVT produces pure crystals without the use of a catalyst. Molybdenum (lab supply, Parshwamani Metals) and selenium (lab supply, Parshwamani Metals) in the elemental form were taken in 1:2 stoichiometric proportions and filled up in a quartz ampoule. The filled quartz ampoule was evacuated in the vacuum chamber and the pressure was reduced to 1.33×10^{-8} bar. Then the horizontal two-zone furnace was filled with the quartz ampoule and sealed. The front zone is the reaction zone where Mo^{4+} and Se^{2-} react to form $MoSe_2$. The back zone in the dual zone furnace is the growth zone where the product of the reaction, $MoSe_2$, gets deposited. Both zones in the furnace were maintained at 1320-1350 K temperature for the appropriate growth of $MoSe_2$ single crystal. The crystal growth took approximately 5 hours. The $MoSe_2$ crystals were used to fabricate the photo-anode (working electrode, **Figure 1**) of the PEC solar cell. A glass tube with fine bore and a flattened end was used for mounting the crystal. The copper wire was used with ohmic contact with the crystal. The thickness of $MoSe_2$ layer was 380 nm, which closely resembles the optimal thickness (20). The area of $MoSe_2$ crystal was approximately 2 cm^2.

The power output, used to calculate P_{max}, was determined using Equation 4.

$$P_{output} = I_{ph} \times V_{ph} \quad (4)$$

The V_{ph} vs. power output curve was plotted to find the maximum power, P_{max}, required for the calculation of efficiency. To find maxima we first predicted the average curve **(Figure 3)**. Equation 5 represents the formula used to obtain the maxima point. A graphing calculator was used to approximate the curve obtained from Equation 5. Using the average graphical function, we also reduced the possible error arisen from least counts of instruments.

$$P_{output} \equiv P_{max} \Leftrightarrow \frac{dP_{output}}{dV_{ph}} = 0 \quad (5)$$

The efficiency of the PEC solar cell was determined by Equation 6. Efficiency relates maximum power output (P_{max}), intensity of incident light (I), and the area of the cross-section of PEC solar cell which receives incident light.

$$\eta\% = \frac{P_{max}}{I \times Area} \times 100\% \quad (6)$$

Where η% represents percentage efficiency of PEC solar cell. Another factor that we calculated to measure the quality of solar cell was fill factor. Fill factor compares the maximum output power to maximum theoretical yield, which is

$$Maximum\ Theoretical\ Yield = I_{sc} \times V_{oc} \quad (7)$$

Where I_{sc} and V_{oc} are short circuit current and open circuit voltage, respectively. Equation 7 depicts the theoretical maxima of power due to the fact that short circuit current is the value of maximum current obtainable (maximum current flows in a circuit when we do not resist its flow with any resistor, i.e., we allow it to flow freely by joining opposite polarities of the battery with wire), and the open circuit voltage is the maximum obtainable voltage difference (flow of current would theoretically reduce the voltage across any electrical appliance). Using Equation 8, we obtained the value of fill factor.

$$Fill\ factor = \frac{P_{max}}{I_{sc} \times V_{oc}} \quad (8)$$

The One-Way ANOVA test was used in the first experiment to find the statistical significance of the proportionality between efficiency of PEC solar cell and inverse 0.9th power of intensity when 100 mL of electrolyte was used. All calculations and graphs were made in XLSTAT computer application.

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