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Reduction of Silver Use in Low-Temperature Interconnection of Solar Cells

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Abstract. New solar cells' technologies such as silicon heterojunction and tandem solar cells raise new challenges such as temperature-induced degradations, which are problematic for the cells' interconnection. Electrically Conductive Adhesives (ECAs), usually made of silver particles embedded in a polymeric matrix, seem a promising technology to allow low-temperature interconnection, but raise concern about a high consumption of silver in the manufacture of photovoltaic (PV) panels. In this work, the fabrication of copper-based ECAs is presented as an approach to reduce the silver consumption of the interconnection process. Core-shell copper-silver particles with dendritic shapes that showed resistant to oxidation up to 220°C were used as conductive fillers. Their integration into an acrylate fast-curing matrix was performed in ratios of 59 wt% and 69 wt% fillers, respectively accounting for silver contents of 3.0 wt% and 3.5 wt%. The resulting pastes have resistivities of $3.5 * 10^{-3}$ Ω*cm and $9.0 * 10^{-4}$ Ω*cm respectively, globally similar to those of commercial ECAs dedicated to the interconnection of solar cells. Their fast-curing process was measured to occur upon heating at 105°C, and be completed upon heating at 160°C during 20 seconds, enabling high throughputs. First trials of these ECAs into strings of solar cells were made: the stencil-printed patterns had a good quality, but a poor adhesion between the solidified ECAs and the ribbons prevents their further use.

Keywords: Interconnection, Low-Temperature, Conductive Adhesive, Copper, Fabrication

1. Introduction

New solar cells such as silicon heterojunction (SHJ) and tandem cells enable high conversion efficiency but deteriorate when heated to high temperatures. Their interconnection is challenging due to limitations of the processing temperatures below 200°C for SHJ cells [1] and below 140°C for tandem cells [2].

Electrically Conductive Adhesives (ECAs), usually made of silver particles embedded in a polymeric matrix, seem a promising technology to allow interconnection at low temperatures [3]. However ECAs usually contain more than 50 wt% silver to ensure low electrical resistivity [4]. As they could participate to a high silver consumption in the photovoltaic (PV) manufacturing industry, efforts need to be made to reduce the silver content of ECAs. A first approach to reduce the silver consumption of ECAs consists in optimising the fillers used by tuning their shapes, size distributions, etc. However, the lowest silver contents obtained in this way remains higher than 20 wt% [5].

Our approach consists in substituting silver with another conductive material, namely copper. Pure copper particles embedded into ECAs oxidise, contributing to a fast degradation of their electrical conductivity [6]. On the other hand, core-shell copper-silver (Cu@Ag) particles were reported to be more resistant towards oxidation, promoting low electrical resistivity, even upon aging [6]. This paper presents the fabrication of copper-based ECAs dedicated to the low-temperature interconnection of solar cells.

2. Materials and methods

Cu@Ag particles (5 ± 1 wt% silver, 200 nm thick silver shell) were purchased from ABCR. 2-butanone was purchased from Technic Micropur. Isobornyl acrylate, 1,6-hexanediol diacrylate, Poly(methyl methacrylate) (PMMA, 120 kg/mol), and 2,2′-Azobis(2-methylbutyronitrile) (AIBN) were purchased from Sigma-Aldrich.

Based on the composition of commercial PV-dedicated ECAs and reference [7], the products were mixed in the ratios shown in Table 1 with the following procedure. A solution of PMMA solubilised into 2-butanone (ratio 1:3) was mixed with the acrylate monomers. The 2-butanone was removed using a rotary evaporator (Heidolph). Cu@Ag particles and AIBN were sequentially added to the solution, followed by mixing during respectively 24h and 2h using a roller shaker (Stovall). The samples obtained were stored in a freezer.

Table 1. *Composition of the homemade ECAs presented in this work*

Material	Role	Content in ECA A (wt%)	Content in ECA B (wt%)
Isobornyl acrylate	Monomer	17	13
1,6-hexanediol diacrylate	Monomer	17	13
2,2′-Azobis(2-methylbutyronitrile) (AIBN)	Free-radical initiator	1	1
Poly(methyl methacrylate) (PMMA, 120 kg/mol)	Viscosity modifier / dispersant	6	4
Cu@Ag particles	Conductive fillers	59	69

A Leo 1530 Scanning Electron Microscope (SEM) was used to characterise the morphology of the particles (high voltage = 10 kV, working distance = 10 mm). Thermogravimetric coupled with Differential Scanning Calorimetry (TG-DSC) characterisations were carried with a SDT Q600 (TA Instruments), at heating rates of +5°C/min under air for the particles, and +10°C/min in a closed alumina pan for the ECAs. ECAs were deposited on a glass slide and cured at 160°C on a hot plate to confirm their fast-curing behaviour.

For electrical characterisations, samples were deposited in moulds measuring 12 * 12 * 0.7 mm^3. They were cured at 150°C for 15 min under a mechanical loading of 1 kg. Their thickness was measured with a Mitutoyo Absolute micrometer and averaged on five different locations. Resistivities were measured on three samples for each ECA with a four-points probe setup (Napson) and averaged.

For the interconnection experiments, ECAs were deposited through masks on half M2 SHJ solar cells by stencil-printing. The masks contain full-lines patterns composed of openings of 300 µm width. The quality of the printed patterns was examined with a Keyence optical microscope. Interconnection with textured silver ribbons (width = 0.6 mm) was performed by annealing the samples 20 s at 160°C under a mechanical loading.

3. Results and discussion

Firstly, an analysis of the commercial Cu@Ag particles purchased was performed. Figure 1.a shows that the particles have elongated shapes and sizes in the order of 10 µm. Figure 1.b reveals that their mass increases upon heating in air, which corresponds to the oxidation of copper atoms into Cu_2O and CuO. No increase of mass was observed up to 220°C, indicating that the particles can withstand oxidation up to this temperature. Therefore, they are suitable for the low-temperature interconnection of cells below 200°C. Upon complete oxidation of copper into CuO, the mass of the particles increased by +23 wt%, in agreement with their theoretical composition of 95 wt% copper.

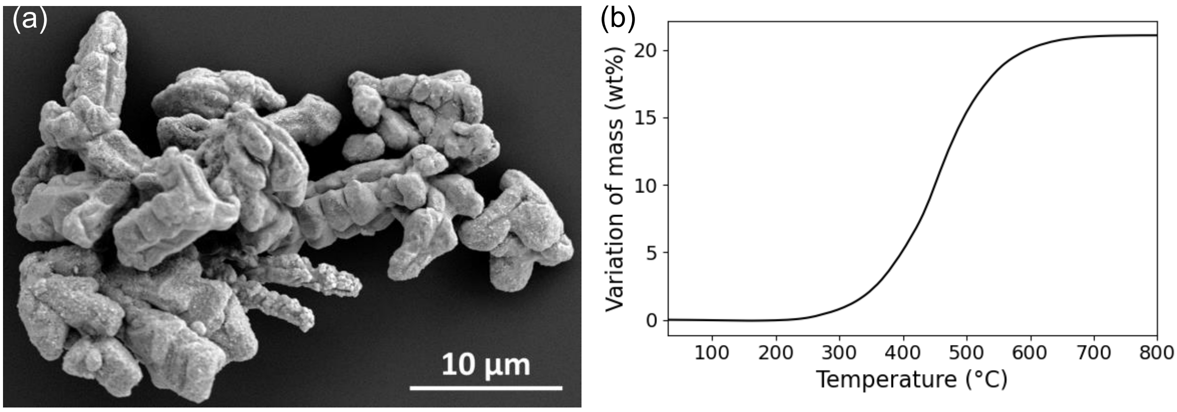

Figure 1. *(a) SEM image and (b) thermogravimetric analysis of the particles used*

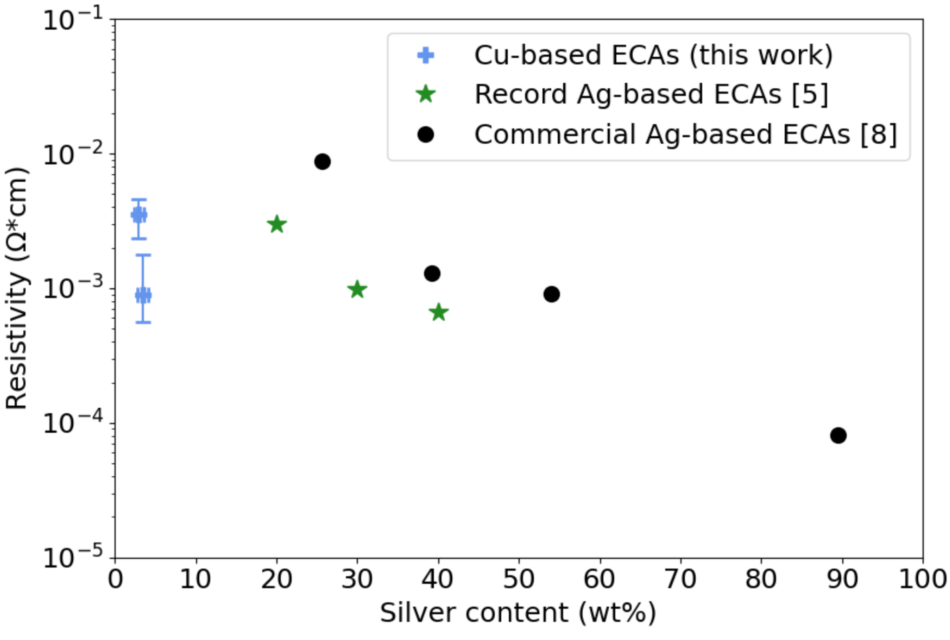

Figure 2. *Electrical resistivities of ECAs A and B. Comparison with the electrical resistivities of ECAs with low silver content and commercial PV-dedicated ECAs [5], [8]*

These particles were used to make copper-based ECAs A and B, containing respectively 59 wt% and 69 wt% fillers, and thus extremely low silver contents of 3.0 wt% and 3.5 wt%. Measurements of the electrical resistivities of these ECAs revealed that the resistivity of ECA A is $3.5 * 10^{-3}$ Ω*cm while that of ECA B is $9.0 * 10^{-4}$ Ω*cm. As highlighted in Figure 2, these resistivities are comparable to both the resistivities of commercial PV-dedicated ECAs and those of silver-based ECAs with the lowest silver contents reported. Thus the copper-based ECAs made are promising for future substitution of silver with copper.

ECAs dedicated to the interconnection of SHJ solar cells should be compatible with a fast-cure processing in less than thirty seconds at low temperature below 200°C [9]. Each of ECA A and ECA B was deposited on a glass slide and annealed at 160°C on a hot plate. After 20 seconds kept at 160°C, both ECA pastes had solidified, proving their compatibility with fast-curing processes. The curing behaviour of ECA A was further investigated with TG-DSC, and is presented in Figure 3. Upon heating, the DSC signal exhibits an exothermic peak, starting at 95°C and with its maximum at 105°C. This peak corresponds to the curing of the monomers, this reaction being launched by the decomposition of the free-radical initiator. In the meantime, the TG signal shows only a small loss of mass of -2 wt% when the sample is heated to 200°C. It can be concluded that both the monomers and their reaction products are relatively stable up to 200°C.

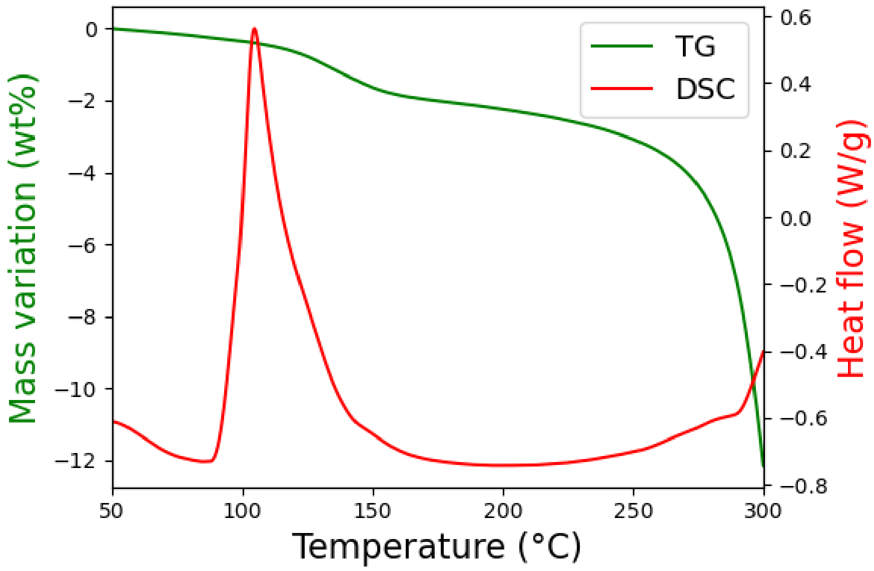

Figure 3. *TG-DSC analysis of ECA A*

The integration of ECAs A and B into PV modules was tested. Firstly, the ECAs were stencil-printed on SHJ solar cells. 16 mg to 20 mg of ECAs were deposited per half M2 cell, which is consistent with usual values for full-lines patterns. Similar quality of printing was observed for both ECAs. An example of stencil-printed ECA A displayed in Figure 4 reveals that the pattern is well-respected, while no splashing was obtained thanks to the viscous behaviour of the ECAs.

Textured silver ribbons were deposited on the top of the ECA patterns, and annealing at 160°C during 20 seconds was performed to interconnect the solar cells. Unfortunately, the adhesion between the ECAs and the silver ribbons is weak, causing the strings fabricated to break when handled.

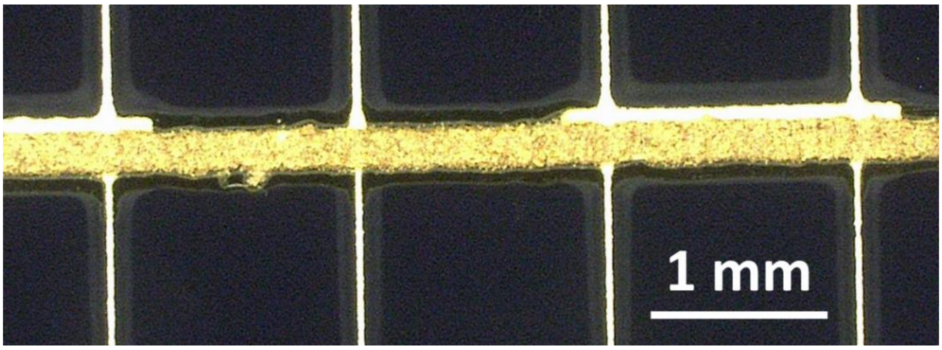

Figure 4. *Optical image of ECA A stencil-printed on a SHJ solar cell*

According to Kinloch [10], the adhesive fracture energy of a glued assembly G_C can be expressed as:

$$G_C = G_0 + \psi \quad (1)$$

Where G_0 is the intrinsic adhesive fracture energy determined by the bonding forces across the interface, and ψ is the energy dissipated viscoelastically when a crack propagates. As a consequence, both terms should be maximised to enhance the adhesion of the home-made ECAs. Namely, the use of coupling agents could increase the strength of the bonding forces existing between the ECAs and the ribbons [11], increasing overall the adhesion of the assembly. Meanwhile, careful tuning of the polymeric matrix can lead to enhanced dissipation phenomena [12]. Thus, future works can focus on the integration of such compounds into the formulation of home-made ECAs.

4. Conclusion

The reduction of silver consumptions in the PV industry is a challenging topic to be addressed urgently. In this work, the fabrication of copper-based ECAs for the low-temperature interconnection of solar cells was realised. The copper-silver particles used proved to be resistant to oxidation up to 220°C, and thus are promising candidates for the fabrication of ECAs that cure below 200°C. Two pastes, containing 59 wt% and 69 wt% of these particles were fabricated. They correspond to pastes containing 3.0 wt% and 3.5 wt% silver respectively. Resistivities down to 9.0 * 10^{-4} Ω*cm were measured, fulfilling typical conductivity requirements for such pastes.

These ECAs have also demonstrated compatibility with automated solar cells interconnection. Their curing occurs around 105°C, and the pastes solidify upon annealing at 160°C during 20 seconds. Initial trials conducted with industrial-type processes showed efficient deposition of the pastes on the solar cells, ensuring pattern accuracy and preventing splashing. The curing also led to full solidification of the pastes. However, the adhesion of the strings obtained was weak. Future works will be focused on the modification of the polymeric matrix to enhance both its ability to dissipate mechanical energy and its ability to bond with metallic ribbons.

Author contributions

Nathalie Ronayette: Conceptualisation, Data curation, Formal analysis, Investigation, Validation, Visualisation, Writing – original draft, Writing – review & editing, **Sonia Sousa Nobre**: Formal analysis, Writing – review & editing, **Sandrine Barthélémy**: Data curation, **Olivier Poncelet**: Conceptualisation, Writing – review & editing, **Daniel Bellet**: Writing – review & editing, and **Rémi Monna**: Conceptualisation, Resources, Writing – review & editing

Competing interests

The authors declare no competing financial interests.

Data availability

Data may be made available upon request.

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