Reliability of electrically conductive adhesives

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Abstract

The application of electrically conductive adhesives (ECAs) is a promising alternative to the soldering process for cell interconnection in today's solar module production. ECAs provide an environmentally friendly solution and offer several other advantages over the conventional solder interconnection technology, such as lower processing temperature, higher mechanical flexibility and replacement of toxic lead. When it is proposed to switch from soldering to adhesive technology in a critical process such as the production of solar cell strings, it is necessary to perform a thorough preliminary analysis of the properties of the materials involved, the material compatibilities and the long-term stability of the interconnections within the PV modules. An investigation has therefore been conducted with regard to the performance, quality and reliability of: 1) isolated bonded joints using ECAs, and 2) the interconnections of ECA-bonded cells within the test modules. Moreover, new formulations with increased flexibility of the polymeric binder within the ECA have been developed in order to increase the resistance to thermomechanical loads and delamination. To better understand the relevant material interactions and the influencing factors, a comprehensive test plan was set up. A characterization of the pure adhesive with respect to the outgassing and migration behaviour of volatile low-molecular compounds was performed using thermogravimetric analysis (TGA) and thermodesorption/gas chromatography–mass spectrometry (TD/GC–MS). Single-cell test modules with different combinations of ribbons and encapsulation and backsheet materials were investigated to achieve the targets. The test modules were exposed to different combined stress factors in accelerated ageing tests, namely damp heat (DH), irradiation with sunlight, and thermal cycling (TC). Even though the screen printing of the ECA on the cell, as well as the module layering, was done manually, the results from the electrical characterization showed excellent reproducibility. All ribbon types (Ag coated, bare Cu, SnAgCu coated) could be processed in the PV module lamination without any problems. Upon accelerated ageing, slight power losses between 1 and 4% were measured for the ECA-connected one- and six-cell sample modules. ECAs are therefore a promising alternative to the soldering process used in cell stringing. Outgassing of molecular compounds was found to be low, and no material incompatibilities of the adhesive with the various types of ribbons and encapsulants were observed.

Introduction and objectives

Resin-based interconnection materials for electronic packaging and interconnection technologies are currently widespread in the fabrication of electronic devices, whereas they are only rarely used in crystalline silicon PV modules for the interconnection of cells.

Creating interconnections via the print application of electrically conductive adhesives (ECAs) offers several advantages over the conventional solder process for electrical interconnection, such as the possibility of low-temperature processing, the potential for higher-resolution printing and easier handling.

The soldering process requires temperatures of 210°C for conventional tin–lead solders, or even higher for lead-free solders. These high temperatures often cause cell breakage and the introduction of microcracks in the crystalline Si cells. The main limitation in the goal to reduce wafer thickness is therefore imposed by the soldering process. The curing reaction of ECAs, on the other hand, usually takes place below 180°C and can be tailored by modifying the basic polymeric binder. Thus, switching to an adhesive interconnection technology allows further reductions in wafer thickness and opens the door to innovative possibilities in cell design. Another advantage is that the adhesives can be applied by screen printing directly onto the finger grid of the cell, without using additional busbars on the front side of the cell.

Compared with lead-based solder alloys, the use of ECAs is an environmentally cleaner solution for interconnection tasks [1]. By replacing the toxic lead-containing solders, the accompanying challenges concerning waste management and recycling can be avoided. Furthermore, the possibility of using non-solderable materials – such a silver-coated ribbons, which are used as light-capturing ribbons – opens up new possibilities in novel cell and PV module designs. Nevertheless, the replacement of the soldering process by ECAs also has some limitations. A major drawback is the high silver price, which is why the highly filled adhesives are much more expensive than solders; this can be partially offset by cutting down on the number of busbars. Another challenge is the ability to withstand harsh environmental conditions in certain climate zones, where some issues with limited impact resistance, weakened mechanical strength and increased contact resistance (when unsuitable ribbon coatings are chosen) have been observed.

Generally speaking, ECAs are composite materials based on a conductive filler and an insulating polymeric adhesive. Here, thermosetting as well as thermoplastic resins can be used as the matrix material; epoxy resins, silicones or polyurethanes are widely used thermosets, while polyimides are typical examples of thermoplastic resins used in ECAs. Among the conductive fillers, silver (Ag) is

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the most commonly employed; it has the highest electrical conductivity, with an ability to retain its high conductivity, even when the silver particles are oxidized. However, copper coated in gold (Au), nickel (Ni), copper (Cu), tin (Sn), SnBi or SnIn, in various sizes and shapes, is also found in applications as a filler material [2].

Depending on the loading level and type or shape of the electrically conductive filler, ECAs are divided into isotropic conductive adhesives (ICAs) and anisotropic conductive adhesives (ACAs). Because of their high filler content (50–80 wt%), ICAs provide an electrical conductivity in all directions throughout the material (x, y and z directions). Here, the resin is generally cured at higher temperatures to provide the shrinkage force to increase the conductivity, adhesion strength and chemical and corrosion resistance. ICAs are commonly used to replace the traditional SnPb solder alloys in electronic interconnections. In contrast, ACAs provide conductivity only in the vertical direction; this is achieved by using very low amounts (5–20 vol%) of conductive filler with a spherical shape in the adhesive [2–4].

An important property to consider for the use of ECAs in PV modules is the fracture toughness of the cured resin. With regard to high-efficiency cell concepts and reduced cell thicknesses, the consideration of mechanical straining is essential. Pander et al. found that the application of ECAs in silicon solar cells yields a reduction in strain within the silicon compared with the solder route [5]. There are several possible ways of achieving a more flexible (less cross-linked) network in the cured adhesive, for example the use of reactive diluents, which are basically monoepoxide compounds that can react with the curing agent to become part of the cross-linked epoxy system. Another possibility is to use long-chain hardeners to reduce the effective cross-linking density in order to achieve less tight networks [6–10].

The main objective of the work presented in this paper was to investigate the performance, quality and reliability of different types of electrically conductive adhesives, as well as test modules with ECA-connected cells. Of special interest were:

1. Outgassing behaviour of the cured ECA.
2. Material interactions with the encapsulant.
3. Characterization of fatigue behaviour.
4. Performance of ECA-bonded test modules.

Single-cell modules were used for the studies of material interactions after stress impact, such as temperature, humidity and irradiation, as well as for sequential irradiation-humidity tests, whereas six-cell modules were utilized for thermal cycling (TC) tests.

Name	Resin type	Hardener	Mixing ratio
ECA 1	Epoxy based on novolak, type A	Hardener 1	100:11
ECA 2	Epoxy based on bisphenol A/F, type B	Hardener 1	100:10
ECA 3	Epoxy based on bisphenol F, type C	Hardener 1	100:10

Table 1. ECAs under investigation.

Name	Type	Cross-linking	Formation of acetic acid
E1	Ethylene vinyl acetate (EVA)	Yes	Yes
E2	Thermoplastic polyolefin (TPO)	No	No
E3	Polyolefin elastomer (POE)	Yes	No

Table 2. Encapsulation materials used for test modules.

Name	Type	Coating	Solderable
R1	Silver	Ag	No
R2	Bare copper	-	No
R3	Lead free	SnAgCu	Yes

Table 3. Ribbon materials used for test modules.

Type	Duration	Temperature	Humidity	UV dosage	Remarks
Damp heat	1,000h; 2,000h; 3,000h	85°C	85% RH	No	
Irradiation	1,000h; 2,000h	50°C	< 50% RH	120W/m^2 (300–400nm) Metal halide lamps (300–2,500nm)	
Sequential	S1: 330h S2: 8h S3: 8h	85°C 50°C 30°C	85% RH < 50% RH 85% RH	No 120W/m^2 (300–400nm) No	Test procedure: S1 + 40× (S2 + S3)

Table 4. Accelerated ageing tests.

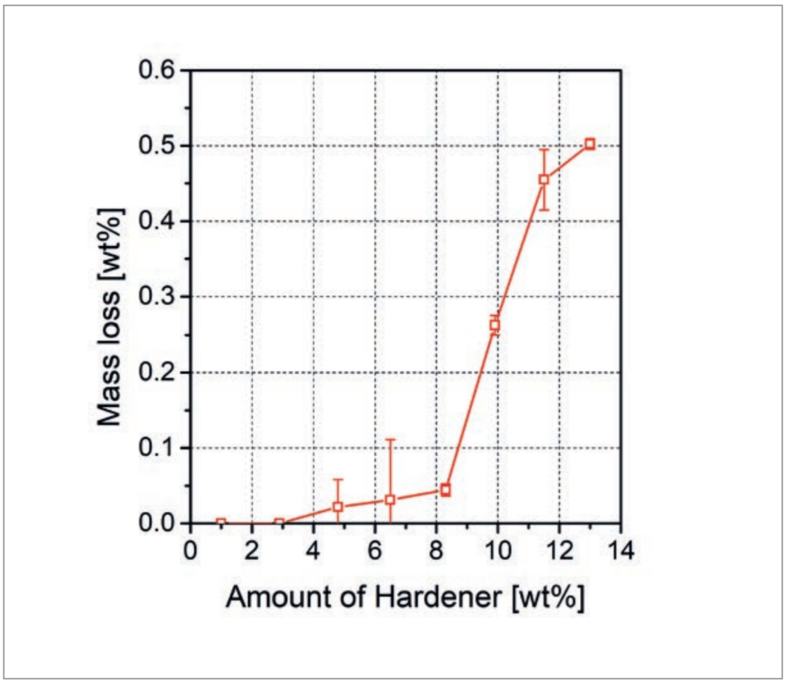

Figure 1. Percentage mass loss of the cured adhesives of type ECA1 with varying hardener content, received from TGA measurements, at a constant temperature of 150°C for 30 min.

Experimental

A main aim of the studies was to investigate the compatibility of ECAs with other module materials, especially different encapsulation and ribbon materials, and to describe possible new ageing-induced failure modes in PV modules using ECA interconnections.

First, the outgassing behaviour of three different ECA formulations (ECA1, ECA2, ECA3) comprising different epoxy components was investigated; the details are given in Table 1. Additionally, the fatigue resistance of ECA1 and ECA3 was investigated by performing cyclic fatigue tests.

In the next step, possible interactions with different encapsulation materials were investigated by printing the three different ECA formulations on glass and encapsulating them with three different encapsulation materials (EVA, TPO, POE) and a cover glass.

Finally, the performance, ageing behaviour and ageing-induced material interactions were investigated in test modules with different combinations of ECAs, ribbons and encapsulation films. As the reference, a soldered test module with

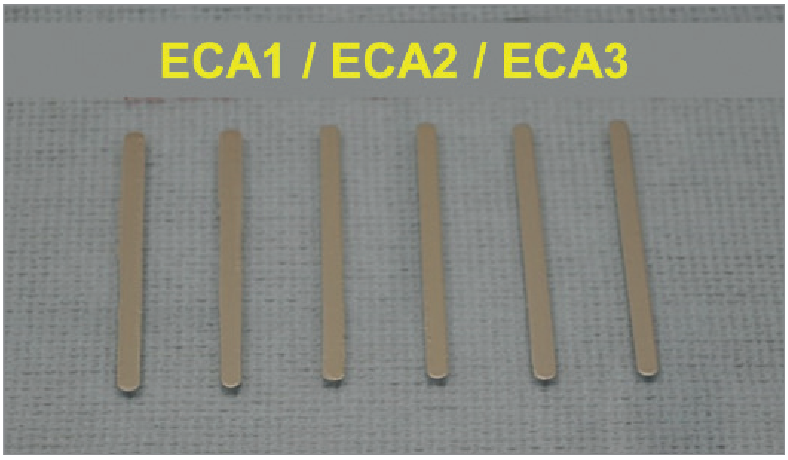

Figure 2. Printed ECA lines on glass substrate and laminated with encapsulants and glass covers.

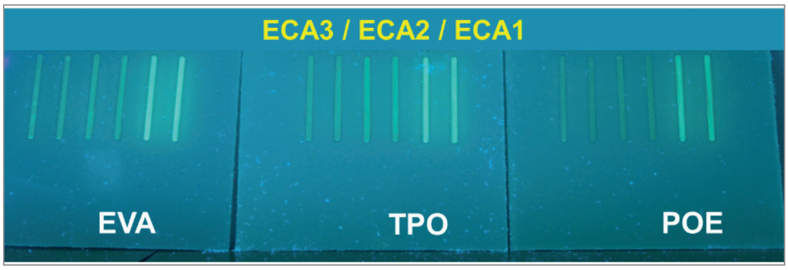

Figure 3. UV fluorescence image of printed ECA specimen (encapsulated) after lamination.

> "The lamination process and accelerated ageing do not induce any distinct harmful interactions or degradation modes between the investigated ECA and encapsulant types."

a standard EVA encapsulation Cu/SnPb ribbon was used. Three different encapsulation materials (see Table 2) were chosen in order to investigate in particular the effect of acetic acid (which is a degradation product of EVA) and the added cross linker. In addition, three different ribbon types were used for the module tests (see Table 3). The accelerated ageing tests are summarized in Table 4.

Results and discussion

The main objective was to investigate the performance, quality and reliability of the adhesive bonds, but also of the test modules with ECA-bonded cells.

Since the outgassing behaviour of adhesives applied in PV modules is of great importance, a number of volatile products of the cured adhesives generated at 150°C using thermogravimetric analysis (TGA) were investigated. The temperature of 150°C was chosen because this is the temperature applied during the lamination process for PV modules.

The values of mass loss received from TGA measurements of the different formulations of ECA1 (with varying amounts of hardener) are shown in Fig. 1; it can be seen that the mass loss at 150°C decreases with decreasing amount of hardener. The mass loss decreases steadily, and the formulation with the lowest amount of hardener approaches a mass loss of almost 0 wt%. This finding is in accordance with the results of thermodesorption/gas chromatography–mass spectrometry (TD/GS–MS) measurements, demonstrating that the thermal extractable compounds of the ECAs derive from the hardener. These outgassing products have not been found with formulations with a hardener amount of less than 6.5 wt%.

Possible interactions of the ECAs with the encapsulation materials EVA, TPO, POE were investigated (see Fig. 2). From a visual inspection, no migration of silver particles was observable. With the use of confocal Raman spectroscopy, no corrosion products containing Ag (e.g. silver acetate) were seen in the encapsulant close to the ECA and the ribbon. It can therefore be concluded that the PV module lamination process did not induce a diffusion of Ag particles into the encapsulant.

Fig. 3 shows the UV fluorescence image of the specimens for different encapsulation materials. Because of the various chemical compositions of the ECAs (different polymeric epoxy component – see Table 1), the fluorescence signals of the ECAs were different, with ECA1 showing the highest fluorescence.

A comparable UV fluorescence was also observed in the area next to the ribbon after accelerated ageing tests for the single-cell modules using ECA1 and POE as the encapsulant. This indicates migration of the hardener, which was not converted in the curing reaction, into the surrounding encapsulation material. No correlation with power loss of the test module, however, was observable. In the case of the other ECA encapsulant combinations, no migration effect was observed. It is therefore assumed that the lamination process and accelerated ageing do not induce any distinct harmful interactions or degradation modes between the investigated ECA and encapsulant types.

The fatigue resistance of ECA1 and ECA3 was investigated using a single lap shear (SLS) specimen. Cyclic fatigue tests were performed under a tensile load at a frequency of 10Hz and a stress ratio *R* of 0.1. (The stress ratio is defined as the ratio of minimum stress to maximum stress in one cycle of loading in a fatigue test.) Tensile stresses are considered positive and compressive stresses negative. Mean stress levels (between 5 and 20MPa) were derived from tensile tests using the SLS specimen.

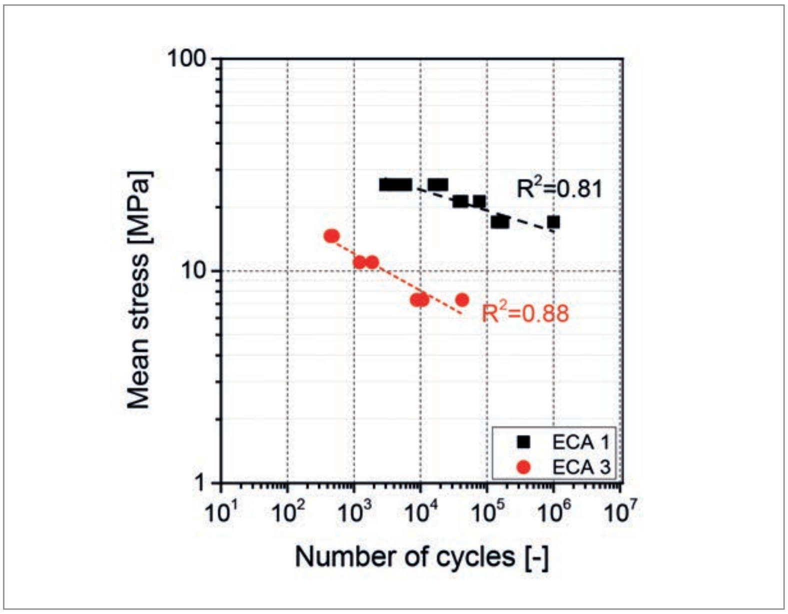

Figure 4. S–N curves for the investigated ECA types.

Fig. 4 shows the S–N curves (also known as *Wöhler curves*) of the investigated adhesives. ECA 1 exhibits significantly better fatigue resistance than ECA 3. One explanation for this can be found in the intrinsic fatigue resistance of the materials; however, the nature of the sample preparation may also be a contributing factor to the lower fatigue resistance. In the investigation of the fractured surfaces of ECA 3, bubbles and a lower degree of curing were detected, which could have strongly influenced the fatigue behaviour.

Only a limited number of papers dealing with the fatigue behaviour of cell interconnection have been published so far [10–12], and these give contradictory values. Pander et al. [11] studied the fatigue of solar cell interconnections, and designed the loading profile during the fatigue test in such a way as to achieve the same strain amplitude in the cell gaps as that found in a full-size module simulation under ±1,000Pa, which corresponds to the IEC proposal. Dietrich et al. [10] also investigated fatigue in solar cell interconnections, and chose the test amplitude so that the failure occurs before 10,000 cycles. However, those authors did not give information on the load levels applied in their fatigue tests. Zarmai et al. [12] studied the thermomechanical damage and fatigue life of solar cell solder interconnections, and reported the calculated value of maximum stress concentration in the solder joint to be of the order of 21MPa. This value was obtained within the thermal cycling test in the temperature range –40°C to +85°C, in compliance with IEC 61215.

With regard to the cyclic fatigue behaviour of the investigated ECA types in the current paper, both S–N curves are either significantly above the mean stress levels that were reported for interconnections in PV modules [10,11], or in a similar range [12]. The reported values for the number of cycles to failure for soldered bonds are also in a similar range.

In a next step, a series of single-cell test modules was investigated. These modules consisted of

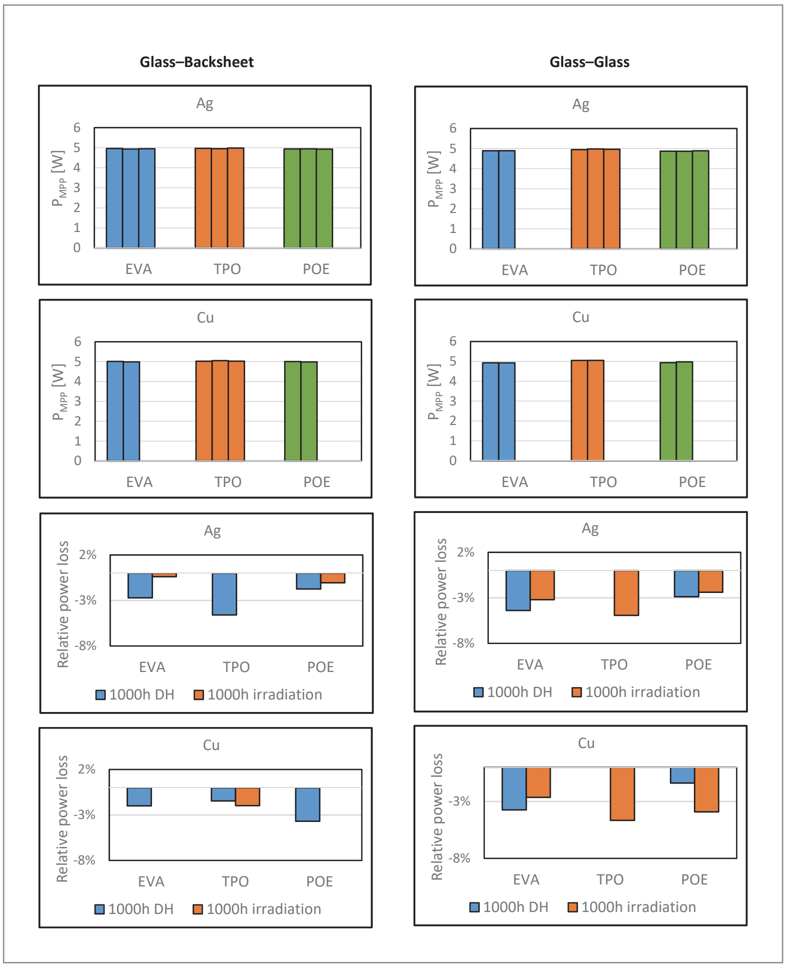

Figure 5. Electrical characterization results for test modules with different ECA-bonded ribbons (Ag, Cu) and different encapsulation materials (EVA, TPO, POE), before and after accelerated ageing (DH and irradiation).

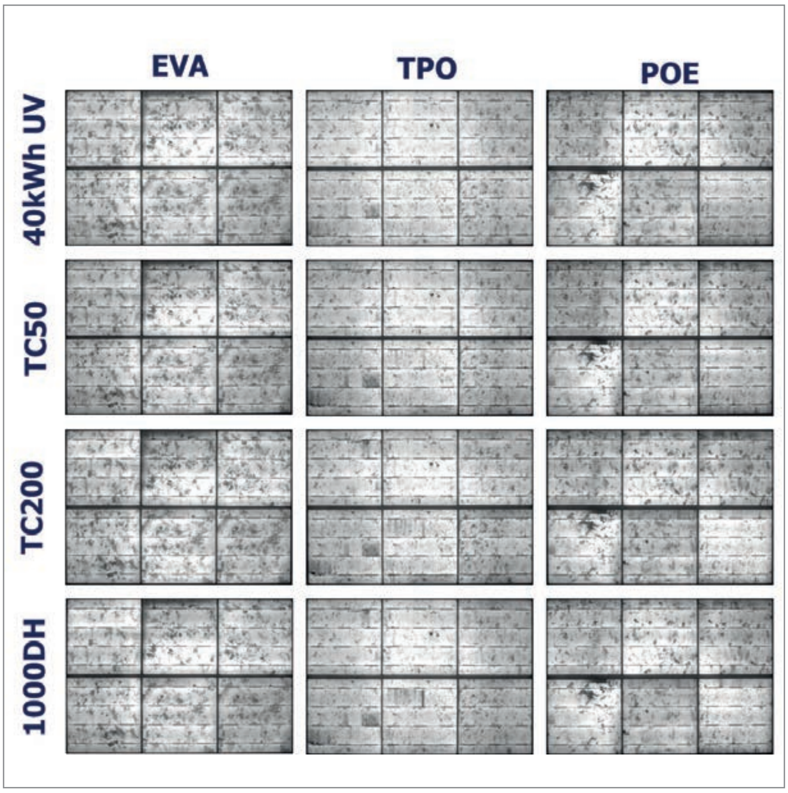

Figure 6. EL images of the six-cell modules with different encapsulation materials, before and after 200 thermal cycles.

identical Si cells, ECA1, front glass, polymeric backsheet (PET laminate) or glass backsheet, and various encapsulants (EVA, TPO and POE) and ribbons (Ag, Cu, SnAgCu) (see Experimental section).

A visual inspection revealed that during module production, ECA1 in contact with all tested ribbons types (Cu, Ag, SnAgCu) did not show any initial problems, such as delamination or discolouration. Fig. 5 presents the electrical characterization results of these test modules before and after accelerated ageing (1,000h damp heat – DH – and 1,000h irradiation). The results showed satisfying reproducibility, even though the strings and modules were fabricated manually. The single-cell modules exhibited maximum power output (P_{MPP}) values between 4.7 and 5W. The glass–glass modules showed slightly lower P_{MPP} values than glass–backsheet modules, which can be explained by the rear reflection of light from the highly reflective white backsheet onto the solar cell.

In regard to ribbon types and encapsulation films (and otherwise identical set-ups), the modules using bare copper ribbons yielded the highest power outputs, followed by the modules with Ag-coated ribbons. This can mainly be explained by the reduced ribbon width and the light-capturing effect. The lowest values were observed for SnAgCu-coated ribbons. The differences, however, were fairly small, at ±0.2%. Furthermore, the modules using TPO as the encapsulant demonstrated the highest P_{MPP} values, which can be explained by the slightly higher transparency of the TPO film than that of EVA or POE. It should be noted that, even though the power measurements were reproducible, with standard deviations below 0.5%, some of the observations may not be significant, since the measurement inaccuracy of the electrical measurements was around $\pm1.5\%_{\text{rel.}}$.

Test modules were then exposed either to 1,000h of damp heat or to 1,000h irradiation. The ageing-induced relative power losses were found to lie between 0.5 and $4.5\%_{\text{rel}}$. Missing values in the figures correspond to test modules that suffered glass breakage during the accelerated ageing test (untempered glass was used for the single-cell test modules). No visible changes – such as discolouration, delamination or cracking – were observable after the accelerated ageing tests. Generally, the glass–glass modules showed higher power loss than the glass–backsheet modules. Other correlations between stress factors (i.e. the ageing test), ribbon type and encapsulation films were not observed, partly because of missing values, and partly because of inconsistent test data. The exact cause of the power loss is therefore still unknown.

In order to investigate the mechanical stability of cell interconnections, six-cell modules were manufactured and subjected to thermal cycling tests. The modules were exposed to the following test sequence:

1. Preconditioning: 40kWh (AM1.5 in compliance with IEC 60904-3).
2. TC: 200 cycles from −40°C to +85°C (in compliance

with IEC 61215), with a first power measurement after 50 cycles.

3. Additional DH: 1,000h of exposure at 85°C/85%RH.

After this test sequence, the modules were characterized by visual inspection, power measurement and electroluminescence (EL) imaging.

Figs. 6 and 7 show the relative power loss and the EL images of the six-cell modules. The visual inspection did not reveal any delamination, while the EL images showed slight brightness differences and only a few crack propagations after the applied test sequences. The identified cracks already existed before the different tests were performed (see Fig. 6), and the reason for the brightness differences in the EL images are explained by the change in contacting properties of the strings. It is assumed that this damage was caused by production failures (e.g. manual handling of the ECA-bonded strings) and/or during transport.

The relative power losses of the six-cell modules (three identical modules for each module design, nine modules in total) after each test sequence are illustrated in Fig. 7. For all test modules, UV preconditioning resulted in a power loss of around 1%. Furthermore, the behaviour after 50 and 200 temperature cycles was similar for all identical test modules; power loss values of around 2 and 4% were measured after 50 and 200 cycles respectively. The *I*–*V* curves revealed a decrease in open-circuit voltage (V_{oc}) and in short-circuit current (I_{sc}), and an increase in serial resistance (R_s). The increase in R_s is an indication of an increased contact resistance between ribbon, adhesive and cell. Another explanation for the increased contact resistance could be the cross connector, which was manually soldered. Interestingly, an additional DH exposure led to power regeneration for modules with TPO and POE encapsulant. Generally, modules with TPO yielded the best performance.

Conclusion

The main objective was to investigate the performance, quality and reliability of ECAs, but also of test modules with ECA-bonded cells. Of special interest was 1) the outgassing behaviour of cured ECA; 2) the material interactions with encapsulants; 3) the characterization of thermomechanical and fatigue behaviour; and 4) the performance of ECA-bonded test modules.

No harmful interactions were found between the investigated ECA formulations and the different encapsulant films after lamination and ageing tests. The main outgassing products were identified as fragments of the hardener. In addition, no migration of silver particles was detected. ECAs were found to be compatible with all tested ribbons types (Cu, Ag, SnAgCu), since no delamination or discolouration after lamination or accelerated ageing tests was observed. For ECA-bonded test modules, a slight power loss after thermal cycling, damp heat and irradiance exposure was identified; this power loss can be attributed not only to failure of the ECA bond but also to additional factors such as sample preparation and cell damage that was present from the start. To summarize, cell interconnection using ECAs provides an interesting alternative to standard soldering approaches; however, additional work needs to be done, especially with regard to a root cause analysis of the power loss after thermal cycling.

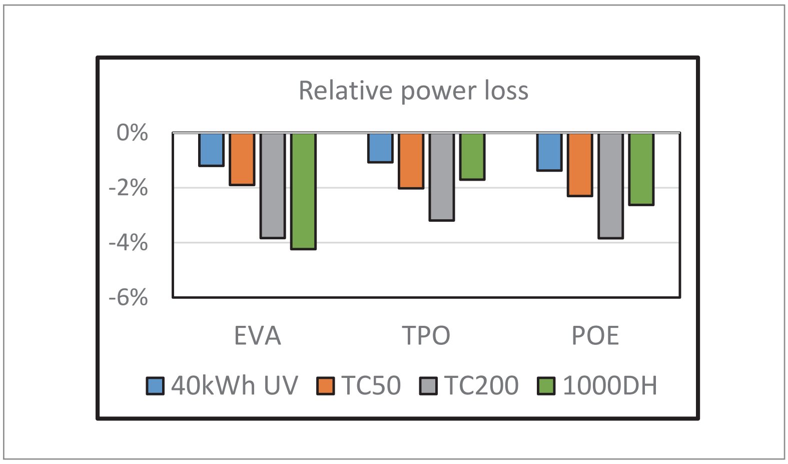

Figure 7. Relative power losses of the six-cell modules after each test sequence.

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