ZnO Films Deposited Using the Ultrasonic-Assisted Successive Ionic Layer Adsorption and Reaction Method for Dye Sensitized Solar Cell Applications

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Abstract

Zinc Oxide (ZnO) photoanode has attracted great attention due to its favorable electronic and optical properties. The deposition of ZnO films using the ultrasonic-assisted successive ionic layer adsorption and reaction (SILAR) method has been done successfully on fluorine-doped tin oxide and glass substrates at various dipping cycles. The structural, morphological, and optical properties as well as the dye sensitized solar cell (DSSC) performance of the ZnO photoanodes were investigated. The ultrasonication process is used to enhance the compactness, crystallinity, shape, and size of the prepared ZnO films. The fabricated DSSC device with 150 SILAR cycles shows the highest efficiency of 0.78% with a $J_{sc}$ of 4.56 mA/cm$^2$, $V_{oc}$ of 0.45 V, and FF of 0.38. The joint employment of the SILAR-dipping and ultrasonication process provided an effective way to prepare excellent photoanodes for DSSC application.

Keywords: DSSC, N3 Dye, UA-SILAR, ZnO.

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1. Introduction

Recently, the universe has faced one of the most persistent issues: an energy crisis. Fossil fuels are being exhausted continuously. Using fossil fuels contributes to global warming, air pollution, and greenhouse gases. Finding clean energy resources and alternatives to nonrenewable energy sources is now becoming a top priority in research.11 Sunlight is the most copious renewable energy source on earth. However, proper utilization of solar energy with financial benefits is a great challenge for researchers. Solar energy can be extracted by a photovoltaic (PV) system. PV systems deal with the alteration of light energy into electrical energy on the basis of the photovoltaic effect. To date, various generations of PV cells are present: a) First generation includes Si wafer-based solar cells; b) Second generation includes CIGS, CdTe, and GaAs; and c) Third generation includes organic solar cells, quantum dot solar cells, DSSC, and perovskite solar cells.23 DSSCs are third-generation solar cell with a low cost. Currently, DSSCs with nanocrystalline mesoporous metal oxide films are receiving more attention.10 Because of its low price and easy assembly technique, DSSC has a remarkable impact. DSSC’s working principle is similar to the natural photosynthesis principle.44 The efficiency of DSSC was reported to be approximately >11%. DSSCs are composed of four main parts: 1) photoanode (semiconducting metal oxide), 2) electrolyte, 3) dye (photosensitizer), and 4) counter electrode. The efficiency of DSSCs is hampered because of the dark current produced because of charge recombination at different interfaces. Metal oxides such as TiO$_2$ and ZnO are semiconducting materials. In the context of photovoltaic cells, TiO$_2$ and ZnO are used as electron transport layers. When sunlight is absorbed by the cell, electrons are excited and can move through the TiO$_2$ or ZnO layer to reach the other side of the cell, where they can be collected to generate electricity.13 The photoanode is one of the main parts of the DSSC device. It plays a vital role in charge transfer. Nanocrystalline semiconducting materials with wide bandgaps, such as ZrO$_2$, ...
ZnO and TiO$_2$ etc. are used as photoanodes. Photoanodes are anchored by dye molecules, and they provide a platform to absorb dye on their surface. In comparison to TiO$_2$, a material used to synthesize photoanodes, ZnO nanomaterials have high electron mobility. High electron mobility causes fast charge transport, which generates power. Therefore, dye-loaded ZnO photoanodes for DSSC applications need to be investigated. The synthesis of ZnO nanostructures is more energy effective and lucrative because of its slow growth process, but it has higher electron mobility and lifetime. Here, we deposited a ZnO films and used this film as photoelectrode to assemble DSSC solar cells. A cost-effective wet chemical method, i.e., Successive Ionic Layer Adsorption and Reaction (SILAR) was used to synthesize ZnO films on Glass and FTO substrate.

The next important part of DSSCs is the photosensitizer (dye). Photosensitizers imbibe a major role in absorbing light and producing photoexcited electrons at the semiconductor interface. A variety of dyes are used as photosensitizers, such as metal complex dyes or natural dyes. Natural dyes have a nontoxic and environmentally friendly nature, and they are inexpensive and simply accessible. Natural dyes exhibit low efficiency in comparison with artificial dyes. Chlorophyll, betalain, and anthocyanin pigment-based dyes are reported to be the best natural photosensitizers. Metal complex dyes show the highest efficiency, but they also have some drawbacks. These include high cost, toxicity, degradation in water contact, carcinogenic tendencies, etc.

In the current efforts, the UA-SILAR method deposited ZnO photoanode, N3 dye as a photosensitizer and platinum counter electrode with polyiodide electrolyte was used to fabricate DSSC solar cells. Solar simulator with a Keithley source meter is used to calculate power conversion efficiency (PCE) calculations of fabricated devices.

### 2. Experimentation:

#### 2.1 Material

In this study as purchased analytical grade chemical were used for experiment. This includes an extra pure ZnSO$_4$·7H$_2$O (Sisco Research Lab), FTO substrate (Sigma Aldrich, USA) Ethanol (Changshu Hongsheng Free Chemical Co. Ltd.), liquid ammonia (Specific gravity of 0.91 and approximately 25% NH$_3$), and N3 dye (Sigma Aldrich).

#### 2.2 Methodology

##### 2.2.1 Preparation of precursor sources

- **Anionic precursor:** Deionized water was used as an anionic precursor.
- **Cationic precursor:** 0.05 M ZnSO$_4$ was used as a cationic precursor. Ammonia solution (approximately 25% NH$_3$) is used as complexing agent added to the ZnSO$_4$ solution at 300K. Ammonia was supplemented dropwise with stirring, until a white gelatinous precipitate of zinc hydroxide is dissolved and the pH of the solution became ~12. This solution is then used as a cationic precursor.

##### 2.2.2 Deposition of photoanode

Zinc oxide films were grown using the UA-SILAR method over the conducting surface of FTO. First, the conducting substrate is properly washed using ethanol. The SILAR method includes alternative dipping of the FTO into the precursor sources. Here, we use the three-beaker system for the UA-SILAR method. The adsorption of the zinc complex takes place on the cleaned FTO when immersed in a cationic source solution. Subsequently, the FTO was immersed in an anionic precursor, where reaction of the complex takes place with deionized water. Finally, the substrate was subjected to rinsing with deionized water to eliminate loosely attached ions. For deposition, the adsorption time, reaction time, and rinsing time were 20, 15, and 30 sec, respectively. Such cycles were repeated several times, and ZnO films of 50, 100, 150 and 200 cycles with thicknesses such as 1.11, 1.77, 2.11 and 1.55 μm respectively were deposited. The Schematic arrangement for this mode is displayed in Fig. 1. Annealing of as deposited films was carried out at 250 °C for 2 hours in an air furnace. Annealing converts the zinc hydroxide phase of the material to the oxide phase and enhances crystallinity.

![Fig. 1 Schematic setup of the UA-SILAR method.](image-url)

##### 2.2.3 Film Formation and Reaction Mechanism

The UA-SILAR method includes dipping the substrate alternatively in cationic and anionic bath solutions. A cationic source composed of tetra ammonium Zn (II) complex [Zn(NH$_3$)$_4$]$^{2+}$ formed by adding a small amount of excess ammonia. Here, the twofold role of ammonia is to control the pH of the solution to ~12 and as a complexing agent. When we added ammonia dropwise to ZnSO$_4$ solution, initially white-colored gelatinous precipitate of Zn(OH)$_3$ was obtained. This precipitate disappeared by adding a small excess amount of ammonia. Thus, ammonia helps to avoid the precipitation of Zn(OH)$_2$ in a cationic solution. Anionic sources contain deionized water. The temperature of the anionic source was sustained at approximately 80 °C in UA-SILAR method. In complete one cycle of the UA-SILAR method, various reactions take place at different intervals as shown by Equation (1) to Equation (4). Zinc complex gets adsorbed on the surface of the substrate when dipped in a cationic precursor bath, and the zinc complex is then converted to zinc hydroxide during a reaction in an anionic solution. Finally, at annealing,
zinc hydroxide is converted to ZnO. Annealing also improves film crystallinity, and we obtained a compact ZnO film on the FTO substrate.

\[ \text{ZnSO}_4 + 2\text{NH}_3 \cdot \text{H}_2\text{O} \rightarrow \text{Zn(OH)}_2 \downarrow + (\text{NH}_2)_2\text{SO}_4 \]  

(1)  

\[ \text{Zn(OH)}_2 + 4\text{NH}_3 \cdot \text{H}_2\text{O} \rightarrow \text{Zn(NH}_3)_4^{2+} + 2\text{OH}^- + 4\text{H}_2\text{O} \]  

(2)  

\[ [\text{Zn(NH}_3)_4]^{2+} + 4\text{H}_2\text{O} \rightarrow \text{Zn(OH)}_2 \downarrow + 4\text{NH}_4^+ + 2\text{OH}^- \]  

(3)  

\[ \text{Zn(OH)}_2 \rightarrow \text{ZnO} + \text{H}_2\text{O} \]  

(4)

2.2.4 Ultrasonic cavitation effect on film formation

Ultrasonication helps in well dispersion of particles and it also reduces particle size by implosion effect which makes film more crystalline and compact. The major impact of ultrasonication treatment is a uniform deposition and even distribution of particles. In ultrasonication, acoustic cavitation effect produce extremely high uniform transient pressure. This uniform transient pressure disperses the particle and relaxes the crystal structure.[14] Also, it avoids agglomeration and accumulation of particle which form compact films and increase adherence of films. Sonication treatment causes inhibition of the growth of ZnO grains which results in the attachment of nanoparticles over the entire surface and forms homogeneous films without agglomeration of nanoparticles. Generally, specific surface area is associated with particle size. The smaller the size of the particle, the larger is specific surface area. The ultrasonic cavitation effect contributes to avoid agglomeration and forming small-sized particles which reduces crystallite size and increases surface area which is in good agreement with XRD results. In this deposition process, the temperature of the anionic source was sustained at approximately 80°C which helps to dissociate water to release hydroxyl ions (OH\(^-\).[11]

2.2.5 Device fabrication

A DSSC device was fabricated by assembling a photoanode, photosensitizer, electrolyte, and counter electrode into a sandwich-like structure. The platinum counter electrode was then mounted over surface of the dye-loaded photoanode. Iodide/triiodide liquid electrolyte was injected between them which formed a sand-witch like structure.

2.3 Material Characterization

A UV–visible spectrophotometer (JASCO·V-670, Germany) was utilized to understand the optical absorption characteristics of the ZnO film photoanode. Additionally, the bandgap of the photoanode was calculated. To study the surface morphology of the as-prepared ZnO films, scanning electron microscopy (SEM) (JEOL-JSM 6360-A USA) was used. Photoluminescence spectroscopy: Horiba Fluorolog-QM with an emission range with a standard PMT of 185 nm to 900 nm was used to measure the bandgap energy, presence of impurities, and structural defects in the material. Raman spectroscopy was studied by using a Renishaw model with a spectral resolution of 0.5 cm\(^{-1}\), Raman spectral range of 100 - 4000 cm\(^{-1}\), and spectrometer range of 200-2000 nm, and a multimodal argon ion laser was used to study the chemical structure, phase, and crystallinity of material. Photovoltaic parameters of the fabricated device were calculated with the help of a Solar Stimulator (Oriel sol-2A, Newport, USA). An AM1.5 sunlight filter with illumination intensities of 50 mW/cm\(^2\) and 33 mW/cm\(^2\) LED (Nevis Clean Tech, India) was used to measure the photovoltaic parameters of the fabricated device. A Keithley source meter 2420 (Keithley Instruments Ltd., USA) was used to measure the J-V characteristics.

3. Results and discussion

3.1. Thickness variation analysis

To analyze the variation in thickness, ZnO film was deposited at various cycles for UA-SILAR method. SILAR cycles were repeated 50, 100, 150, and 200 times, and the thickness per variation was measured (see Fig. 2). All these cycles for the ultrasonication mode are denoted as ZnO-S50, ZnO-S100, ZnO-S150, and ZnO-S200. Deposited ZnO films appear to be white in color and adhere tightly to the substrate surface. It was found that films remained stable under environmental conditions.[12,13] The calculated thicknesses of the deposited ZnO films were approximately 1.11, 1.77, 2.11, and 1.55 μm for 50, 100, 150, and 200 cycles, respectively. A trivial reduction in the width of the zinc oxide film was detected after 150 cycles. This reduction in the thickness of ZnO films may be due to increased stress, which causes peeling off of the film after the optimum thickness. The temperature of the anionic precursor in ultrasonication mode was maintained at approximately 80°C. Deionized water was used as an anionic precursor. Temperature helps to break attractive forces and bonding present between ions of the anionic source and become adsorbed on FTO.[14,15]

![Fig. 2 Thickness variation of prepared ZnO films at various UA-SILAR deposition cycles.](chart.png)
spectra along with \((ahv)^2\) versus light energy \((hν)\) plots of pure zinc oxide deposited using the UA-SILAR method are presented below in Figs. 3(A) and 3(B), respectively. Pure ZnO crystals show a wurtzite structure with a 3.33 eV direct bandgap.\[16] From this ZnO film’s UV-Visible spectrum, it can be concluded that due to the wide bandgap, ZnO shows low absorbance in the visible region. ZnO shows maximum absorption at 390 nm. The band gap value was determined by analyzing absorption data using Tauc’s equation. Tauc’s equation demonstrates the conventional relation between light energy \((hν)\) and absorption coefficient \((α)\). The absorption coefficient measures how deep the light of a particular wavelength penetrates the substance before it is absorbed into it.\[17] The band gap values for ZnO films deposited at various cycles were calculated to be approximately 3.24, 3.23, 3.19, and 3.22 eV for the ZnO-S50, ZnO-S100, ZnO-S150, and ZnO-S200 samples, respectively. The optical absorption edge shows a slight shift toward a higher wavelength with an increase in the count of UA-SILAR repetitions, which might occur because of the scattering due to grain boundaries. This shift shows a decrease in the bandgap, which confirms thickness enhancement and growth in grain size with increasing UA-SILAR cycles.\[18,19] From Fig. 3B, it is observed that a slight reduction in the band gap value with a surge in film thickness. The surge in band gap energy may be because of a reduction in lattice deformation of the film during the growth of ZnO. The second reason for the increment in band gap energy may be because of the potential periodicity of the crystal lattice of ZnO, which is created by electrons in the ZnO metal oxide semiconductor.\[20,21] The slight diminution in the band gap of ZnO films may be caused by oxygen vacancies, as studied by Ansari et al.\[22] Here, they reported that when ZnO is grown in water, oxygen vacancies increase, which causes a decline in the band gap of zinc oxide.

3.3 X-ray diffraction study
The XRD spectra of ZnO films deposited by the UA-SILAR method are denoted as ZnO-S50, ZnO-S100, ZnO-S150, and ZnO-S200 in Fig. 4. From the XRD data of deposited films, the polycrystallinity of ZnO films was reflected. The observed XRD peaks at approximately 20 values of 32°, 35°, and 36° confirm the diffractions at (100), (002), and (101) planes, respectively which shows the formation of the wurtzite (JCPDS No. 030752) crystal structure of the zinc oxide material.\[18,19] The relative crowning intensity of planes (100), (002), and (101) increased with increasing film thickness. The 20 values for the (002) crystallographic plane of three deposited ZnO thin films for samples ZnO-S100, ZnO-S150, and ZnO-S200 are larger compared to the other hkl planes, which implies that the prepared thin films are exposed to compressive strain in the c-axis direction.\[23,24] Here, samples deposited for 150 cycles show the highest diffraction peak intensity. This result indicates that the enhancement in ZnO film crystallinity was due to SILAR repetitions. This improved crystallinity of the films can be assigned to the atom rearrangement and defect removal because of stratified deposition in the UA-SILAR method. Crystal analysis of deposited ZnO was performed by the Debye-Scherrer formula by using Equation (5).\[25]

\[
D = \frac{k \lambda}{\beta \cos \theta}
\]

where \(D = \text{crystallite size (nm)}, \ K = 0.9 \text{ (Scherrer constant)}, \ \lambda = 0.15406 \text{ nm (wavelength of the X-ray source)}, \ \beta = \text{full-width at half maximum (FWHM)}, \ \text{and} \ \theta = \text{peak position/diffraction angle.}
\]

The average crystallite sizes corresponding to major peaks (100), (002), (101) of the deposited ZnO films are approximately 30, 28, 27, and 27 nm for 50, 100, 150, and 200 cycles, respectively. Large crystal sizes of material provide long pathways for the movement of electrons at the surface of the material. This long pathway supports active charge transport in DSSC devices.
3.4 Raman spectroscopic study

Raman spectroscopy is one characterization methodology for the examination of the purity and phase of nanostructure materials. It provides information on the vibrational characteristics of ZnO nanocrystals. Raman scattering is very subtle to the nanocrystalline material. The standards of the ZnO film material were investigated by Raman scattering. Hexagonal wurtzite crystal structure of ZnO corresponding to the (C\textsubscript{6}V\textsubscript{3}) space group. The optical mode at the Γ point of the Brillouin zone was adopted by group theory\textsuperscript{[26]} and represented by the Equation (6).

\[
\Gamma_{(opt)} = 1A_1 + 2B_1 + 1E_1 + 2E_2
\]

The E\textsubscript{1} and A\textsubscript{1} branches show Raman and IR active modes. Two E\textsubscript{2} branches are only Raman active, and two B\textsubscript{1} modes are Raman and IR inactive. For the ZnO unit cell, twelve vibrational modes exist. Among these active modes, E\textsubscript{1} and A\textsubscript{1} are further separated as Longitudinal Optical phonons (LO) and Transverse Optical phonons (TO). And E\textsubscript{2} mode is again torn apart into two diverse frequencies: E\textsubscript{2} (low) associated with the Zn-sublattice and E\textsubscript{2} (high) accompanied by an oxygen atom. Raman spectra of ZnO film deposited by UA-SILAR method at various cycles are illustrated in Fig. 5. All these samples show an eminent peak at 438 cm\textsuperscript{−1}, which is endorsed to the E\textsubscript{2}(high) active mode, is linked with the oxygen atom, and is characteristic of the nonpolar, optical phonon, hexagonal phase of ZnO materials.\textsuperscript{[27]} This is attributed to structural defects due to oxygen vacancies. At 200 SILAR cycles, peak intensity was found to be the highest. This is assigned to the increased crystallinity of ZnO and coverage of all surfaces of the substrate.\textsuperscript{[28]}

3.5 Scanning Electron Microscopy (SEM)

Figure 6 displays, SEM and Image J software images of all ZnO samples deposited by the UA-SILAR method. The consequence of the variation in UA-SILAR cycles on the morphology of the deposited films is seen in terms of the orientation and directionality of ZnO microrods. As the number of cycles increases, almost the entire substrate surface area is covered by ZnO particles. For ZnO deposited by ultrasonication, in the ZnO-S50 sample, several voids are seen. Here, the grown microrod shows a small inclination and compact aggregation observed at a few places. Relatively fewer voids are observed in ZnO-S100 cycles. Needle-like microrods with nearly equal lengths grow randomly and cover almost all the surface area of the substrate. For the ZnO-S150 sample, uniform and homogeneous microrods with an increased diameter are observed. Except for a few, most of the microrods grow with inclination. A well-aligned, and uniformly grown rod structure is seen in the case of the ZnO-S200 sample. Micro rods grew abruptly on the surface of the substrate, densely covered all surfaces of the substrate, and showed granular morphology. Here, we use an ultrasonic-assisted anionic precursor that enhances the homogeneity of the ZnO film. Ultrasonication alters the deposition methodology and endorses the superiority of the deposited film by the cavitation effect.\textsuperscript{[29]} A vertical view image of each sample is displayed in the inset of each SEM image to measure the thickness of the samples. From these optical microscopic images, the estimated thickness for sample ZnO-S50 was 1.11 μm, for ZnO-S100 it was 1.77 μm, for sample ZnO-S150 it was 2.11 μm and for Sample ZnO-S200 it was 1.55 μm. The average diameter of particles ranges from 200 nm to 500 nm as calculated from ImageJ software. Images b, d, f, and h in Fig. 6 gives the surface topography of deposited films. These images reflect that the surface of deposited films are porous which help in the absorption of dye molecules. From these thickness values, it can be concluded that film thickness enhancement was observed with an escalation in the count of UA-SILAR repetitions.
3.6 Photoluminescence spectroscopic study
Photoluminescence spectroscopy is an adaptable technique for speculating photoactive centers in a material. PL offers valuable insights into the charge carrier recombination, defect traps, optical quality of materials, and lattice defects present. Fig. 7 represents the PL spectra of the prepared zinc oxide film. All the samples of different cycles show a peak at the same wavelength in the PL spectra. It is visualized from the PL spectra of all ZnO films that all samples exhibit luminescence centers at 381, 473 and 569 cm\(^{-1}\).[30] A strong and sharp near UV emission peak occurs at 381 cm\(^{-1}\). It is assigned to near-band edge emission (NBE). It has resulted from the recombination of the charge carrier.

The peaks at 473 and 569 cm\(^{-1}\) are because of the existence of structural imperfections such as Zn interstitials, oxygen impurities, and some free impurity atoms. For example, oxygen and Zn interstitials are present due to the presence of free impurity atoms.[32–34] Here, it is interesting to note that with an escalation in the count of repetitions, PL intensity also increases. That is, as the number of SILAR repetitions rises, the film thickness is enhanced, which causes an increase in charge carriers, eventually increasing the PL intensity.

3.7 Current-voltage (I-V) measurements
Photovoltaic properties of the fabricated DSSC were also calculated with the help of a solar simulator. DSSC device fabricated by ZnO film deposited by UA-SILAR method, platinum counter electrode, polyiodide electrolyte, and N3 dye. Various solar cell parameters, i.e., \(V_{OC}\), \(I_{SC}\), FF, and PCE were determined and reported in Table 1, shown below, along with a literature survey of the performance efficiency of DSSCs. Here is the study of the execution efficiency of the fabricated device by dye loading for 4 hr at room temperature on a ZnO photoanode deposited at various cycles. A study of the

Fig. 6 SEM and Image J software images of ZnO films prepared via the UA-SILAR method (a) & (b) at 50 cycles, (c) & (d) at 100 cycles, (e) & (f) at 150 cycles, and (g) & (h) at 200 cycles.
### Table 1. Literature survey of the performance efficiency of DSSC solar cells along with present work.

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<th>Sr. No.</th>
<th>Photo-anode</th>
<th>Method</th>
<th>Dye and loading time</th>
<th>Jsc (mA/cm²)</th>
<th>Voc (V)</th>
<th>FF</th>
<th>η (%)</th>
<th>References</th>
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<td>0.44</td>
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**Fig. 7** PL spectra of ZnO films prepared via the UA-SILAR method.

**Fig. 8** I-V curves of ZnO films prepared via the UA-SILAR method.

thickness variation of ZnO films for DSSC solar cells was performed. Fig. 8 illustrates the I-V graphs of deposited ZnO films at different UA-SILAR cycles, such as 50, 100, 150, and 200 cycles. The I-V curve under illumination displays a rise in current density with film thickness. An enhanced thickness of ZnO films will help to improve the interaction possibilities between the semiconductor-electrolyte junction and charge carriers. Improved junctions probably help in harvesting higher numbers of charge carriers. With the increase in the film thickness, the effective band gap of the material decreases because of the absorption of long wavelength photons. Here, the current density increases with the increasing thickness of the material, and Voc decreases as the rate of charge carrier recombination increases.

**4. Conclusion**

DSSC fabricated by ZnO photoanode deposited by UA-SILAR method, platinum counter electrode, polyiodide electrolyte and N3 dye has been performed successfully. Preparation of ZnO films by the UA-SILAR method at various cycles was performed, and these films were further used as photoanodes for DSSC device fabrication. The anionic precursor was ultrasonicated to enhance the compactness, shape and size of the prepared ZnO films. It has been justified that the cavitation effect of ultrasonication enhances the morphology and crystallinity and causes uniform deposition of ZnO films. The morphology of deposited ZnO microstructure also has a direct influence on DSSC activity. Film thickness also increases with the rise in the repetitions of
SILAR cycles, which offers a large surface area for dye absorption. The DSSC device fabricated by the ZnO-S150 sample for 4 hr. of dye loading shows the highest efficiency of 0.78% with a $J_{SC}$ of 4.56 mA/cm², $V_{OC}$ of 0.45 V, and FF of 0.38.

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Conflict of Interest
There is no conflict of interest.

Supporting Information
Not applicable.

References


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