

Volume holographic gratings in acrylamide-based photopolymer to provide selective light as an added input for improving the performance of dye-sensitized solar cells

A. B. Sreebha¹, S. Suresh¹, C. O. Sreekala² and V. P. Mahadevan Pillai^{1,*}

¹Department of Optoelectronics, University of Kerala, Thiruvananthapuram 695 581, India

²Department of Physics, Amrita Vishwa Vidyapeetham University, Amrithapuri, Kollam 690 525, India

A transmission holographic grating with good storage life and diffraction efficiency is fabricated in an indigenously prepared red-sensitive photopolymer. Stability retained by the photopolymer transmission holographic grating in its diffraction efficiency upon long-term exposure to temperature indicates the suitability of the fabricated photopolymer holograms in solar applications. Holographic elements diffract different wavelengths to different regions and dye sensitized solar cells (DSSCs) can absorb the desired range of wavelengths from the diffracted light. Hence, we exploit this property of the holographic grating to enrich the performance of DSSCs by enhancing its input and output power, particularly under low photometric conditions. Improvement in maximum power output of DSSCs is observed in all cases, with a maximum enhancement of about 100% for the lowest level of input intensity.

Keywords: Diffraction efficiency, dye sensitized solar cells, photopolymer, transmission holographic grating.

SOLAR cell technology is on the cusp of revolution for a shift towards a green energy supply system for the 21st century. Yet, it does not meet more than about 1% of the global energy demand due to the high cost of photovoltaic (PV) cells and inadequate energy conversion efficiency under low irradiance/illuminance conditions¹⁻⁴. Real-time environment under which solar cells operate will be different from standard laboratory conditions and more often, solar cells have to function below peak illuminance under which the output performance of the cell is low⁴. Since solar energy is a low-energy density resource^{4,5}, there is need for photovoltaic systems that can also work under low illuminance/irradiance conditions.

Photovoltaic systems that comprise dye-sensitized solar cells (DSSCs) can work under diffuse and low light conditions⁶. DSSCs which rely on light-absorbing dye molecules are considered as good alternatives to the expensive conventional solar cells. Unlike conventional

silicon-based solar cells, DSSCs do not use toxic chemicals for their manufacture and working, and hence they are environmental-friendly^{6,7}. Furthermore, DSSCs do not need any thermal or vacuum treatments in their production and so prove to be cost-effective^{6,7}. In order to enhance the performance of DSSCs, sensitizing dye should absorb maximum incoming light⁸. A grating element is employed for the purpose of allowing the sensitizer to absorb more light by diffracting selective wavelengths of polychromatic light that falls on it towards the DSSCs^{9,10}. The grating element is fabricated by holographic technology. Using this technology, a grating structure is created in a suitable recording medium by the phenomenon of interference between object wave and reference wave. The record called a hologram when illuminated with the reference wave reconstructs the object wave¹¹. Holographic grating element (HGE) can be fabricated and placed in such a way to bring light of selected wavelengths towards the solar cell and diffract heat radiations away from the cell^{9,10}. Hence the amalgamation of HGE in photovoltaic system will not generate supplementary heat to the solar module.

When too much sunlight does not fall directly on the solar cells, the proposed HGE will direct light towards these cells from its surrounding sides. For example, in the morning and evening when the solar panel at the rooftop is kept flat and there is no tracking, incorporation of HGE can enhance the amount of light falling on a photovoltaic cell. The proposed method does not block direct photon incidence on the cell. Furthermore, HGE directs the otherwise inaccessible portion of light to a photovoltaic cell by utilizing its diffraction property (Figure 1a). HGE can diffract different wavelengths to different regions and DSSCs can absorb the desired range of wavelengths from the diffracted light that match up with the absorbance of the dye incorporated in it. Hence, the proposed scheme based on holographic elements is suitable for better functioning of wavelength-selective photovoltaic cells like DSSCs. In Figure 1a, HGEs are shown only at two peripherals of a DSSC. More such HGEs can be incorporated into the system to enhance the overall power output of DSSC.

*For correspondence. (e-mail: vmpillai9@gmail.com)

As photopolymers have proved to be a highly versatile material for holography, an extensively studied photopolymer material with acrylamide (AA) monomer and polyvinyl alcohol (PVA) base matrix was indigenously prepared in our laboratory for the fabrication of HGEs. Photopolymer-based holographic concentrators are widely exploited for PV applications^{12–19}, where they focus the incident light to PV cells. This article deals with the coupling of holographic element which is not a concentrator. The function of the holographic grating is different from that of a holographic concentrator. HGE does not supply concentrated light to the solar cells. It only directs additional light from the peripherals to the cells. HGE functions by the property of diffraction and hence its performance is evaluated by its diffraction efficiency (DE). To employ the photopolymer-based HGE for better functioning of DSSCs, consistency in its DE at elevated temperatures is necessary.

Studies have already been conducted on the effect of temperature on AA holographic photopolymers^{20–22}. Layer shrinkage is observed upon thermal stress for a photopolymer combination of AA and diacetone acrylamide monomers with two photo-initiators²¹. Increment in polymerization rate of a thermally excited AA monomer has been widely studied^{23–26}. Naydenova *et al.*²⁰ studied the application of AA-based polymer as a relative humidity (RH) indicator at elevated temperatures. They have also reported the stability of the position of peak wavelength of diffracted light upon temperature exposure²⁰. Possibility of chain transfer events²⁴ and increment in the polymer chain termination of AA at high temperatures²⁵ have also been studied.

Experimental section

The photopolymer material fabricated under normal laboratory conditions holds 0.375 molar AA, 0.25 molar

triethanolamine (TEA) initiator and 0.02 molar *N,N'*-methylene-bis-acrylamide (BAA) cross-linker in a 10% transparent PVA solution. The polymer composition is then sensitized with 0.003 molar methylene blue (MB) dye solution in distilled water. A photopolymer film with an optimized dye concentration of 4×10^{-5} molar is then fabricated in PVA/AA matrix by gravity settling method. The films having an average thickness of 200 μm (measured using Bruker's Dektak – TX Stylus thickness profilometer) are fabricated on cleaned glass slides of dimension 75 mm \times 25 mm \times 1.35 mm, and are dried for two weeks.

Well-dried photopolymer samples are taken for fabrication of volume HGEs that reconstruct strongly in the first diffraction order²⁷. HGEs are recorded with a laser radiation of wavelength 632.8 nm from He–Ne laser using standard two-beam transmission geometry²⁷. The recording is done by maintaining the inter-beam angle (angle between object and reference beams) at 27.5°, exposure energy at 100 mJ/cm² and keeping the irradiance of both the beams equal. First-order DEs of the gratings are measured using the standard set-up for such measurements, with the same laser light used for recording holographic gratings²⁷.

HGE thus recorded is coupled with a DSSC fabricated in our laboratory having an area of 0.20 cm². The DSSC consists of mesoporous nanocrystalline titanium dioxide (TiO₂) as photoanode, platinum-coated fluorine-doped tin oxide (FTO) as counter electrode, tri-iodide/iodide redox couple as electrolyte and N719 dye sensitizer.

Light input from an Oriel 3A solar simulator tested with an NREL-calibrated silicon solar cell under the illumination of a 1000 W/m² AM1.5G spectrum is passed through various optical density (OD) filters to create various low photometric (less than AM1.5G) conditions. The input power from the simulator is measured using an intensity meter (Newport model No. 91150 V, S/N: 399/0149). Photopolymer HGE is then placed in the plane of illuminance of the solar simulator in such a way that diffracted light from HGE also falls on the intensity meter without blocking direct illuminance. The power of incident light (direct light + diffracted light from HGE) that falls on the intensity meter is then measured. Figure 2 is a schematic representation of the experiment to study the enhancement in input power by HGE coupling.

In order to find the enhancement in output performance of the DSSC which can occur due to HGE coupling, it is initially placed under a solar simulator (Newport's Class 3A, model 67005) with various reduced photometric conditions (as mentioned before). HGE is then inserted between the DSSC and solar simulator in such a way that diffracted light from it falls on DSSC without blocking the direct photon incidence. The experiment is repeated for various low-input intensity conditions. Figure 3 is a schematic representation of the coupling of HGE with DSSC.

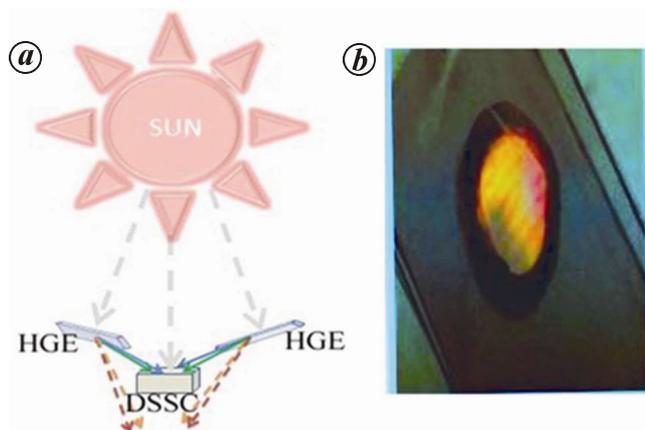


Figure 1. *a*, Coupling of the holographic grating element (HGE) with a solar cell. The selected wavelength components of the solar spectrum are diffracted towards the solar cell, while the undesired wavelength components are diffracted away from it. *b*, Photograph of the HGE recorded in photopolymer material.

Results and discussion

The holographic transmission grating recorded in the fabricated photopolymer exhibits a DE of 80.80%. Once the grating is recorded in the photopolymer, it is immediately subjected to long-term fixing by storing it in dark environment for 48 h. DE of the grating is monitored and a slight reduction from 80.8% to 76.4% is observed during this period. This may be due to the diffusion of unreacted monomers from the region of destructive interference to the region of constructive interference²⁸. The variation in DE of long-term fixed holographic gratings is continuously monitored on a daily basis for a period of six months. The grating element shows a relatively good stability in its DE (Figure 4a) and it is plotted. Even six months after fabrication, photopolymer gratings exhibit a DE of 75%.

When light is incident on the fabricated photopolymer material which is sensitized with MB dye, the photosensitive MB molecules absorb photons to get excited to

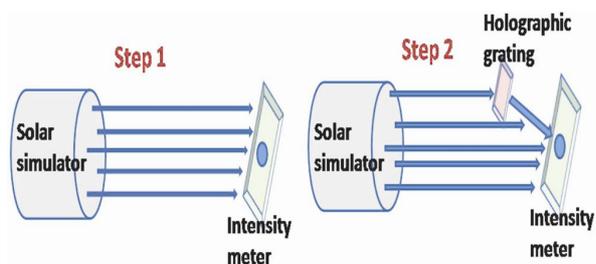


Figure 2. Schematic representation of the experiment to study the enhancement in input intensity by HGE coupling. Step 1: Light from the solar simulator with reduced intensity falls on the intensity meter. Step 2: Light with reduced intensity from the solar simulator together with light diffracted from HGE fall on the intensity meter.

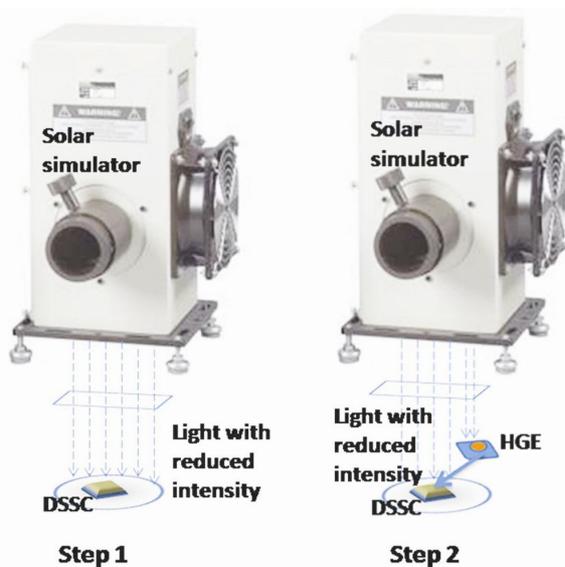


Figure 3. Schematic representation of the experiment to study the enhancement in output characteristics of DSSC when it is coupled with HGE. Step 1: Light from the solar simulator falls on the DSSC. Step 2: Light from the solar simulator together with light diffracted from HGE fall on the DSSC.

singlet-state MB* (refs 29–31). These singlet-state dye molecules can undergo inter-system crossing to get transferred to the more stable triplet state ³MB* (refs 29–31). The dye in the triple excited state can lose hydrogen atoms to convert to leuco form. Constancy achieved in the DE of holographic gratings recorded in the fabricated photopolymer may be due to stability of the leuco form³². Here, the MB dye molecule in fabricated photopolymer can get trapped inside the photopolymer material which is cross-linked with BAA to attain stability of the leuco form. Thus, stability in DE is observed when HGEs are stored in normal laboratory conditions.

To evaluate the DE of HGE at elevated temperature, long-term fixed photopolymer holographic gratings are subjected to a temperature of 70°C by mounting a glass slide identical to substrate on the photopolymer slide to prevent peeling of polymer layer from the glass substrate. This sandwiched assembly is thermally exposed for different time-durations (1–60 min). DE of the thermally exposed photopolymer gratings is measured once it is cooled naturally to room temperature. Figure 4b shows the influence of temperature exposure on DE of the grating. After 1 min of temperature exposure, DE of the grating is reduced from 76.40% to 70.10% and after 5 min, it is further reduced to 64%. At an exposure period of

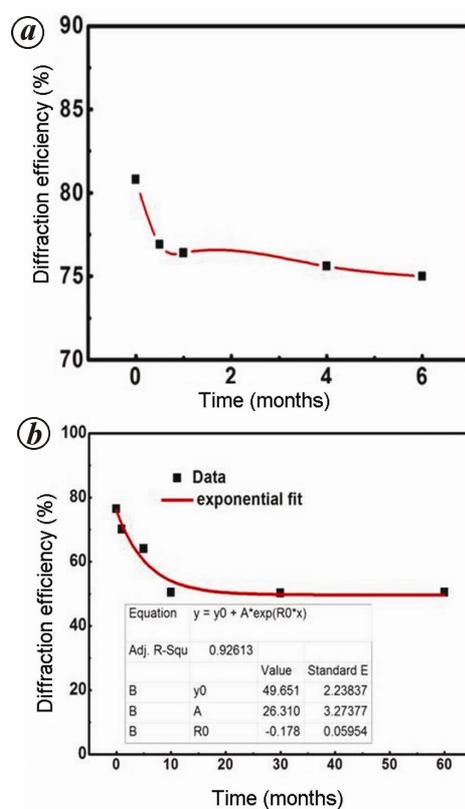


Figure 4. a, Variation of diffraction efficiency with ageing of the photopolymer holographic grating. b, Variation of diffraction efficiency with time for the photopolymer holographic grating subjected to a temperature of 70°C.

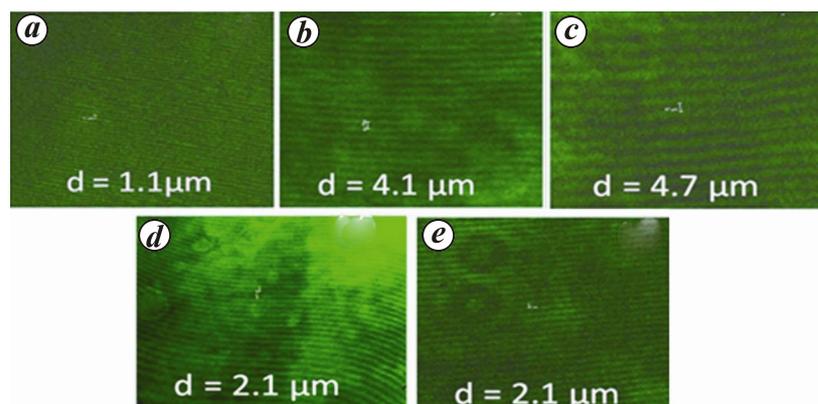


Figure 5 a–e. Confocal microscopic images of interference fringes formed in HGE exposed to a temperature of 70°C for 0, 1, 5, 10 and 60 min respectively.

Table 1. Holographic grating element parameters upon thermal exposure at 70°C

Time duration (min)	Average thickness of photopolymer, D (μm)	Interference fringe spacing, d (μm)	Diffraction efficiency (%)
0	200	1.10	76.40
1	240	4.10	70.10
5	240	4.70	64.00
10	150	2.10	50.40
30	150	2.10	50.20
60	150	2.10	50.40

10 min, DE is reduced to around 50%. The variation in DE of HGE with thermal exposure time shows an exponential decrease (Figure 4 b).

It is observed that the blue colour of the photopolymer film is changed to yellowish-orange when the duration of thermal exposure is 10 min or more. However, DE of the grating remains almost constant at around 50%. This stability of DE of photopolymer holographic grating upon long time thermal exposure may be regarded as an indication of the suitability of the fabricated photopolymer to be used in conjunction with DSSC. Since initial thermal exposure results in increase in fringe width, the Bragg replay angle also gets shifted. Hence, HGE is slightly tilted while measuring DE to find the maximum value.

The effect of temperature on fringe width and thickness of holographic gratings is analysed. The spacing of interference fringes of holographic gratings upon thermal exposure is measured using typical 2D confocal microscopic images (CMIs) (Figure 5 a–e).

Before subjecting the grating to thermal exposure, the fringe spacing measured is 1.1 μm and it increases to 4.7 μm when the photopolymer grating is subjected to 5 min thermal exposure. Thermal exposure should result in increase in fringe width. However, the observed increase in fringe width is higher than that expected. As the thickness of the photopolymer material is rather high, enhancement in its volume effects upon thermal exposure may occur³³. The enhanced volume effects could be the reason for this large increase in fringe width. Fringe spacing decreases to 2.1 μm when the photopolymer grating is

thermally exposed to 70°C for 10 min. Further increase in the extent of thermal exposure does not change the fringe spacing.

The average thickness (D) of the photopolymer grating subjected to thermal exposure is measured. Before thermal exposure, D is 200 μm . It increases to about 240 μm when the extent of thermal exposure is enhanced from 1 to 5 min. This initial increment in the thickness of photopolymer grating upon temperature exposure is due to the bulging of photopolymer material with temperature³⁴. Further enhancement in the duration of thermal exposure reduces the average thickness of the photopolymer grating to about 150 μm . This reduction in thickness with long-term thermal exposure may be due to the formation of a thermal cross-link upon temperature exposure. With thermal exposure, a thermal cross-link may develop³⁵ and as exposure time increases, the cross-linked structure may become so rigid to make further enlargement impossible. Hence, average thickness of the photopolymer grating maintains a constant value when the extent of thermal exposure is 10 min or more.

Table 1 shows the interference fringe spacing, average thickness and DE of the fabricated photopolymer grating for different thermal exposure times. It is obvious that the variation in thickness of the photopolymer is reflected in the variation of fringe spacing of the grating fabricated on the material³⁴. The combined effect of change in thickness along with the change in fringe width of the grating can have a consequent impact on the performance of gratings^{34,36}. When subjected to a thermal exposure of 10 min

Table 2. Comparison of input intensity before and after coupling with HGE

Optical density of filter	Power before inserting HGE (mW/cm ²)	Power after inserting HGE (mW/cm ²)	Percentage enhancement in input power
0.1	76.58	76.86	0.37
0.2	61.69	62.30	0.99
0.5	32.91	33.50	1.79
0.6	26.70	27.35	2.43
1.0	12.65	13.15	3.95
3.0	1.35	1.70	25.92
4.0	1.12	2.36	110.71

Table 3. Dye-sensitized solar cell parameters before and after insertion of HGE

Insertion of HGE	Input power (mW/cm ²)	Open circuit voltage (V)	Short circuit current density (mA/cm ²)	Maximum power (mW)	Enhancement in P_{\max} (%)
Before	1.12	0.466	0.205	0.009	100
After	2.36	0.517	0.396	0.018	
Before	12.65	0.553	0.523	0.022	59.1
After	13.15	0.573	0.642	0.035	
Before	26.70	0.584	0.908	0.053	50.90
After	27.35	0.634	1.479	0.080	
Before	61.69	0.661	3.661	0.273	22.34
After	62.30	0.666	4.665	0.334	

or more, the photopolymer grating attains stability in DE, fringe width and thickness. This is an important observation.

Thus, the prepared photopolymer holographic grating is stable under ambient conditions for long periods and retains stability in DE, fringe width and thickness upon long-term thermal exposure. As the above results indicate the suitability of the fabricated photopolymer for better functioning of DSSC, HGE recorded in the fabricated photopolymer is coupled with DSSC which is sensitized with dye N719 that mostly absorbs light in the low wavelength portion of the visible spectrum³⁷.

Since the absorption band of N719 dye incorporated with DSSC falls within the blue to green wavelength range of visible spectrum³⁷, mostly the diffracted blue/green components are allowed to fall on the intensity meter and DSSC to find the enhancement in input and output power respectively, without blocking the direct photon incidence. Enhancement in the input power is found in all cases of HGE coupling (Table 2). Also, it can be observed that the enhancement is higher for low photometric conditions. This enhancement in input power can cause corresponding enhancement in output characteristics of the solar cell to which HGE is coupled.

Open circuit voltage (V_{OC}), short circuit current density (J_{SC}), maximum power (P_{\max}), etc. are some of the key parameters to determine the output performance of any solar cell³⁸. The maximum voltage available from a solar cell is V_{OC} and the maximum current density is J_{SC} (ref. 39). There is a position in between V_{OC} and J_{SC} where the solar cell produces maximum power. This is the maximum power point P_{\max} . P_{\max} is the location around which the ideal function of a solar cell occurs³⁸ and it is con-

nected to the load. Hence, any system which can enhance P_{\max} of a solar cell enriches the overall performance of the cell. The integration of photopolymer HGE in the DSSC module enhances V_{OC} , J_{SC} and P_{\max} of DSSC (Table 3). It is also observed that the percentage enhancement of all these parameters is high for very low input intensity conditions. Hence the integration of HGE with DSSC is effective under very low intensity conditions. By incorporating more number of HGEs in the region roughly surrounding a single DSSC but without blocking the direct solar incidence towards the solar cell, it is possible to markedly enhance the output performance of the DSSC module under each input intensity condition.

Conclusion

A red-sensitive photopolymer holographic grating element with good diffraction efficiency and storage life is fabricated and coupled with a green-sensitive DSSC. The coupling done under various low-intensity conditions shows the improvement of output parameters of DSSC in all cases. This result shows the possibility of using holographic gratings to enhance the performance of solar cells even when they work in low photometric conditions. The stability in diffraction efficiency of photopolymer HGE under long-term temperature exposure indicates its suitability for solar applications.

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