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Solar Energy

DOI:

<https://doi.org/10.1016/j.solener.2020.08.046>

Published: 15/09/2020

Peer reviewed version

[Cyswllt i'r cyhoeddiad / Link to publication](#)

Dyfyniad o'r fersiwn a gyhoeddwyd / Citation for published version (APA):

Menendez, M. F., Martinez, A., Sanchez, P., Andres, L. J., Gomez, D., Haponow, L., Bristow, N., Kettle, J., Korochkina, T., & Gethin, D. T. (2020). Development of intermediate layer systems for direct deposition of thin film solar cells onto low cost steel substrates. *Solar Energy*, 208, 738-746. <https://doi.org/10.1016/j.solener.2020.08.046>

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1 **Development of intermediate layer systems for direct**
2 **deposition of thin film solar cells onto low cost steel**
3 **substrates**

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11

12

Abstract

13 The functionalisation of low-cost steel over large areas with low cost intermediate layers (ILs)
14 for utilisation as substrates in thin film solar modules is reported. Three approaches for the
15 deposition of ILs are demonstrated and evaluated; a thick SiO_x sol-gel based on a one-step acidic
16 catalysis applied by spray technique, a commercial screen-printable dielectric ink, and an epoxy-
17 based material (SU8) deposited by screen printing or bar coating. These ILs demonstrated the
18 properties of surface levelling (quantified by mechanical profilometry), electric insulation (tested
19 using breakdown voltage and leakage current) and acted as an anti-diffusion barrier (demonstrated
20 with glow discharge mass spectrometry). Moreover, the performances of amorphous silicon (a-
21 Si:H) and organic photovoltaic (OPV) thin film solar cells grown on carbon and stainless steels
22 (a-Si:H: 5.53% and OPV: 2.40%) show similar performances as those obtained using a reference
23 glass substrate (a-Si:H: 5.51% and OPV: 2.90%). Finally, a cost analysis taking into account both
24 the SiO_x sol-gel and the dielectric ink IL was reported to demonstrate the economic feasibility of
25 the steel/IL prototypes.

26

27 **1. Introduction**

28 Building Integrated Photovoltaics (BIPV) has emerged as an important strategy for
29 domestic energy production with commercial interest driven by the EU's 2020 targets [1,
30 2] and the consideration that all new buildings should be nearly zero-energy by the end
31 of 2020. Although BIPV still represents a small share of the photovoltaic (PV) market, a
32 number of demonstrators have shown the feasibility of BIPV systems and their superiority
33 over conventional roof installation (Building Attached Photovoltaics (BAPV))
34 approaches in terms of overall cost and aesthetics [3, 4]. BIPV concepts have been applied
35 within a diverse range of building sector products including for public/commercial and
36 domestic buildings, both for retrofitting and for new buildings (see, for example, [5]).

37 Most BIPVs are made using silicon-based materials; however, further improvements in
38 product innovation and cost are still required in order to compete with mainstream PV
39 technologies. As a result a number of different authors [6, 7, 8, 9] have proposed: (i) the
40 integration of thin film solar cells (TFSC) as alternatives to crystalline silicon (c-Si), and
41 (ii) the development of BIPVs directly onto conventional building materials instead of
42 glass substrates.

43 In order to introduce the TFSC technology in the market in a competitive fashion, a
44 number of challenges need to be overcome. Firstly, there is a need to demonstrate the
45 feasibility of low-cost substrates that are already used by the construction industry; for
46 example to adopt steel substrates, low cost alternatives are needed as existing work is based
47 on expensive "solar-grade" stainless steel (SS), which is too costly for the construction
48 industry. Therefore, it is more cost effective if existing construction material is adapted
49 for BIPV usage, but this requires specific preparation of the steel surface. The preferred
50 solution is the use of an intermediate layer (IL) system between the steel and the TFSC.
51 The steel industry has substantial experience of coating insulating layers so this is a
52 feasible option for implementation. The main requirements for this IL layer are: (i) to
53 smooth the steel surface in order to prepare it for the application of TFSC nanolayers and
54 to prevent direct short-cuts in the future integrated solar cells; (ii) to prevent the diffusion
55 of chemical elements (e.g. Fe, Cr, Ni, etc.) from the steel to the TFSC during the solar
56 cell process fabrication; (iii) to electrically insulate the TFSC back contact electrode from
57 the steel substrate to allow the development of TFSC modules by the interconnection of
58 monolithic serial cells using laser ablation scribing processes; and (iv) to be stable and

59 not delaminate during post-processing for semiconductor layer deposition as well as
60 during storage.

61 Recently, several works concerning IL system deposition on steel substrate to develop
62 chalcopyrite (CIGS) or silicon TFSC have been reported. Whilst promising, their work
63 used relatively thin IL which are unlikely to be scalable for solar modules. L. Zortea *et al.*
64 *al.* [10] investigated the behavior of an electroplated bi-layer (Ni and Cr) on a mirror
65 surface finished (average roughness, $R_a < 0.1\mu\text{m}$) mild steel foil. Kyoung-Bo *et al.* [11]
66 developed monolithic CIGS on SS applying, by screen-printing, a $10\mu\text{m}$ silicon dioxide
67 (SiO_2) IL. However, the resulting R_a was $0.21\mu\text{m}$ and only poor levelling was achieved
68 through the sol- SiO_2 ($R_a=0.157\mu\text{m}$). Thongkham *et al.*[12] applied aluminum oxide
69 (Al_2O_3) ($2\mu\text{m}$ thick) by physical vapour deposition (PVD) on different SS substrates
70 (types 301, 316L and 430) and reported a very poor adhesion due to the different
71 coefficient thermal expansions (CTE) of the IL compared to the CIGS thin films.

72 Concerning silicon based TFSC, Yun *et al.* [13] developed SiO_2 and $\text{Al}_2\text{O}_3/\text{SiO}_2$ sol-gel
73 stack as IL for SS. The results showed poor adhesion of the layers and etching the surface
74 with hydrochloric (HCl) acid solution prior to application of the IL was needed to increase
75 the roughness and improve the adhesion. Lee *et al.* [14] reported the improvement of the
76 performance of silicon TFSC on SS substrate by an electrochemical mechanical polishing
77 method. This process created chromium oxide (Cr_2O_3) and iron oxide (Fe_2O_3) passive
78 films which act as anti-diffusion layers, blocking Cr and Fe diffusion into the silicon
79 TFSC. However, electrical insulation was not achieved using these passive films.
80 Martinez *et al* [9] developed a SiO_x sol-gel multilayer to make automotive hot-dip
81 galvanized steel compatible with amorphous silicon (a-Si:H) TFSC. A power conversion
82 efficiency (PCE) of 3.03% was achieved, but the scalability to larger areas was not
83 addressed. Lopez *et al* [15] combined SiO_x sol-gel and SiO_x plasma enhanced chemical
84 vapor deposition (PECVD) films in two and three layers stacks on SS foils, reporting
85 highly dielectric properties (breakdown voltages $> 900\text{V}$) and compatibility with CIGS
86 TFSCs.

87 In general, CIGS and silicon-based thin film technologies are the most widely reported
88 and mature PV systems which have been developed onto steel substrates, although there
89 have also been some attempts with dye sensitized [16], cadmium tellurite [17], organic
90 photovoltaic (OPV) [18-21], and kesterite solar cells [22]. Many of the reports, such as
91 Ding *et al* [21] were focused on non-scalable approaches to IL deposition for OPVs using

92 spin-coating. As OPVs are likely to be made commercially by roll-to-roll (R2R) production
93 methods, similar processing strategies must be adopted for the application of ILs.

94 As can be seen in the works above, steel/IL systems show several drawbacks for an
95 industrial application, such as the necessity to process the steel surface to reach a very
96 low roughness [10, 14], poor IL adhesion [12, 13], the lack of electrical insulation [14],
97 and the difficulty in implementing multilayer stacks [9, 15].

98 Moreover, most of these examples use relatively expensive steels (e.g. stainless and only
99 very thin substrates, around 25 μ m thick), and in most of these cases the up-scaling of the
100 process in terms of manufacturing yield, potential or cost is rarely reported. This creates
101 a paradoxical situation; by using an expensive steel substrate, the end product is difficult
102 to be introduced to the building sector, as it is more common to use a lower cost steel
103 such as dipped or galvanised low carbon steels for building infrastructure.

104 In this paper several low-cost IL strategies (ceramic sol-gel and polymer-based) deposited
105 onto low-cost structural steel grades are reported. The developments were carried out with
106 the view to possible further industrial application, so only single layer IL strategies were
107 pursued to make compatible steel substrates and TFSC. In that sense, one challenge of
108 the present work concerned the sol-gel IL, as its main drawback is that its maximum
109 achievable thickness without stress cracks forming is low (typically, 1 μ m).

110 In addition, only high throughput coating techniques were employed in order to
111 demonstrate the potential for upscaling the approach. The IL systems were characterised
112 morphologically and electrically, and different TFSC (a-Si:H and OPV) were grown and
113 compared with glass substrates as reference to demonstrate the feasibility of our approach
114 for the thin-film solar community. The results show that the behaviour of amorphous
115 silicon cells and OPVs on intermediate layers of SiO_x sol-gel and SU8, respectively, are
116 indistinguishable from cells grown on glass, which constitutes the most innovative part
117 of this work.

118

119 **2. Experimental**

120 **2.1 Substrate selection**

121 The initial selection of steel substrates has been made by consideration of four main
122 factors: (i) chemical inertness, (ii) thermal coefficient of expansion with respect to the IL

123 materials, (iii) surface quality in terms of absence of defects and low initial roughness,
124 and (iv) pre-existing usage within the construction sector.

125 Four steels were selected which met these criteria: a bare low carbon steel (DC01), two
126 hot-dipped metallic low carbon steels (zinc-coated DX51D+Z and aluminium-silicon
127 coated DX51D+AS) and a ferritic stainless steel (AISI430). Chemical composition of
128 each type is given in Table I. Initially, all the steels were rolled by high speed steel (HSS)
129 grinding rolls in order to reduce the initial peak to-peak (R_z) surface roughness down to
130 1.3 μm . This roughness value was considered as a good compromise between the rolling
131 cost and the intermediate layer approach considered. The steels were rolled to a thickness
132 of 0.3mm as this is compatible with lab-scale coating techniques and leads to a 10 times
133 reduction in the steel costs when compared to the industrial standard stainless steel used
134 in previous attempts to integrate steel and TFSCs [23].

135

136 **2.2 Intermediate layer (materials, processes and characterization)**

137 Once the steel substrates were prepared, ILs were applied to fulfil the following
138 requirements: (i) to level and minimize the surface roughness for high quality thin film
139 solar deposition, (ii) to electrically isolate the steel substrate for safety and to enable
140 monolithic integration, (iii) to act as a barrier for ionic diffusion and to avoid TFSC
141 contamination from the steel, (iv) to give thermal and mechanical stability to the PV cell
142 or module above the steel substrate, (v) to show a long shelf life during storage, and (vi)
143 to be compatible with subsequent PV deposition processes. The selected ILs and the
144 deposition methods are listed below:

145 **2.2.2 SiO_x-based sol-gel coatings**

146 **2.2.2.1 Sol-gel synthesis**

147 Sol-gel is a well-established technology for thin-film coatings in the steel industry;
148 materials are applied in a mixture that leads to a hydrolysis-condensation of the precursors
149 and enables coatings through low cost/temperature deposition techniques. Sol-gel
150 technology offers a good control of the thickness and a high degree of homogeneity
151 through the creation of an inorganic matrix. The main drawback for sol-gel is that its
152 maximum achievable thickness without stress cracks forming is low (typically, 1 μm).
153 Cracks are mainly produced during evaporation of solvents due to mechanical and

154 thermal stresses between the layer and the substrate. The introduction of tension releasing
155 agents in sol-gel matrixes constitutes an interesting approach to reduce cracking and
156 increase coating thickness. Special attention to this strategy has been considered in this
157 work. In this work, the sol-gel formulation followed a one-step acid catalysis using
158 methyltriethoxysilane (MTES 99%, Aldrich) and tetraethyl orthosilicate (TEOS 98%,
159 Aldrich) as precursors. Water was incorporated as a solvent, polyethylene glycol 6000
160 (PEG-6000, Panreac) as a tension releasing agent and HNO₃ as a catalyst. The molar
161 ratios were optimised in order to reach a suitable homogeneity, adhesion and pin-hole
162 free layer after application and curing. These were TEOS/MTES: 1.35,
163 H₂O/(MTES+TEOS): 4.7, and PEG-6000/(MTES+TEOS): 0.02. The synthesis was
164 carried out at room temperature with vigorous magnetic stirring. The addition of HNO₃
165 initiates the hydrolysis, increasing the temperature by 25°C. During the sol phase
166 preparation, the ambient conditions have been controlled, being the average temperature
167 registered of 23°C and the relative humidity of 48%. This sol mixture was aged for one
168 hour under stirring before application.

169

170 **2.2.2.2 Sol-gel deposition and curing**

171 Spray coating was performed with a commercial air atomizing spray gun (Spraying
172 Systems Inc.) with a nozzle cone angle of 60°. It was mounted on an automatized spray
173 system and moved transversely with respect to the sample movement. The 30cm x 30cm
174 samples were coated using the following parameters: (i) SiO_x sol-gel dilution 1:0.5 in
175 ethanol, (ii) a sol-gel liquid pressure between 0.4bar and 0.5bar, (iii) a holder speed of
176 1.6m/min, (iv) a nozzle/sample distance of 8cm, and (v) a one scan spray.

177 Curing was carried out by means of near-infrared (NIR) technique. The fast heating of
178 the steel substrate combined with the relative transparency of the sol-gel film to NIR
179 radiation allows a rapid, indirect annealing of the sol-gel layer. In addition, the prospect
180 of curing from the inner to the outer part of the coating enables a correct extraction of the
181 solvents. The experiments were performed using a NIR equipment (AdPhos, model 336-
182 250) composed of eight NIR lamps (0.8kW each) each 250mm x 42mm, which outputs a
183 maximum power density of 76kW/m². The distance between the NIR lamps and the
184 steel/IL prototypes was kept constant at 20cm and the process was developed in sheet-to-
185 sheet mode. At an irradiation power of 90%, a cure time of 2 minutes was applied in all

186 cases (reaching a peak metal temperature of 198°C), which represents a clear time
187 reduction when compared to conventional hot-plate curing (close to one hour, typically).

188 In order to study the suitability of the sol-gel coatings for the coating of selected steels,
189 the surface tension was measured using a Dynamic Absorption and Contact Angle Tester
190 (FIBRO DAT 1100), and showed a value of 26.19 ± 0.14 mN/m. Under the same
191 experimental technique, the surface free energy of the four steel substrates was also
192 evaluated using the Owens-Wendt-Rabel-Kaelble method. The values obtained were
193 between 38 mN/m and 40 mN/m for all steel types, and confirm the suitability of the sol
194 formulation for coating onto these steels. Additionally, viscosity has been calculated
195 using a Bohlin-Gemini rheometer. The obtained value was 0.005 Pa.s at 230 1/s. This
196 ensures the solution is suitable for dip and spray coating technologies and should lead to
197 relatively thin coatings.

198 **2.2.3 Organic commercial coatings**

199 Organic coatings have also been trialed as ILs. These materials allow easier processing,
200 thicker coatings with better dielectric behaviour and potentially better planarization.
201 Organic materials were sought on the basis of the following priorities: low cost, proven
202 dielectric properties and usage in other industries i.e. electronics, printability/coatability,
203 good insulation and good adhesion to substrates. Two materials were trialed:

204 **2.2.3.1 Blue Ink**

205 Blue Ink is a commercially available dielectric ink also known as ‘blue dielectric paste’
206 (D2140114D5) from Sun Chemical, USA. This material was screen printed by means of
207 a sheet-to-sheet semi-automated system (DEK248) suitable for handling 35cm x 35cm
208 (up to 6 mm thick) steel sheets. The layers were applied directly onto the steel through
209 square stencils. After coating, the ink layer was then dried using a hot air conveyor dryer
210 at 150°C for 20 minutes.

211 **2.2.3.2 SU-8**

212 SU-8 is an epoxy-based material, which contains a bisphenol A novolac epoxy that is
213 dissolved in cyclopentanone and mixed with 10% of mixed triarylsulfonium salt which
214 acts as the photoacid generator. It is widely used in the microelectronics and micro electro
215 mechanical systems (MEMS) industry as it crosslinks under ultra-violet (UV) irradiation.
216 After coating it can provide a suitable interface between a steel substrate and a PV cell.

217 The SU-8 epoxy layer (the formulation used is SU8-2050, supplied from Chestech Ltd.,
218 UK) was bar coated onto the steel samples (size 18mm x18mm). In contrast to the earlier
219 work of Ding *et al* [21], the SU-8 deposition approach is scalable to larger areas, therefore
220 potential enabling module production. The film was then cured at 150°C for 15 minutes
221 then hard baked at 250°C for 10 minutes.

222

223 **2.2.3 Characterization of the steel/IL systems**

224 Three main characterization techniques have been considered in this work. Firstly, the
225 impact of the IL on surface roughness, which has been determined through roughness
226 analysis by mechanical profilometry (Model XP-1, Ambios Technology, USA).
227 Profilometry was selected for surface roughness measurements because it allows for
228 surface roughness measurements to be conducted over large areas. A large surface area
229 needs to be mapped because a small number of surface topography issues could lead to
230 electrical shortages. Profilometry allows for large processing anomalies to be identified,
231 but also allows for the surface roughness to be accurately characterized as the resolution
232 of our system is 5Å.

233 Secondly, the anti-diffusive behaviour of the layer has been characterized by solid state
234 Glow Discharge Time of Flight Mass Spectrometer (GD-ToF-MS). GD sources coupled
235 to MS provides rapid and highly sensitive characterisation of major and trace elements in
236 homogeneous solids as well as depth profiling analysis of coating materials [24]. The
237 radiofrequency (rf) GD-ToFMS instrument consists of a rf-GD bay unit from Horiba
238 Jobin Yvon coupled to a fast orthogonal time-of-flight mass spectrometer (Tofwerk) with
239 a microchannel plate detector [25]. A pure copper modified Grimm-type GD source
240 (EMPA), with a 4 mm diameter anode and a 2.5 mm inner diameter flow tube was used.
241 The power is supplied to the GD through the back side of the sample by an rf-power
242 generator operating at 13.56 MHz. Experimental conditions (120 Pa argon discharge
243 pressure, 70 W rf forward power) were chosen as a compromise between high sensitivity
244 and good depth resolution. Finally, the dielectric behaviour has been analysed. Two
245 approaches were developed. Concerning the breakdown voltage, aluminium (Al) pads
246 (1cm²) have been first deposited by thermal evaporation (Pfeiffer, model Classic 500) on
247 the IL using a shadow mask. The tests were then performed varying a voltage in a range
248 of 0V to 53V (using a direct current source) between a copper probe connected to the

249 steel substrate and another probe placed onto the metallic Al pad. The breakdown voltage
250 occurs when visual shunts suddenly appear at the IL surface. This simple check also
251 verifies the IL quality by detection of defects such as small voids/cracks; if this were the
252 case, the breakdown voltage occurs at very low values (<10V). To evaluate the maximum
253 voltage which the IL will have to endure, a value of 40V has been chosen considering
254 30cm x 30cm panels, typical 1cm width serial interconnected cells, output voltages of
255 current thin film commercial modules and by applying a security factor of 50% over-
256 voltage. A second method to determine the dielectric behaviour of the ILs has been
257 through the measurement of the leakage current. Although this test requires a low current
258 source measurement unit, it enables a quantification of the IL insulation. For these tests,
259 aluminium electrodes have been deposited in a thermal evaporator (Edwards 306). A
260 shadow mask was used to define an active area for the electrodes. A Keithley 237 High
261 Voltage Source Measurement Unit (SMU) was used to measure the leakage current
262 between the back electrode and steel substrate. The bias range was selected from -50V to
263 +50V and the SMU is capable of measuring leakage current at a resolution of 10^{-13} A.
264 Small contact areas were used (5mm diameter circle) and contacted using electrical
265 probing.

266

267 **2.3 PV technologies**

268 **2.3.1 Amorphous silicon**

269 Amorphous silicon solar cells were manufactured onto steel with an n-i-p a-Si:H
270 structure, instead of the more commonly used p-i-n junction configuration, which is
271 widely adopted for transparent substrates (see Figure 1a and Figure 1b). The bottom and
272 top electrode materials have been reversed in order to improve their electrical
273 conductivity and the electron extraction. Therefore, a bottom electrode of aluminium (Al)
274 and indium tin oxide (ITO) was chosen. The Al and ITO layers were deposited by thermal
275 evaporation (Pfeiffer 500 System) and rf sputtering (AJA International, model ATC Orion
276 8HV), respectively. Both processes were performed at room temperature. ITO samples
277 were measured for transparency using UV-NIR spectrophotometry (Avantes, model
278 AvaSpec2048-USB2) with the wavelength range from 400nm to 800nm. Transmittance
279 higher than 90% was achieved in all cases. ITO sheet resistance (R_s) was determined by

280 the four-point probe method (Lucas-Signatone, model Pro4) and values of $20\Omega/\square$ and
281 $8\Omega/\square$ respectively for back and front contacts.

282 The n-i-p a-Si:H layers were deposited by (Very-High Frequency) VHF-PECVD in a
283 cluster configuration system which has separate chambers for the deposition of intrinsic,
284 n-type and p-type layers in order to avoid cross contamination (Elettrorava, model
285 V0714). The silicon deposition process was performed using a plasma gas reaction using
286 SiH_4 as the plasma gas and applying a power density of $16\text{mW}/\text{cm}^2$ at a fixed frequency
287 of 13.56MHz . The gas mixtures, the substrate temperature and the chamber pressure
288 during the n-i-p layers are detailed in previous work [9]. In order to achieve better
289 extraction of the generated electrons, a silver grid busbar was deposited on the top ITO
290 electrode in the samples. All samples were made with an active area of 1cm^2 .

291 The performance of the a-Si:H TFSC was determined by measuring the power conversion
292 efficiency (PCE) from current-voltage (I-V) curves generated by the TFSC devices under
293 global AM1.5G spectrum ($1000\text{W}/\text{m}^2$, 25°C) solar simulator (Oriel, model 91193)
294 through the standard IEC 60904-3:2008. In parallel to a-Si:H TFSC grown on our steel/IL
295 samples, n-i-p inverted cells were also manufactured on glass substrate as a reference.
296 This configuration allows us to benchmark the performance of PVs made on IL-coated
297 steel with those made on the more conventional glass substrate and provide information
298 to the PV community about the relative performance of PVs on steel as opposed to glass.
299 All reported performances are averaged from 6 devices.

300

301 **2.3.2 Organic Photovoltaics**

302 OPVs were fabricated on IL coated steel substrates composed of an opaque bottom
303 electrode, a thin-film semiconductor, and a transparent top electrode (see Figure 1c). For
304 device fabrication, a similar process to the work of Ding *et al.* was used [21] with the
305 following inverted layer structure: Steel-Al-Cr-ZnO/-3HT:PCBM-CLEVIOS HTL-
306 AgNW. The IL coated substrates were used for the deposition of the solar cell. Al/Cr
307 metal back electrodes were thermally evaporated onto the steel/IL system through a
308 shadow mask in an Edwards 306 thermal evaporator. A zinc oxide electron transporting
309 layer was deposited from zinc acetate dehydrate (109mg) dissolved in 2-methoxyethanol
310 (1ml) and ethanolamine (0.03ml) solution. This was spin-coated on the metal electrode
311 and annealed in the presence of atmospheric air at 150°C for the zinc acetate to calcinate

312 into zinc oxide. Initially, active layer blends using poly(3-hexylthiophene) (P3HT) and
313 [6,6]-Phenyl-C61-butyric acid methyl ester (PCBM) with weight ratios 5:4 were prepared
314 and mixed with chlorobenzene solvent with a concentration of 40mg/ml, and allowed to
315 dissolve for 24 hours on a hot plate at 60°C. Samples were stored in a nitrogen atmosphere
316 glovebox ($[O_2] < 1\text{ppm}$, $[H_2O] < 100\text{ppm}$), where the active layer was applied by spin-
317 casting from the 60°C solution to form a 220nm layer. This is thicker than earlier work
318 and is required in order to ensure higher yield as the SiO_x sol-gel layers create more
319 roughness prior to solar cell deposition. The active layer was annealed at 140°C for 30
320 minutes before the hole transport layer (HTL, i.e. Clevios HTL PEDOT) was spin-coated
321 at 4000rpm. The transparent top electrodes were fabricated by spray coating PH1000
322 PEDOT:PSS (purchased from Ossila ltd., UK) and 0.5mg/ml silver nanowire (Ag NW)
323 (L-50, purchased from ACS Materials Inc, USA) in ethanol subsequently onto the HTL
324 in a fume hood through a shadow mask with an air brush. All samples were made with an
325 active area of 1cm² and the results presented are averaged from 6 devices.

326 Alongside the OPVs on steel substrates, conventional inverted cells were manufactured
327 on ITO coated glass substrates. The properties of the ITO-glass were: $R_s=18\Omega/\square$,
328 transparency=84% with glass, transparency=94% without glass, and were purchased from
329 Xinyan Ltd. The samples were cleaned using deionised water, acetone and isopropanol
330 in an ultrasonic cleaner and then treated in a UV-ozone reactor with oxygen plasma for
331 10 minutes and were then used to fabricate OPVs with the following layer structure:
332 ITO/ZnO/P3HT:PCBM/CLEVIOS HTL/Ag. Except for the ITO and the thermally
333 evaporated Ag top electrode, all the other layers were the same as opaque steel substrate
334 cells. As with the a-Si:H cells, the reference sample provides a comparison between PVs
335 made on steel substrates with conventional processing routes. In addition, testing was
336 conducted using the methodology explained in the a-Si section.

337 **3. Results and discussion**

338 **3.1 Levelling and barrier properties of ILs on steel**

339 The three IL coatings were applied onto the four selected steels types, leading to 12
340 steel/IL combinations. The optimised deposition procedure is highlighted in section 2 for
341 the different layers. The measured thicknesses of the IL coatings were independent of the
342 steel substrate and were between 3-4µm for sprayed sol-gel coatings, ~30µm for the
343 BlueInk and ~70µm for SU8. Thicknesses were initially measured using profilometry,

344 but confirmed with cross-sectional field emission secondary electron microscopy (FE-
345 SEM) as shown in figure 2. The images show the consistency of the IL layers with low
346 surface roughness and are virtually defect free in the bulk. For the organic layers, a
347 moderate optimisation process was needed in order to apply the materials onto steel
348 substrates. However, for sol-gel coatings, a significant optimisation was required to
349 ensure that the right combination of formulation and process spray parameters were used
350 to obtain a consistent and robust coating. However, once fully optimised, the process was
351 readily reproducible and the results explained herein are based upon the optimised
352 deposition processes reported in section 2.

353 For all IL coatings, it is observed that a significant reduction in surface roughness is
354 achieved when compared to the initial roughness of the substrate. Figure 3 shows the
355 measured average roughness (R_a , Figure 3(a)) and peak-to-valley roughness (R_z , Figure
356 3(b)) for all the systems, denoting a clear reduction of these parameters when the IL are
357 deposited. Based upon results conducted for this paper, we estimate that a reduction of R_a
358 to $<60\text{nm}$ and R_z below $<300\text{nm}$ is needed for high performing a-Si:H and OPV solar cell
359 deposition. However, the performance of OPVs seem more sensitive to high roughness,
360 so in this case SU8 represents the best choice since this coating material has led to
361 $R_a < 15\text{nm}$ and $R_z < 90\text{nm}$.

362 Concerning dielectric isolation, Table II summarizes the obtained results for all the
363 steel/IL systems in terms of leakage current after the voltage breakdown test. As it can be
364 seen, leakage currents are below 50nA/cm^2 for all the samples.

365 Further studies were conducted to evaluate whether the IL layers limit the impurity
366 diffusion from the steel into the PV device. Figure 4 shows qualitative depth profiles of
367 the steel substrates coated with the SiO_x sol-gel IL and an Al electrode to replicate the
368 back contact of a solar cell. This electrode has been deposited in order to study reverse
369 diffusion from the back contact through the SiO_x layer, and potentially into the solar cell.

370 For each steel/ SiO_x /Al system, a comparison between a non-annealed and an annealed
371 process has been carried out. The annealing process was dictated by the thermal processes
372 needed for the TFSC technologies considered in this work; therefore, the time-
373 temperature cycle (150min at 210°C) for a-Si:H has been selected as the annealing cycle
374 because it represents the most demanding annealing condition.

375 For each qualitative depth profile, the Al layer shows an abrupt interface with the SiO_x
376 coating. As the sputtering time increases, there is an increase in the silicon and oxygen
377 content; an increase in carbon is also observed, owing to the PEG releasing agent that has
378 not been fully degraded at the 210°C curing temperature.

379 Related to the DC01, DX51D+AS and AISI430 substrates, it is clearly observed that no
380 steel element (Fe, Cr) and Al/Si elements from the metallic coating are diffusing into the
381 SiO_x sol-gel, demonstrating that this IL limits the diffusion of unwanted impurities from
382 the steel into the PV structure. Concerning the DX51D+Z substrate, a minor diffusion of
383 the zinc is present, although this is unlikely to interfere with the Al back contact due to
384 the low intensity and penetration depth. It is also observed that no diffusion of Al into the
385 steel substrates is apparent, so it appears that the SiO_x sol-gel layer does act as an ionic
386 blocking layer in both directions. These qualitative in-depth profiles also show the
387 levelling effect of the SiO_x coating. All the profiles possess an abrupt interface between
388 the Al and SiO_x, but the corresponding SiO_x/steel interface is extended indicating a much
389 rougher surface finish on the steel.

390 The anti-diffusion behaviour of the organic coatings, i.e. the *BlueInk*, has been also
391 studied under the same experimental conditions as above. For this coating, the elemental
392 identification is much more complex due to the highly insulative behaviour of the coating
393 which limits the signal into the mass spectrometer until the depth profiling has reached
394 the steel substrate. However, it is possible to infer from the data that no elements from
395 the steel are diffusing through the dielectric, which is evidenced by the paucity of signals
396 detected in between the steel and surface, demonstrating that the *BlueInk* also acts as a
397 good anti-diffusion barrier. For SU8, these characterizations have not been carried out
398 due to the high charging as a result of the highly insulative properties of the layer.

399 **3.2 Dielectric behaviour of ILs**

400 The dielectric behaviour of the ILs has been evaluated by measuring the leakage current
401 and by detection of a breakdown potential. Through breakdown tests, it was shown that
402 the dielectric coatings (sol-gel, *BlueInk* and SU8) passed well above the threshold of 40V.
403 In the case of sol-gel samples, values of 53V were obtained. *BlueInk* coatings showed
404 breakdown potentials above 500V. As qualitative test, the leakage current was measured
405 by voltage sweeping from -40V to +40V in steel/IL systems with evaporated silver or
406 aluminium electrodes. Table II shows the achieved values for all the tested samples. This

407 supports the breakdown tests confirming that these ILs are suited as electrical insulator
408 layers for BIPVs on steel substrates, with few pin holes present in the films.

409 **3.3 Photovoltaic cells**

410 Solar cells (1cm^2) were manufactured on all four types of steel and three different IL
411 coating combinations. This was undertaken for both a-Si:H solar cells and OPVs. The
412 results are summarised in Table III and example current density-voltage (JV)
413 characteristics are shown for solar cells manufactured on SiO_x -coated AISI430 in figure
414 5 under AM1.5G illumination. It is worth noting that the efficiencies of both a-Si:H solar
415 cells and OPVs are slightly lower than the state of the art in this field. The reason for this
416 is twofold; firstly the solar cells are made on relatively large areas (1cm^2) so are more
417 representative of module performance; secondly the device processing could be further
418 optimised for this configuration.

419

420 **3.3.1 Performance of a-Si:H cells on IL-coated steel**

421 Solar cells based on a-Si:H were successfully deposited in steel/IL systems with SiO_x sol-
422 gel as the IL. As shown in Table III, a relative difference of less than 10% was found
423 when compared to a solar cell made on the reference (glass) substrate using the same
424 experimental procedure. These results confirms the suitability of the IL/steel substrate for
425 a-Si:H solar cell integration. Although this result had been achieved before [9], it is
426 important to remark that, in this case, the layer was deposited from an upscale process
427 using a single spray step and a high speed curing cycle, instead of a three dip coating
428 processes followed by a 1h30 thermal treatment.

429 In the case of organic coatings (*BlueInk* and SU8), the steel/IL systems were firstly
430 introduced in a PECVD high vacuum chamber in order to evaluate possible desorption of
431 organic compounds that could induce future delamination of the solar cells. When the
432 steel/IL systems with the *BlueInk* IL was introduced into the vacuum system, a loss of
433 one order of magnitude in vacuum pressure resulted (2.1×10^{-6} Torr instead of the base
434 pressure of 2.0×10^{-7} Torr). A high vacuum treatment at 180°C was carried out for 2 hours
435 within the chamber. This thermal treatment allowed the improvement of the vacuum in
436 the PECVD chamber to values close to 3.1×10^{-7} Torr, which is within the limits of the
437 deposition window. Subsequently, the a-Si solar cells were deposited using the same

438 experimental procedure as for those made onto the SiO_x sol-gel examples. As shown in
439 Table IV, it has been possible to manufacture working cells. However, the obtained
440 efficiencies are around 1.67%, much lower than the efficiencies corresponding to the
441 glass reference. This result proves the viability of *BlueInk* as an IL for a-Si:H deposition,
442 although the roughness appears to limit the values of efficiency, in particular showing a
443 relatively low fill factor and poor shunt resistance.

444 Steel/IL samples using the SU8 IL layer were also introduced into the PECVD high
445 vacuum chamber. However, a high desorption was registered just after the sample was
446 introduced into the PECVD chamber (the pressure rose to 1.2x10⁻⁵ Torr from the limit of
447 2.0x10⁻⁷ Torr). As a result of the poor desorption properties, it was judged that the SU8
448 epoxy was not suitable for a-Si:H TFSC.

449 **3.3.2 Performance of OPVs on IL-coated steel**

450 Table III reports the power conversion efficiencies achieved for OPVs on the four steels
451 coated with SiO_x sol-gel IL by spray coating. It is evident that the efficiencies achieved
452 on steel (2.00-2.40%) are lower than achieved on a standard OPV fabricated on glass
453 (2.90%). The results provide a comparison of the same active layer material system, but
454 very different architecture owing to the different substrates and different direction of
455 irradiation onto the cell. Despite the lower efficiency, the result highlights the
456 compatibility of OPV onto steel substrate with a scalable SiO_x sol-gel IL. Glass is a low-
457 cost and slightly higher performing option but is not R2R compatible which is a
458 disadvantage for OPVs, as most groups tend to fabricate OPVs from solution processing
459 on R2R processing lines. In the case of the SU8 IL layer, the efficiency achieved is
460 relatively similar to that achieved on the SiO_x IL (2.42%). OPVs are unlikely to be made
461 commercially onto glass substrates, as glass adds a limitation to two of the key advantages
462 which the OPV technology provides: flexibility and low embodied energy per module.
463 Therefore, a more commercially relevant comparison of OPV performance is conducted
464 relative to substrates such as polyethylene terephthalate (PET) or polyethylene
465 naphthalate (PEN), as is shown in Table IV. In comparison, the performance of OPVs on
466 PEN and PET is 2.31% and 2.2%, respectively. By comparison, OPVs made on steel have
467 an intermediate efficiency; they are not better than those made on glass, but appear to
468 work better than those manufactured on plastic substrates. Concerning organic coatings,
469 the use of *BlueInk* led to unsuccessful results for the manufacturing of OPVs. This is

470 likely to be due to the high surface roughness of this layer, which this group estimated as
471 being twice that required for the deposition of OPV.

472

473 **3.4 Economic viability**

474 A cost analysis taking into account both the SiO_x sol-gel (spray process) and the *blue ink*
475 (screen printing process) ILs was finally performed in order to report the economic
476 viability of the Steel/IL prototypes. The detailed study of the economic feasibility of both
477 IL is reported in a previous work [26]. In this work, the process has been extended to
478 ascertain the costs involved in manufacturing a new BIPV product adopting a bottom up
479 approach in alignment with Anderson [27], where each of the individual cost elements
480 (materials, manufacturing fixed costs, plant capacity and operations/maintenance) were
481 identified and included within the cost model. The cost analysis assumes the utilisation
482 of a modern R2R production facility with a manufacturing plant capacity of 30 MW/year
483 [28] to assure the most efficient and cost-effective means to develop the BIPV products.

484 In that sense, a feed rate for both IL materials has been estimated at 12m/min, taking 6.9
485 hours to process a 1500m² roll, or 2.3 hours for a 500m² roll (roll size dependant on
486 material thickness). After estimations of the consumable investment (0.12€/m² for *blue*
487 *ink* and 0.23€/m² for SiO_x sol-gel), the labour (0.52€/m²), the electricity (0.15€/m²) and
488 the materials costs including wastage (1.80€/m² for *blue ink* and 0.33€/m² for SiO_x sol-
489 gel), total processing costs of 2.60€/m² for Blue Ink and 1.24€/m² for SiO_x sol-gel were
490 calculated.

491

492 **4 Conclusions**

493 The development of electrically insulative, ionic blocking, planarising ‘intermediate
494 layers’ (ILs) opens the possibility of using low-cost steels for the deposition of TFSC and,
495 in addition, allows for monolithically interconnected cells on this substrate. The results
496 have been achieved using a wide range of low cost steels (bare low carbon, hot-dip low
497 carbon and ferritic stainless) using an inorganic sol-gel coating and two commercial
498 organic materials, a dielectric ink and a photoresist.

499 In the case of sol-gel IL layers, a-Si:H solar cells have been manufactured with
500 efficiencies ($\eta = 4.59\% - 5.56\%$) close to those achieved on a reference glass substrate (η
501 $= 5.51\%$). The sol-gel/steel combination has been used as a substrate for OPV
502 manufacturing, obtaining efficiencies between 2.00% and 2.40%, marginally lower than
503 the efficiency obtained onto glass ($\eta = 2.90\%$). These differences have been attributed to
504 surface roughness of the coating ($R_a = 30\text{nm} - 60\text{nm}$) and different architecture of the
505 device.

506 When organic coatings have been considered, SU8 coatings show a significant
507 planarizing effect, with mean average roughness below 20nm. This enabled the
508 deposition of OPVs on steel/SU8 systems with high efficiency ($\eta = 2.29\% - 2.42\%$) when
509 compared to a flexible substrate reference (PEN, $\eta = 2.31\%$). In the case of a-Si:H,
510 desorption of this coating under initial vacuum conditions has not allowed the deposition
511 of these cells. *BlueInk* organic coatings represent an alternative approach to obtain high
512 dielectric coatings by means of simple and inexpensive deposition processes (in
513 particular, screen printing). However, the results show that further improvements in terms
514 of planarity and roughness are still required for the deposition of TFSCs. In the first case,
515 the a-Si:H cells show low fill factor ($FF=30\%$) and, consequently, low efficiencies when
516 compared to glass standard.

517

518 **Acknowledgements**

519 The research leading to these results has received funding from the European Union's
520 Research Fund for Coal and Steel (RFCS) research programme under grant agreement
521 no. RFSR-CT-2014-00014.

522

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603

604 **Table captions**

605 **Table I.** Chemical composition of selected steel substrates samples used for this
606 work

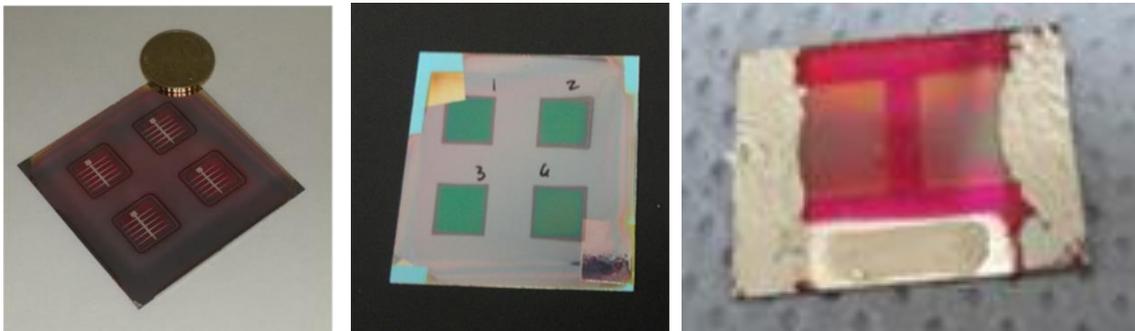
607 **Table II.** Leakage currents (in nA) corresponding the steel/IL systems developed in this
608 work

609 **Table III.** Photovoltaic performance parameters obtained from illumination under
610 1 Sun AM1.5G spectrum for the a-Si:H and OPV cells on different steel/IL
611 systems

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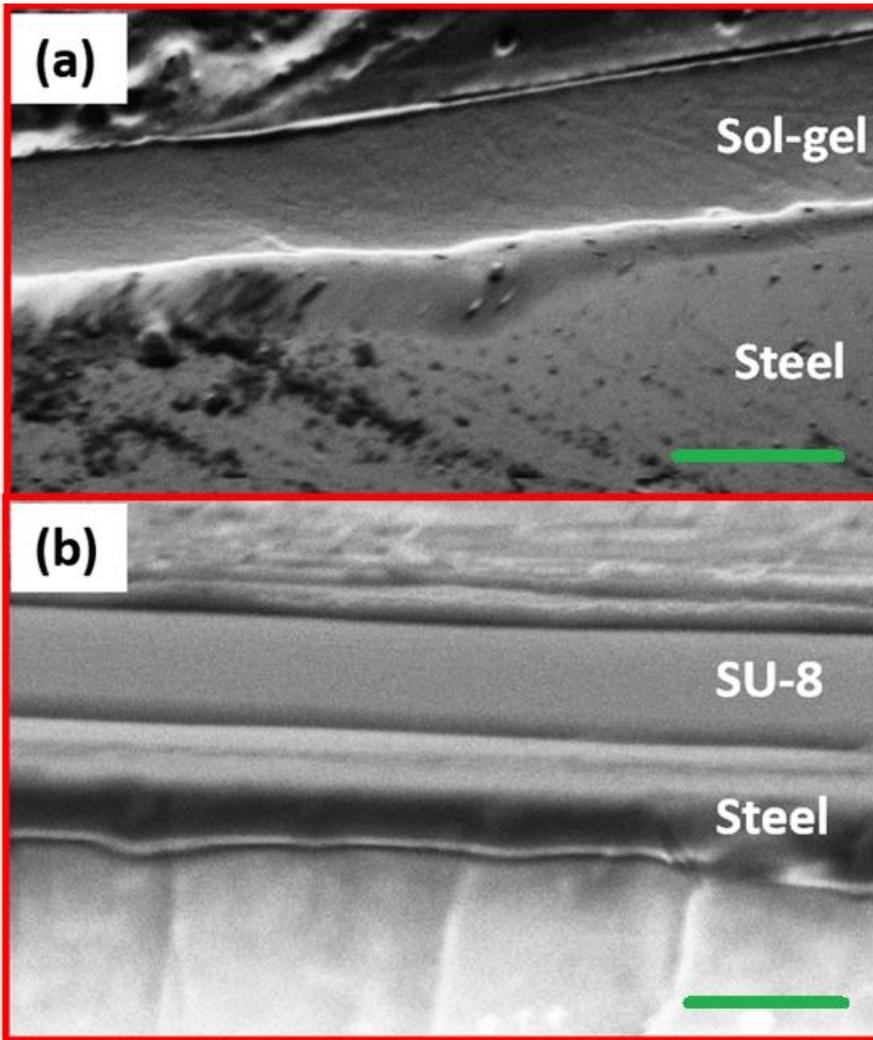
(a)

(b)

(c)

617 **Figure 1.** (a) a-Si:H solar cells deposited on AISI430/SiO_x sol-gel (b) onto BlueInk and
618 (c) Organic photovoltaic deposited onto AISI430/SU8

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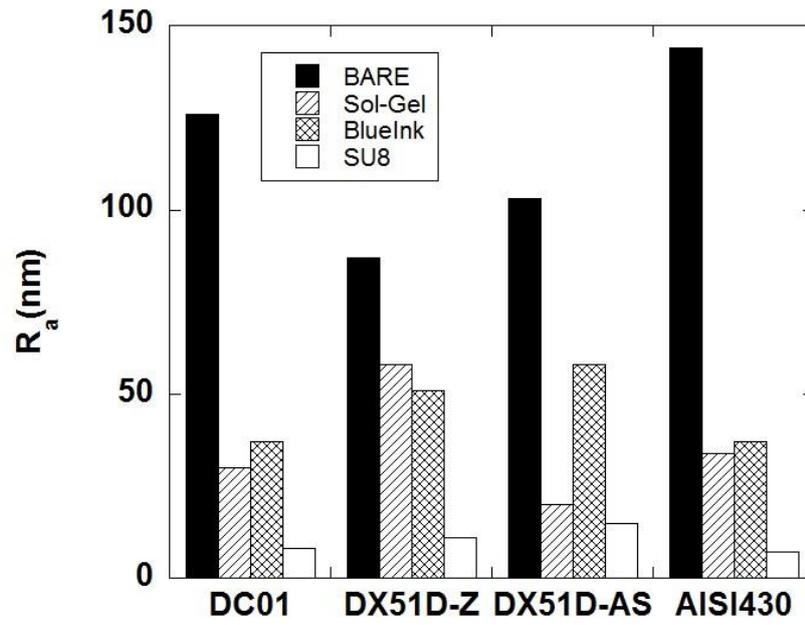


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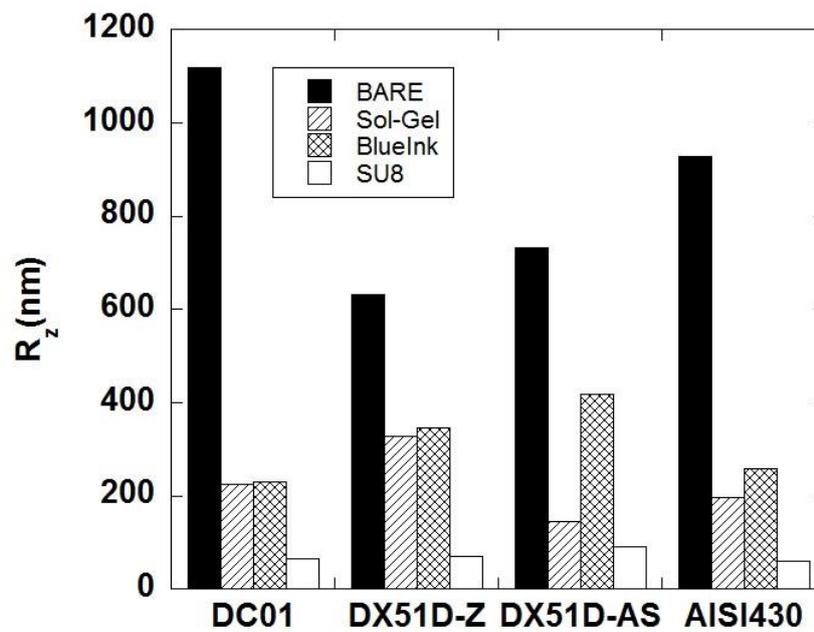
621 **Figure 2.** FE-SEM cross sectional image of AISI430 steel samples coated with (a) SiO_x
622 sol-gel (scale bar = $3\mu\text{m}$) and (b) SU8 intermediate layers (scale bar = $60\mu\text{m}$)

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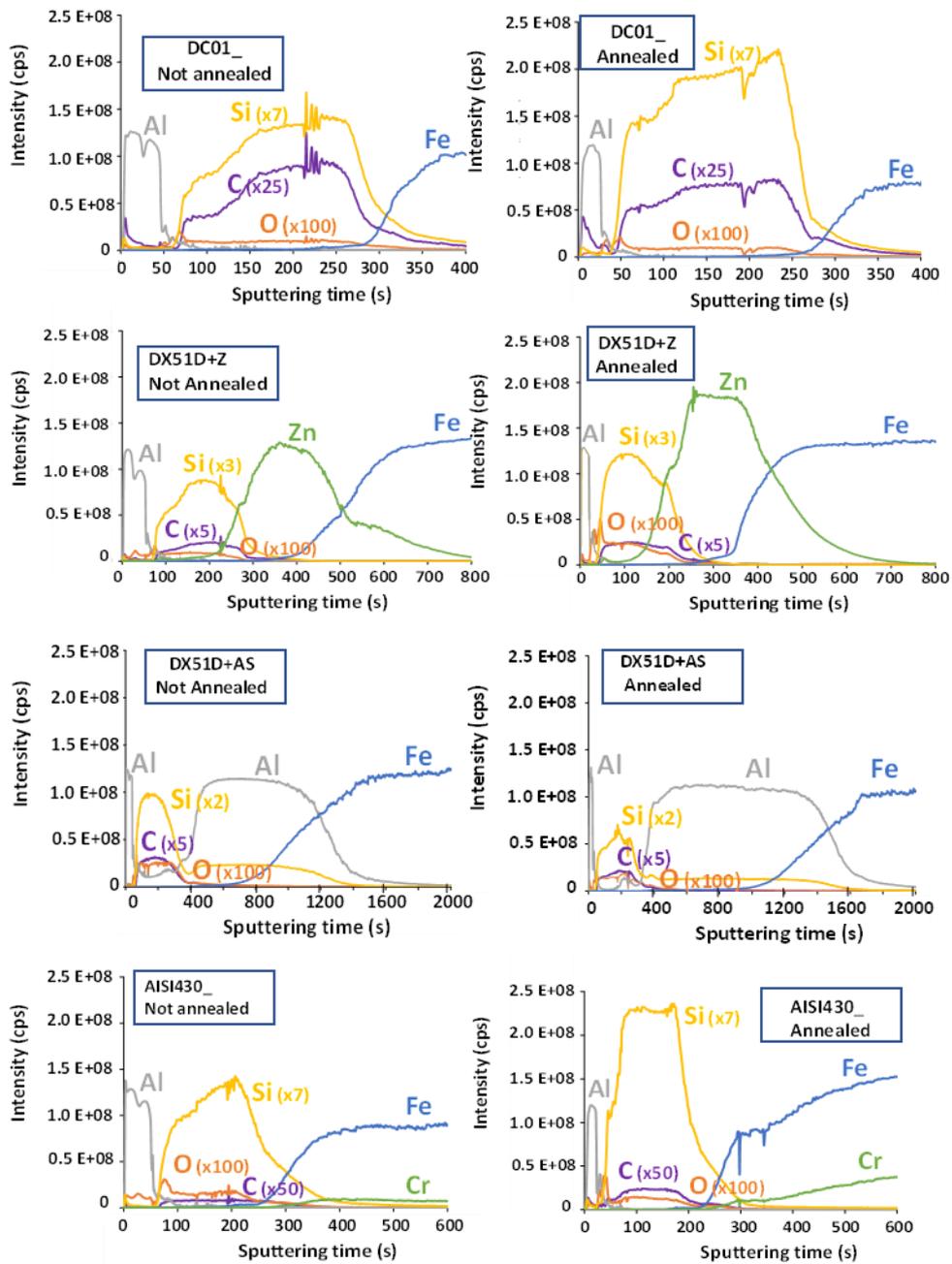


(a)



(b)

Figure 3. (a) Mean (R_a) and (b) peak-to-valley (R_z) roughness of bare selected steels and steel/IL systems developed in this work

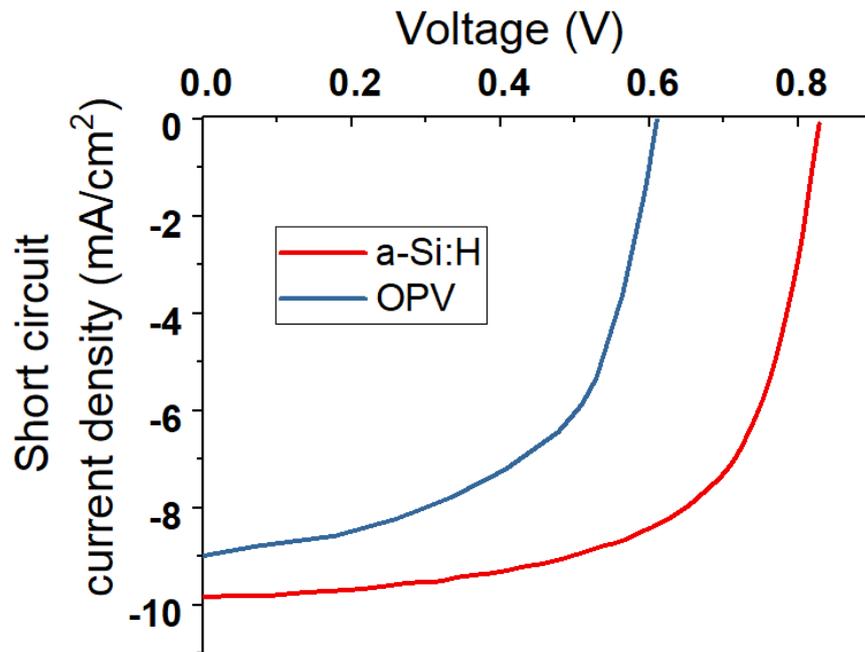


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634 **Figure 4** Qualitative depth profiles obtained using glow discharge time of flight mass
 635 spectroscopy (GD-ToF-MS) of non annealed and annealed for all steels/SiO_x sol-gel
 636 samples used in this work, with Al top electrodes to replicate the structure of the solar
 637 cell. The SiO_x layer shows extremely good barrier layer performance.

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641 **Figure 5.** Sample J–V curves of solar cells using data of a-Si:H solar cells and
 642 OPV onto AISI430 substrates after a SiO_x sol-gel IL has been applied

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Table I. Chemical composition of selected steel substrates

Material	Chemical composition %(wt.)						
	C	Si	Mn	P	S	Cr	Ti
#01 Bare Steel (DC01)	≤ 0.12	≤ 0.3	≤ 0.6	≤ 0.045	≤ 0.045	-	-
#02 Hot-dip zinc coated (DX51D+Z) ¹	≤ 0.18	≤ 0.5	≤ 1.2	≤ 0.12	≤ 0.045	-	≤ 0.3
#03 Hot-dip Al-Si coated (DX51D+AS) ²	≤ 0.18	≤ 0.5	≤ 1.2	≤ 0.12	≤ 0.045	-	≤ 0.3
#04 Stainless Steel (AISI430)	≤ 0.08	≤ 1.0	≤ 1.0	≤ 0.04	≤ 0.03	16.0-18.0	-

648 ¹Hot-dip Zn: Zn≥98%

649 ²Hot-dip Al-Si: Al 85.5%-88.5% + Si 9%-11% + Fe 2.5%-3.5%

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Table II. Leakage currents (in nA/cm²) corresponding the steel/IL systems developed in this work

Material		Leakage current (nA/cm²)					
		Sol-gel (Ag)	Sol-gel (Al)	BI (Ag)	BI (Al)	SU8 (Ag)	SU8 (Al)
#01	DC01	35.17	35.00	-	-	27.29	41.58
#02	DX51D+Z	36.29	36.25	-	-	18.96	30.08
#03	DX51D+AS	31.00	30.83	-	-	16.17	23.13
#04	AISI430	25.92	29.04	-	-	11.54	12.46

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Table III. Characteristic parameters obtained from current-voltage curves corresponding a-Si:H and OPV cells on different steel/IL systems. Averages are taken from 6 devices for all reported in this work and standard deviations are also reported

		a-Si:H*				Organic PV			
		Jsc (mA/cm ²)	Voc (mV)	FF (%)	η (%)	Jsc (mA/cm ²)	Voc (mV)	FF (%)	η (%)
SiO x SG	Reference (glass)	11.03 ±0.35	813±3	55±1	5.51± 0.21	9.88 ±0.20	590 ±2	59 ±4.0	2.90 ±0.34
	DC01	10.62 ±0.94	839±4	56±4	5.56± 0.16	8.46 ±0.30	560 ±5	45 ±4.2	2.15 ±0.46
	DX51D+Z	10.22 ±0.55	832±9	58±4	5.41± 0.12	8.21 ±0.40	550 ±3	44 ±4.9	2.00 ±0.27
	DX51D+A	10.65 ±0.21	835±7	47±2	4.59* *±0.1	8.70 ±0.40	550 ±3	47 ±2.9	2.23 ±0.25
	AISI430	9.92± 0.61	820±17	61±3	5.53± 0.26	9.20 ±0.50	530 ±3	48 ±6.0	2.40 ±0.52
	BI BI	DC01	-	-	-	-			
	DX51D+Z	-	-	-	-				
	DX51D+A	-	-	-	-				
	S	-	-	-	-				
	AISI430	6.93± 1.12	750±82	29±1	1.67± 0.43* *	7.63 ±0.22	510 ±5	38 ±4.0	1.48 ±0.20
SU8	Reference (PEN)					9.01 ±0.18	570 ±2	45 ±5.0	2.31 ±0.30
	DC01					9.54 ±0.19	546 ±10	55 ±5.8	2.42 ±0.22
	DX51D+Z					8.90 ±0.20	546 ±8	47 ±6.0	2.29 ±0.31
	DX51D+A					9.01 ±0.29	540 ±10	47 ±3.0	2.30 ±0.20
	S					8.94 ±0.09	540 ±0.2	49 ±2.5	2.35 ±0.18
	AISI430								

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*a-Si:H area of 1cm² without silver grid and 0.9cm² with silver grid
**no silver grid Irradiance of 0.9W/cm² for the a-Si:H measurements

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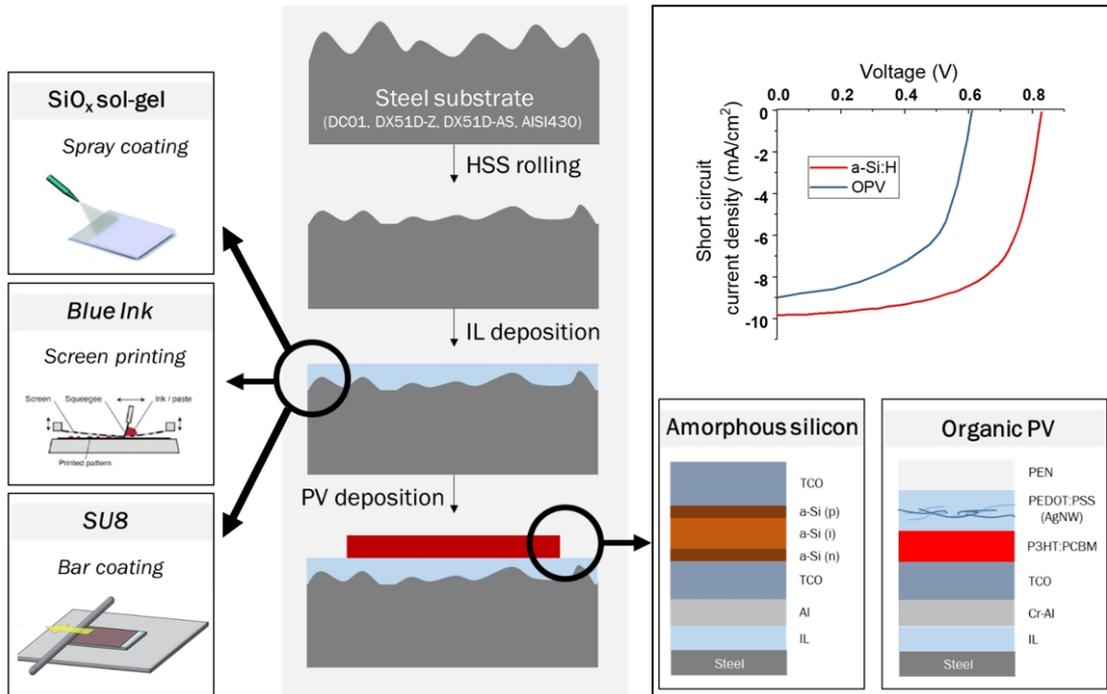
669

Graphical abstract

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