Graphene and its derived compounds such as graphene oxide (GO) and reduced graphene oxide (rGO) are characterized by a single layer of sp²-bonded carbon atoms that provide exceptional electronic, thermal, optical and mechanical properties, thus allowing their application in a wide range of functionalities including flexible displays, photovoltaics, catalysis, smart packaging, high-frequency devices, biosensors, and optoelectronics. Both GO and rGO contain oxygen functional groups on the graphene sheets, but at different oxygenation percentages, which means that it is possible to print any design and size of the desired pattern. In addition, rGO has been considered an interesting alternative material for the fabrication of counter electrodes in DSSCs, mainly because it is based on an abundant and low-cost material. However, there are very few reports in the literature on the preparation of rGO films using the inkjet printing technique. In this work, we have prepared an environmentally friendly rGO dispersion (ink) by means of ultrasound-assisted liquid exfoliation, using isopropanol as a solvent and PVP as a stabilizer. The optimization of inkjet-printing parameters, including the number of printed layers, resulted in the reproducible fabrication of homogeneous rGO thin films with a surface coverage of 1 cm². The rGO films were thermally treated at different temperatures in order to determine the influence of the removal of PVP on the film properties. Inkjet-printed rGO films on FTO with high optical transparency were incorporated as counter electrodes in DSSCs with the [Co(bpy)₃]²⁺/³⁺ redox couple in acetonitrile. The rGO films annealed at 400°C showed excellent electrocatalytic activity toward the reduction of [Co(bpy)₃]²⁺/³⁺ and were effectively integrated as counter electrodes in a TiO₂-DSSC.
10 mL isopropyl alcohol (IPA) (Sigma-Aldrich, anhydrous, 99.5%). Subsequently, the exfoliation process was carried out for 4, 8 and 12 hours using a bath sonicator (Branson 3800). The dispersions were centrifuged at 4000 rpm for 1 hour (Thermo Scientific, Sorvall ST 8), and the supernatant was collected and characterized. The particle size distribution and zeta-potential of the dispersions were evaluated by dynamic light scattering (DLS) using a Zetasizer Nano ZS 3600. The thermal behavior of the optimized rGO ink, rGO powder and PVP was examined by thermogravimetric analysis (TGA) using a TA Instruments-Discovery equipment with a heating rate of 5°C/min from 25 to 1000°C under air flow at 25 mL/min. UV-Vis spectrophotometry analysis (Agilent Instruments) was carried out using 1 mL of the optimal rGO ink in the range of 200–1000 nm.

### Inkingjet printing and characterization of rGO films.

— Before printing, FTO (fluorine-doped tin oxide) TEC-15 substrates were cleaned using 15 mL piranha solution for 5 minutes. Substrates were then rinsed with deionized water and dried at 80°C for 15 minutes. The printing process was carried out by a drop-on-demand Dimatix materials printer DMP-2831 (Fujifilm) and a DMC-11601 piezoelectric cartridge (16 nozzles with 254 μm spacing and 1 μl nominal drop volume) filled with 1.5 mL of optimized rGO ink. Based on visualization of the ink drops using the drop-watcher camera, the jetting voltage applied was 13 V, whereas the appropriate drop spacing was 10 μm, with the platen temperature maintained at 40°C. Using these conditions, homogeneous patterns of 10 × 10 mm with 3, 7 and 10 prints were prepared; after each print, the substrate was heated at 100°C using a hot plate. Finally, the inkjet-printed deposits were thermally treated in ambient atmosphere from 25°C up to 200, 300 or 400°C for 1 hour using a ramp of 3°C/min; the sample identification used hereafter will be rGO film 200, rGO film 300 and rGO film 400, respectively.

Raman analysis was performed with an Alpha 300 WITec equipment using a 50X objective and operated at λ = 488 nm. The morphology was analyzed by scanning electron microscopy (SEM) using a JEOL JSM-7600F field-emission microscope. The UV-Vis spectrophotometry analysis (Agilent Instruments) was carried out on rGO films in the range of 200 to 800 nm using an identical FTO substrate as reference. X-ray photoelectron spectroscopy (XPS) analysis was realized using a K-Alpha Thermo Scientific spectrometer with Al Kα X-ray monochromatic source (1486.6 eV) at 12 kV and 40 W. The analyzed area was 400 μm using an incident relative angle of 30°. Before analysis, the surface of the samples was cleaned by Ar erosion (30 s at 3 kV and 30 W). Survey spectra were obtained from 0 to 1350 eV (1 eV/step and 100 eV pass energy), whereas for high-resolution spectra, 0.1 eV/step and 50 eV pass energy were used. The binding energies were referenced to adventitious carbon (C1s) at 284.8 eV.

### Electrochemistry and dye-sensitized solar cells (DSSCs).

— The electrochemical measurements of 1 mM Co(bpyp)3(BCN)3 (Dyennano, DN-CO4, >95%), 0.1 M TBAPF6 (tetrabutylammonium hexafluorophosphate; Aldrich, 98%) in acetonitrile were obtained by cyclic voltammetry using a potentiostat/galvanostat/ZRA (Gamry Instruments Reference 3000) in a three-electrode electrochemical cell, employing Ag/AgNO3 (0.01 M) as reference electrode and a Pt disk as counter electrode. The working electrode consisted of FTO (TEC 15), platinumized FTO, or FTO covered with inkjet-printed rGO films. All the experiments were performed at 25°C.

Dye-sensitized solar cells were elaborated with TiO2 (Dyesol DSL-18NR-T TiO2 paste) photoanodes deposited by screen-printing onto FTO-covered glass (TEC-15) previously cleaned with piranha solution. The 11 μm TiO2 films were sintered at 530°C for 1 hour, and after cooling to 80°C the films were immersed in the dye solution consisting of 0.2 mM D35-dye (Dyennano, DN-F04) + 1 mM chenodeoxycholic acid (CDCA, Sigma-Aldrich, >97%) solution in acetonitrile/tert-butyl alcohol (1:1 v/v) and sensitized for 24 h. The counter electrodes of the DSSC consisted of FTO, platinumized FTO (prepared by spreading a drop of Platisol T (Solaronix) on the conductive side of the electrode and subsequent heating at 450°C for 5 min), or FTO with inkjet-printed rGO films. The photoanode and the counter electrode were sealed together in a sandwich configuration using a frame of the thermoplastic Meltonix 1170–60 (Solaronix) by heating at 210°C for 5 min. Finally, an acetonitrile-based electrolyte solution consisting of 0.22 M Co(bpyp)3(BCN)3 (Eversolar) + 0.05 M Co(bpyp)3(BCN)3 (Dyennano, DN-CO4, >95%) + 0.1 M LiClO4 (Aldrich, >95%) + 0.2 M 4-tert-butylypyridine (Aldrich, 99%) was introduced into the cell through a pair of holes previously perforated in the counter electrode, which were subsequently sealed with pieces of Meltonix and microscope cover glass pressed under heating. Photovoltaic characterization was performed using an Oriel Sol2A Class ABA solar simulator, calibrated to an irradiance of 90 mW/cm² using a certified 4 cm² monocrystalline silicon reference cell.

### Results and Discussion

#### rGO ink characterization.

In order to optimize the ink properties, the effect of the sonication time and PVP content on particle size was analyzed by means of DLS. It is important to consider the effective particle size because if the dispersions contain rGO particles in the range of 0.2–2 μm, i.e. 100 to 10X smaller than the opening size of the printing nozzle (22 μm); agglomeration could occur, and the particles clog the nozzles complicating the inkjet printing process. Figure 1a shows that the rGO inks containing 5% PVP and sonicated for different durations displayed only one peak, centered at about 275–300 nm. For the case of the rGO dispersions with 10% PVP, the principal peaks are centered at about 250–280 nm, as shown in Figure 1b. The dispersions with 15% PVP present peaks centered at about 270–285 nm, see Figure S1c. However, it was observed that for dispersions containing 10% and 15% PVP additional small peaks representing sizes larger than 1000 nm are detected. For all DLS results, the tables in the insets (in Figures 1a, 1b and S1c) present the principal centered peak values with their standard deviation, as well as the lateral dimension \(<λ>\) calculated according to a previous report.31

On the other hand, Figures S1a, S1b and S1d in the supplementary material depict the results of the particle size distribution of PVP-IPA dispersions: all dispersions of PVP (5, 10 and 15%) show a distribution in a wide size range (from 10–5,000 nm), independent of the sonication time.

These DLS results are very important because they provide real information on the behavior of rGO flakes in dispersion. Therefore, taking into consideration that the lateral size dimension of rGO flakes obtained using only 5% PVP are similar to those using 10% and 15%, and that these are very close to the lower limit of particle size suitable to be utilized in a printing cartridge, the optimal ink was selected to consist of 25 mg rGO powder and 1.25 mg PVP (5%) dispersed in 10 mL IPA, with 8 hours of ultrasound-assisted exfoliation.

For practical applications, the dispersions as an ink need to be highly stable to prevent the precipitation of the rGO flakes. In Figure 1c, zeta-potential and DLS analysis of the optimized rGO ink as a function of time are shown. The pH of the ink is 6.2 and the zeta-potential is −21.2 mV, which is satisfactory according to previous work.32 Based on visual inspection as well as DLS and zeta-potential measurements it can be estimated that the optimal rGO ink presents good stability for at least 6 months after its formulation.

Figure 1d shows the UV-Vis electronic absorption spectrum of the as-prepared ink (as inset a photograph of rGO ink is depicted) presenting absorption principally at λ < 300 nm, characteristic of graphene and its derived compounds GO and rGO. Particularly, the optical absorption of GO increases due to π→π∗ transitions of C=C, presenting bands in the range from 225 to 275 nm (5.5–4.5 eV), and the presence of a shoulder at about 300 nm is often related to π→π∗ transitions of C=O.33 In our ink, the bands at 240 nm and 270 nm are associated principally to π→π∗ transitions, whereas the absence of the band at 300 nm is indicative of a low content of oxygenated functional groups; in addition, the band at 207 nm is related to the carbonyl functional group (ν(C=O) transition) of the PVP ring structure.33 The
concentration of the rGO in the ink is estimated to be 0.032 g/L, which was calculated by means of Beer-Lambert law and considering a $\alpha_{660\text{nm}}$ of 2460 L/g m$^{-1}$. On the other hand, the thermal behavior of the rGO ink was evaluated using TGA. The thermogravimetric and derivative curves for the rGO ink are shown in Figure S2; the curves corresponding to PVP and rGO powders are also presented as reference. As can be seen, the rGO ink presents a constant weight loss reaching about 41%, 57% and 78% at 200, 300 and 400°C, respectively. This is related principally to the removal of PVP, which exhibits two degradation stages. Besides, rGO powder remains completely stable until about 600°C whereas at this temperature PVP is essentially eliminated, as previously reported in Ref. 35.

**rGO film characterization.**—Before printing, the jetting voltage (JV) and drop spacing (DS) were optimized taking into consideration visual and microscopical examination. Applying a JV of 13 V, single and stable drops are obtained, as shown in Figure S3. The optimized value for the DS was determined to be 10 μm corresponding to 10,000 drops/mm$^2$ (resolution of 2,540 dpi). In addition, from Figure S3 the calculated drop velocity was about 8.1 m/s, which is within the range recommended for the inkjet printing process.

After optimization of the printing parameters and conditions, the optimal thickness of the rGO film was established. Square patterns of $10 \times 10$ mm were inkjet-printed with a thickness of 3, 7 and 10 layers, and were then characterized by Raman spectroscopy; Figure S4a shows the corresponding photographs. Figure S4b presents the Raman spectra of the rGO films with different number of printed layers. The intensity of the two characteristic peaks, D (1360 cm$^{-1}$) and G (1580 cm$^{-1}$), increase with the number of prints as expected, providing a clear indication of a good surface coverage and improving film homogeneity with number of prints. Therefore, films were prepared with 10 printed layers for subsequent characterization and evaluation of the electrocatalytic properties. The effect of thermal treatment on the physicochemical properties was assessed, and the photographs illustrate a slightly black color of the films, as shown in in Figure S4c, becoming darker with increasing thickness and annealing temperature. The Raman spectra of the films with 10 prints and annealed at 200, 300 and 400°C are depicted in Figure S4d. All spectra show the distinctive peaks, D at 1360 cm$^{-1}$ related with the number of defects presented on graphene structure with sp$^3$ hybridization, and G at 1580 cm$^{-1}$ originated from vibrations of sp$^2$ carbon and 2D at ~ 2700 cm$^{-1}$, which is the overtone of the D peak usually correlated with the number of graphene layers. A summary of the D, G and 2D peak analysis as well as the calculated I$_D$/I$_G$ ratio as a function of annealing temperature is presented in Table I, observing that the position of the three peaks remains unchanged for all films, which indicates non-functionalization upon annealing.

| Annealing (°C) | D band (cm$^{-1}$) | G band (cm$^{-1}$) | 2D band (cm$^{-1}$) | I$_D$/I$_G$ |
|---------------|------------------|------------------|---------------------|-----------|
| 200           | 1360             | 1577             | 2703                | 0.71      |
| 300           | 1363             | 1581             | 2700                | 0.84      |
| 400           | 1363             | 1581             | 2700                | 0.66      |
| rGO powder    | 1351             | 1569             | 2702                | 0.62      |

Table I. Positions of D, G and 2D peaks as well as I$_D$/I$_G$ values of inkjet-printed rGO films after different heat treatments.

*rGO used for preparation of the ink.
of graphene structure.\textsuperscript{5,36} Particularly, the slight shift of the G peak to higher wavenumber values for the inkjet-printed rGO films compared to rGO powder is a clear indication of the successful exfoliation of rGO layers. The $I_D/I_G$ ratio is related with the size of the $sp^2$ aromatic domain and can be used to quantify the number of defects and disorder present in graphene nanosheets. The calculated $I_D/I_G$ ratio was 0.71, 0.84 and 0.66 for the films treated at 200, 300 and 400°C, respectively; these results suggest an important interaction between rGO and PVP at the lowest temperature. The highest $I_D/I_G$ ratio is obtained at 300°C, which indicates that this rGO film presents a considerable number of defects associated probably to the partial degradation of PVP, originating the presence of many organic species. The diminution of the $I_D/I_G$ value at 400°C (close to that obtained for rGO powder) suggests a decrease in the oxidation level and is a clear indication of the recovery of the aromatic structure of graphene,\textsuperscript{5,37} as PVP and its byproducts have been completely removed at this temperature.

The XPS survey spectra of the different inkjet-printed rGO films are shown in Figure 2a, and the peaks related to O1s, Sn3d and C1s are clearly distinguished; the presence of O and Sn is due to the FTO substrate. On the other hand, Figure 2b depicts the survey spectra of PVP deposited as reference (5% PVP–IPA prepared under the same conditions as the optimal rGO ink, and (c) Deconvoluted XPS spectra of the C1s peak of rGO inkjet-printed films.

Figure 2. XPS survey scans of inkjet-printed rGO films after annealing at different temperatures: (a) rGO inkjet-printed films on FTO, (b) PVP deposited on FTO using a dispersion of 5% PVP–IPA prepared under the same conditions as the optimal rGO ink, and (c) Deconvoluted XPS spectra of the C1s peak of rGO inkjet-printed films.

be noted that the C1s of the PVP molecule presents peaks at 285.0, 285.4, 286.2 and 287.9 eV.\textsuperscript{38} For rGO film 400, the calculated atomic percentage of the C–O and C=O species, deconvoluted blue and yellow lines, respectively, was 13%, which is similar to values previously reported for rGO\textsuperscript{8} and very close to that for pristine rGO powder used for preparation of the ink. These results reveal that the oxygen functionalities present in pristine rGO powder remain unchanged even after exfoliation, printing and thermal treatment, up to the final fabrication of inkjet-printed rGO films.

Electrochemistry and dye-sensitized solar cells (DSSCs).—

Transparent conducting oxides are widely used as electrodes in a variety of electrochemical and electrical devices, including solar cells, sensors, smart windows, etc. In the dye-sensitized solar cell, the most commonly used counter electrode is FTO, however, an electrocatalyst is needed to facilitate the efficient reduction of the oxidizing agent. Although classically the $I_{3}/I_2$ redox couple in an acetonitrile solution is used, recent advances in the increase of the cell open circuit voltage have led to the development of novel redox couples, including Co(bpy)$_3$Cl$_2$/I$_3$ and Cu(dmp)$_2$/I$_2$/Cl$_2$. Carbon materials, including rGO, may be an excellent cost-effective alternative for the generally used platinum catalyst.

The electrochemical behavior of the [Co(bpy)$_3$]$^{3+}$ cation in solution was characterized by cyclic voltammetry on different substrates in a three-electrode electrochemical cell. Figure S5 shows a voltammogram for the reduction/oxidation of the [Co(bpy)$_3$]$^{3+}$/[Co(bpy)$_3$]$^{2+}$ redox couple in acetonitrile using FTO-Pt and FTO working electrodes. Cathodic and anodic peaks were observed at 0.035 and 0.119 V vs Ag/AgNO$_3$, respectively, with a peak separation of 84 mV characteristic of a quasi-reversible one-electron redox system. Figure S5 also shows the voltammogram obtained for the same solution on an FTO substrate. As can be seen, the electrochemical results are quite different to those observed for the FTO-Pt substrate, with a shift of the cathodic peak potential to more negative values and the disappearance of the anodic peak, characteristic of an irreversible system. This result clearly points to the need of an electrocatalytic coating (such as Pt) on the solar cell counter electrode, in order to increase the electron transfer rate to the oxidized species in the electrolyte solution.

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Figure 3. SEM images and UV-Vis spectra of inkjet-printed rGO films obtained at different annealing temperatures. In (a) – (c) representative SEM images are presented, and in (d) the UV-Vis transmittance spectra and the measured value at 550 nm are shown.

Figure 4a presents a set of cyclic voltammograms obtained for the same solution as in Figure S5, but obtained on FTO substrates with the inkjet-printed rGO films treated at different annealing temperatures. As shown in previous sections, the thermal treatment not only has an effect on the amount of PVP present but also on the morphology of the rGO films. Figure 4a illustrates that the electrochemical behavior of the Co-based coordination complex is also dependent on the heat-treatment of the inkjet-printed rGO films. A decrease of the annealing temperature is accompanied with a shift of the cathodic potential peak to more negative values and an increase in the peak separation, hence, irreversibility of the electron transfer process. For the FTO-rGO film 200, which is the most homogeneous film with still a significant presence of PVP, it can be concluded that the electrocatalytic activity is minimal, in fact, the performance is worse than that of clean FTO. This indicates that the remaining PVP blocks the active sites for the electron transfer reaction. The FTO-rGO films treated at 300 and 400°C show anodic and cathodic peaks located at quite similar values as those observed for FTO-Pt, which indicates that rGO is electrocatalytically active for both the reductive and oxidative redox reactions. An interesting observation is that for the FTO-rGO film 300, the cathodic and anodic current densities at the maximum are higher than for the other substrates; this can be explained by the significantly larger surface roughness, hence, active surface area, related to the morphology of rGO film, in agreement with the observations from the SEM images.

Figure 4b shows the current density corresponding to the reduction of the Co(bpy)₃³⁺ complex on the different substrates as working electrodes as a function of the overpotential (Tafel plot). As can be observed, at high overpotentials all systems show a current density related to mass transport limitation. At low overpotential, the current density exhibits an exponential dependence with overpotential, with a similar slope for all substrates indicating a similar charge transfer coefficient. However, the intercept at zero overpotential, which defines the exchange current density J₀, clearly depends on the substrate nature, following the same trends as observed in the cyclic voltammetry results. It can be concluded that the inkjet-printed rGO films annealed at 300 and 400°C are electrocatalytically active for the reduction of the Co(bpy)₃³⁺ complex, with the same J₀ as observed for the platinized FTO substrate, indicating the promise of rGO as electrocatalytic material in this system.

Table II. Short-circuit current density (JSC), open-circuit voltage (VOC), fill factor (FF) and efficiency (η) determined from the current-voltage curves shown in Fig. 4c.

| Sample          | JSC (mA cm⁻²) | VOC (V) | FF   | η (%) |
|-----------------|---------------|---------|------|-------|
| FTO             | 5.7           | 0.79    | 0.20 | 1.0   |
| rGO film 200    | 5.6           | 0.82    | 0.21 | 1.1   |
| rGO film 300    | 5.7           | 0.84    | 0.45 | 2.4   |
| rGO film 400    | 5.8           | 0.82    | 0.51 | 2.7   |
| FTO-Pt          | 5.6           | 0.82    | 0.60 | 3.1   |
Figure 4. Electrocatalytic evaluation and comparative results of inkjet-printed rGO films obtained at different annealing temperatures. (a) Cyclic voltammograms for a 1 mM Co(bpy)3(B(CN)4)3 + 0.1 M TBAPF6 solution in CH3CN. (b) Tafel plots for the reduction of Co(bpy)32+ from the same solution as in (a). (c) Current-voltage curves for TiO2-based DSSCs with the organic dye D35, in 0.22 M Co(bpy)3(B(CN)4)2 + 0.05 M Co(bpy)3(B(CN)4)3 + 0.1 M LiClO4 + 0.2 M 4-tert-butylpyridine; the inset shows a photograph of the TiO2-DSSC assembled with the rGO film 400 as counter electrode. (d) Cyclic voltammograms for the inkjet-printed rGO film 400 in the same electrolyte solution as in (a). All electrochemical measurements in (a), (b) and (d) were performed at 25°C and 50 mV s−1.

point with the JSC and the VOC (Equations 1 and 2):

\[ \eta = \frac{(J_{SC}V_{OC}FF)}{P_{in}} \]  

where \( J_{Max} \) denotes the maximum current density and \( V_{Max} \) denotes the maximum voltage.

\[ \text{FF} = \frac{J_{Max}V_{Max}}{J_{SC}V_{OC}} \]

where \( P_{in} \) is the incident power. Hence, a decrease of the FF leads to a decrease of the cell efficiency, which is generally attributed to an increased resistance in the cell, in this case, an increase of the electron transfer resistance at the counter electrode/electrolyte solution interface.

The FTO and the FTO-rGO film 200 cells showed the lowest FF and efficiency values, while the FTO-Pt shows the best performance. For the FTO-rGO cells a progressive increase of FF was observed as function of the heat-treatment temperature, and the best result was obtained for the rGO film 400, indicating an increase of the electrocatalytic properties of the films as the amount of PVP decreases. The solar cells with the rGO-catalyzed counter electrodes annealed at 400°C show an efficiency of 2.7% versus 3.1% for the Pt-catalyzed FTO, a reduction of 13%. Interestingly, the Tafel plots indicated that the electrocatalytic activity of the rGO 300 and 400 films is equal to that of Pt in a dilute Co(bpy)32+/3+ solution with inert salt in acetonitrile. However, in the more concentrated solar cell electrolyte solution, which not only contains the cobalt complex but also several potential-defining additives including LiClO4 and TBP (4-tert-butylpyridine), the interaction of these compounds with the FTO-rGO films affects the electron transfer rate at the counter electrode/electrolyte interface resulting in a small drop in the FF values compared to the FTO-Pt cell. Finally, Figure 4d shows cyclic voltammograms for the cobalt-based solution on an FTO-rGO film 400 electrode as a function of number of cycles. The electrochemical characteristics remain the same for up to 200 cycles indicating excellent stability, both of the film in the organic electrolyte solution and the redox couple after multiple reduction/oxidation sequences.

It can be concluded that inkjet-printed rGO films are a promising, low-cost alternative for Pt counter electrodes in DSSCs, and we have demonstrated the feasibility of the fabrication of rGO films by using this high-throughput, reproducible and scalable technique. These results illustrate the promise of inkjet-printed rGO films in many important technological applications, such as DSSCs, perovskite-based solar cells, fuel cells, optoelectronics and other electrical and electrochemical devices.

Conclusions

An inexpensive, stable and environmentally friendly rGO ink with a concentration of 0.032 g/L and an average lateral size of 340 nm was formulated for the preparation of inkjet-printed rGO films. We have established the optimal printing conditions allowing a good spatial uniformity of the rGO film after just 10 prints, taking advantage of the automated and high-control drop-on-demand characteristics of the inkjet printing technique. The characteristic oxygen functionalities of rGO powder remain unchanged after exfoliation, printing and thermal treatment at 400°C. Electrochemical studies show similar electrocatalytic features of inkjet-printed rGO films, annealed at 300 or 400°C, compared to drop-casted Pt, both on FTO substrates, for the reversible reduction and oxidation of the Co(bpy)32+/3+ redox complex in acetonitrile, indicating their potential application as dye-sensitized solar cell counter electrode electrocatalyst. The inkjet-printed rGO films demonstrate high chemical compatibility as well as good adherence on FTO when in contact with the organic solvent used, and stable performance for at least 200 oxidation/reduction cycles. The efficiency
of dye-sensitized solar cells fabricated with inkjet-printed rGO films as counter electrode showed a dependence on the thermal treatment applied. Specifically, rGO film 400 exhibited the best electrocatalytic activity as counter electrode reaching an 87% of the efficiency observed for the DSSCs with a Pt-catalyzed counter electrode, demonstrating the possibility of substituting expensive and scarce Pt by low-cost rGO films fabricated using a scalable and highly reproducible inkjet printing process.

Acknowledgments

To CONACYT-SENER for postdoctoral scholarship of R. Martínez-Flores, Cátedras-CONACYT Projects 319 and 1710, as well as CONACYT, SENER and IER-UNAM for funding through the Mexican Center for Innovation in Solar Energy (CeMIE-Sol), Project P-27. Authors also are grateful for the use of the facilities of LANNBIO Cinvestav-Mérida supported by the grants: FOMIX-P-27. Authors also are grateful for the use of the facilities of Mexican Center for Innovation in Solar Energy (CeMIE-Sol), Project well as CONACYT, SENER and IER-UNAM for funding through the

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ORCID

Gerko Oskam https://orcid.org/0000-0002-2105-5874
Miguel A. Ruiz-Gómez https://orcid.org/0000-0001-5263-8151