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Electronic Supporting Information

Novel laser-assisted glass frit encapsulation for long-lifetime perovskite solar cells

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Model Equations

Fourier’s Law

$$\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial y^2}+\frac{\partial^2 T}{\partial z^2}+\frac{\dot{q}}{k}=\frac{\rho C_p \partial T}{k\ \partial t} \qquad (S1)$$

Coupled thermo-elastic-plastic differential equations

$$\int_v \{\dot{\varepsilon}\}^T[C_{ep}]\{\dot{\varepsilon}\}\,dv+\int_v \{\dot{\varepsilon}\}^T\{\gamma\}\dot{T}dv=\int_v \{V\}^T\{\dot{f}\}\,dv+\int_S \{V\}^T\{\dot{p}\}\,dS$$

$$\int_v (\rho_0 C_p+\bar{\gamma})\{T\}^T\{\dot{T}\}\,dv-\int_v \{\bar{\beta}\}^T\{\dot{\varepsilon}\}\,\{T\}dv$$
$$=\int_v D\,\{T\}^T dv+\int_v Q(r)\{T\}dv+\int_v \{T_{,i}\}^T[k]\{T_{,j}\}dv+\int_S \{q\}^T\{n\}\{T\}\,dS \qquad (S2)$$

where ε is the strain, $[C_{ep}]$ is the elastic-plastic matrix, $[k]$ is the thermal conductivity matrix, D is the thermoplastic coupling factor, T is the temperature, ρ is the specific mass, q is the heat flux, γ is the generalized thermal modulus, C_p is the specific heat, β is the thermal modulus tensor, Q is the heat generation, V the rate of displacement, f is the body forces and p is the surface tractions.

Simulation assumptions

The following main assumptions were considered: i) air convection at the sides of the substrates are negligible; ii) emissivity of the glass substrate is 1, and there is no radiation or convection in the space between the two substrates; iii) the laser beam is absorbed at the top of glass frit and the laser beam absorption in the cover substrate is negligible; iv) the glass frit is bonded to the both substrates; v) materials are isotropic and thermal and mechanical properties are constant with temperature; this includes the three layers of glass frit here treated as made of a single material. Quadratic mesh elements were considered, with mesh sizes of < 25 µm for the glass frit and < 1 mm for the rest of the bodies. The properties of the glass frit and substrates are presented in Table S1.

Table S1 – Properties of the glass frit and substrates used in the thermal stress simulation.

	Glass frit	Substrate
ρ [kg m^3]	5000	2500
CTE [10^{-6} °C^{-1}]	8	8.5
E [GPa]	80	50
Poisson’s ratio	0.3	0.18
Thermal Conductivity [W °C^{-1} m^{-1}]	3	1.4
Specific Heat [J kg^{-1} °C^{-1}]	600	750

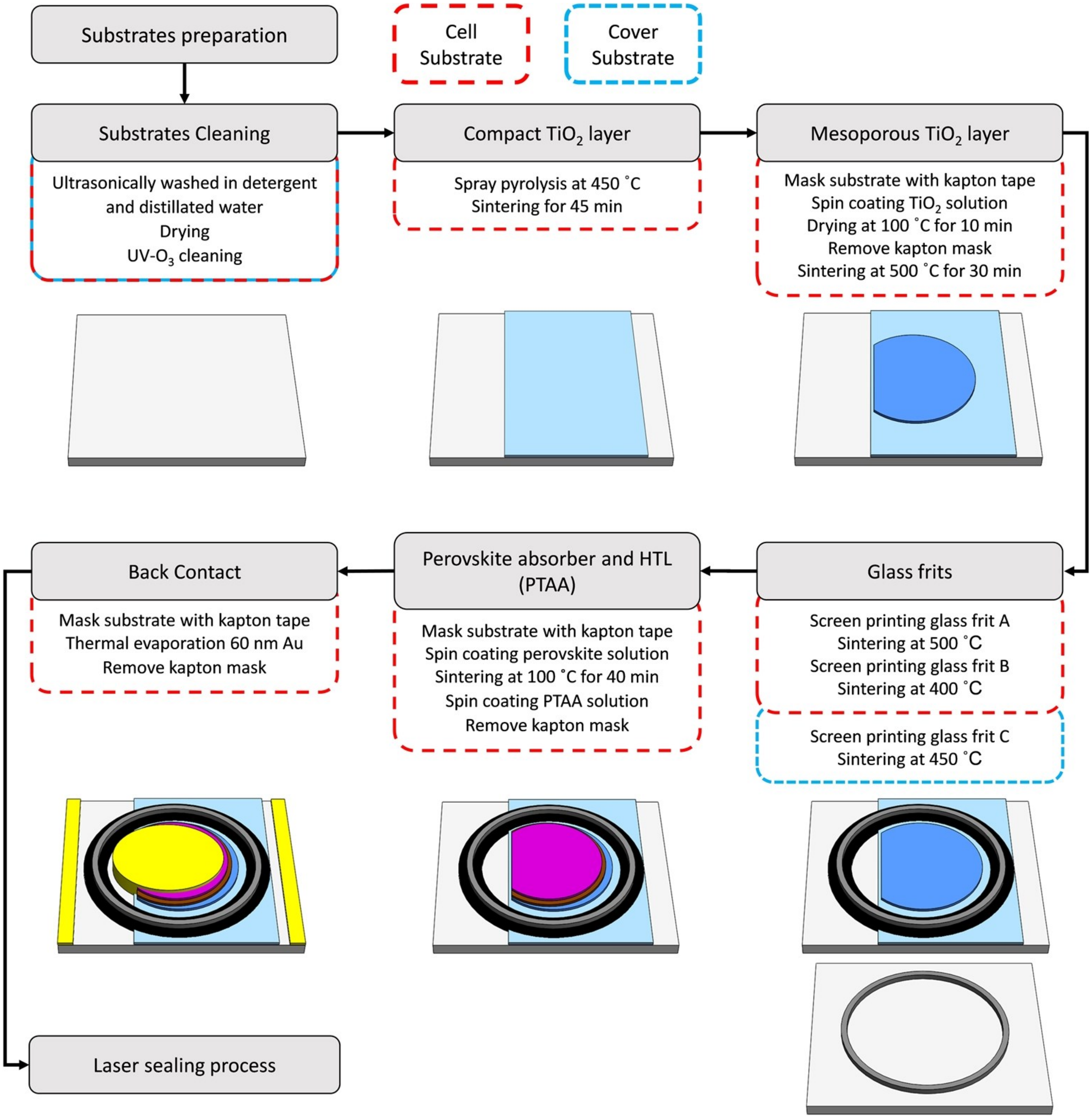


Figure S1 – Schematic diagram of the procedure for the fabrication of laser-sealed PSCs. Note that the masking steps are not included in the schemes.

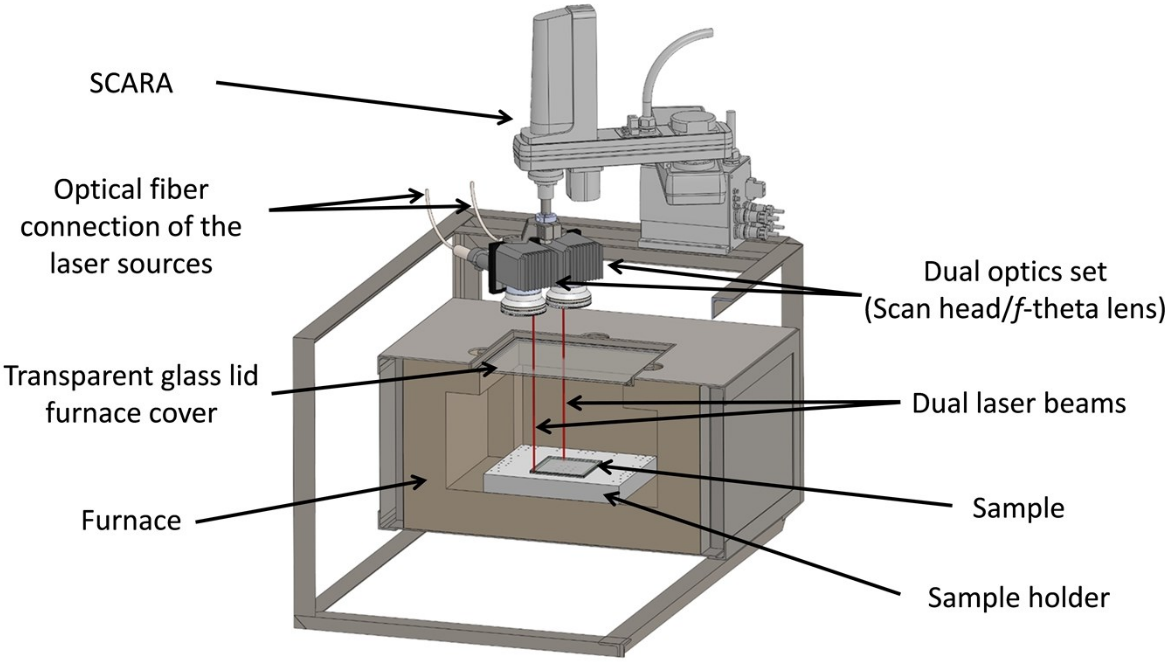


Figure S2 – Schematic view of “LaserStation” used for the laser assisted glass encapsulation.

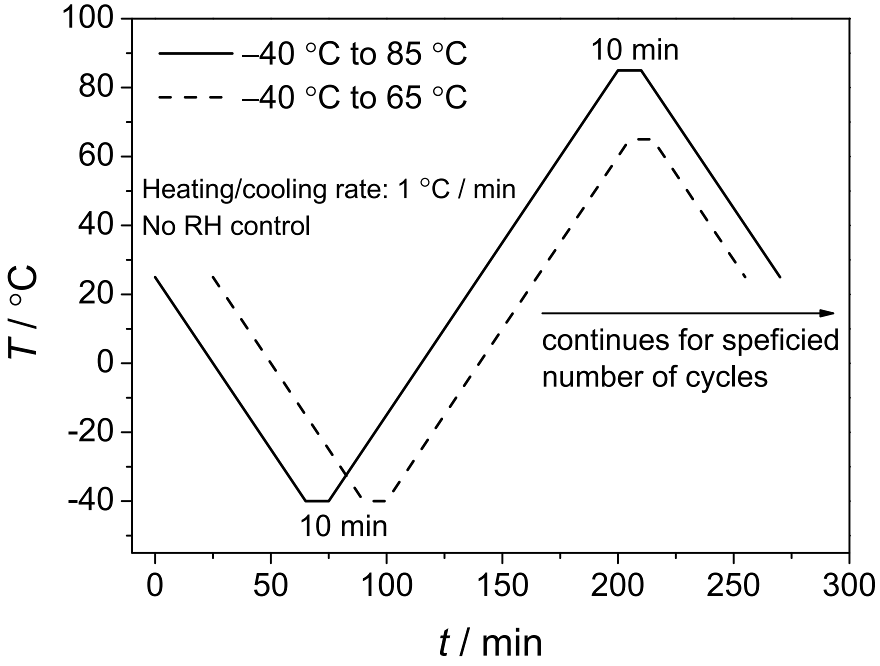
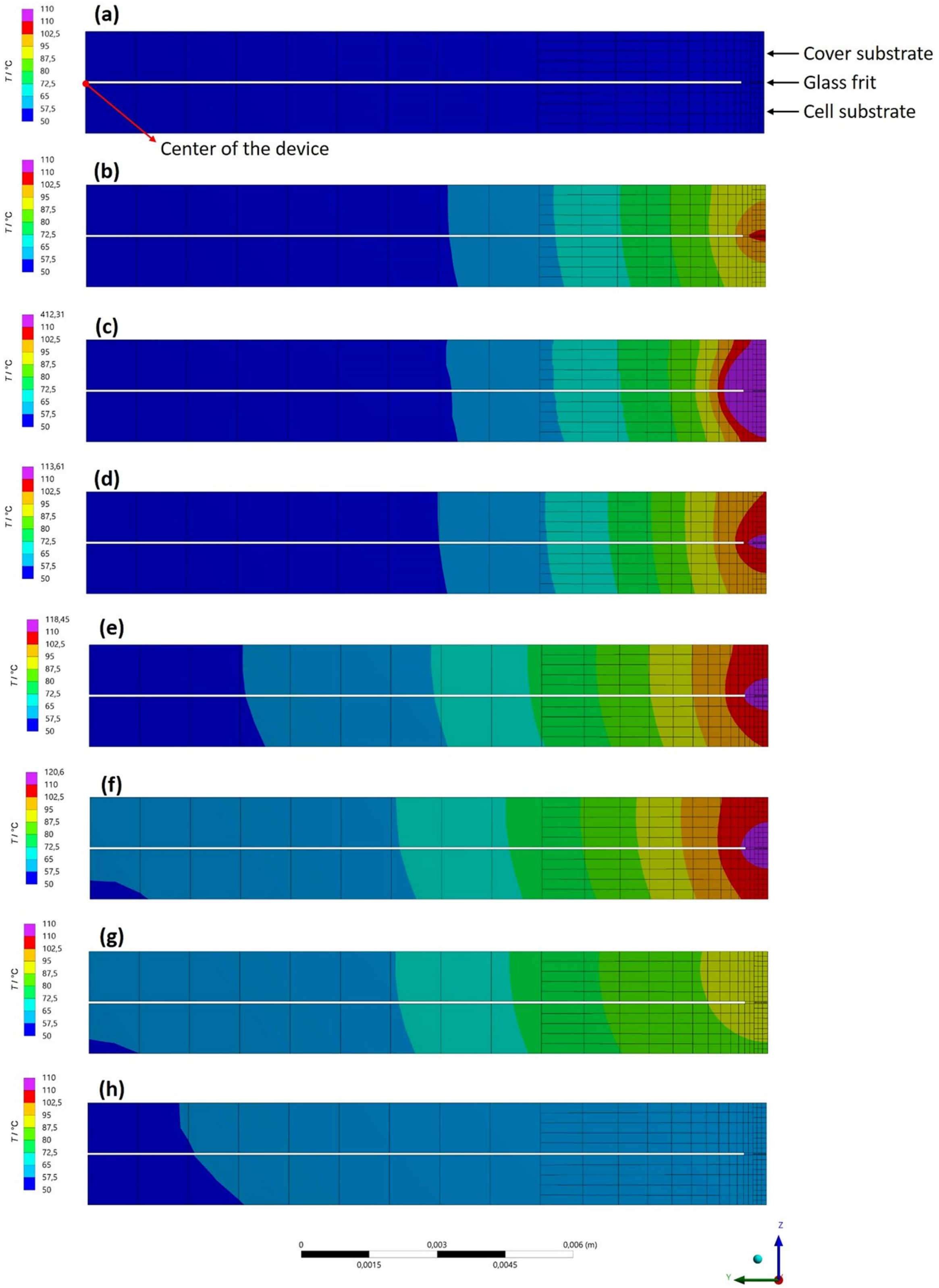


Figure S3 – Temperature history of the two thermal cycling tests.



(a)
T / °C
110
110
102,5
95
87,5
80
72,5
65
57,5
50
Cover substrate
Glass frit
Cell substrate
Center of the device
(b)
T / °C
110
110
102,5
95
87,5
80
72,5
65
57,5
50
(c)
T / °C
412,31
110
102,5
95
87,5
80
72,5
65
57,5
50
(d)
T / °C
113,61
110
102,5
95
87,5
80
72,5
65
57,5
50
(e)
T / °C
118,45
110
102,5
95
87,5
80
72,5
65
57,5
50
(f)
T / °C
120,6
110
102,5
95
87,5
80
72,5
65
57,5
50
(g)
T / °C
110
110
102,5
95
87,5
80
72,5
65
57,5
50
(h)
T / °C
110
110
102,5
95
87,5
80
72,5
65
57,5
50
0
0,003
0,006 (m)
0,0015
0,0045
Z
Y

i)

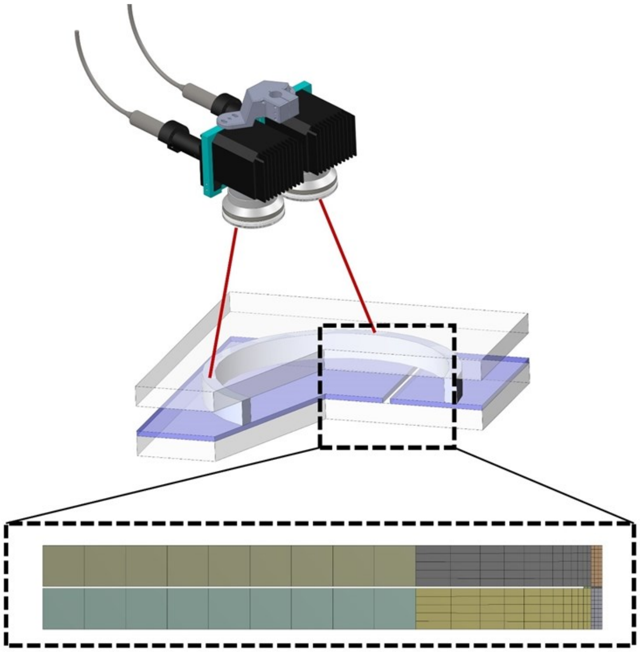


Figure S4 – Simulated temperature during the sealing process at the glass frit and substrates. (**a**) the device is placed in the furnace at the process temperature of 50 °C until a homogeneous temperature is reached (temperature stabilization period is not represented), (**b**) LB_{heat} starts to radiate and the temperature in the sealant increases up to 110 ± 10 °C (**c**) LB_{bond} is emitted and the, temperature in the glass frit reaches up to > 380 °C, bonding the substrates, (**d**, **e** and **f**) LB_{bond} is turned off and LB_{heat} continues emitting, maintaining the sealant material at 110 ± 10 °C, (**g** and **h**) laser-sealing conclusion, LB_{heat} is turned off and the temperature in the sealant decreases to the process temperature, **i)** scheme of the cross-section view of the simulation model.

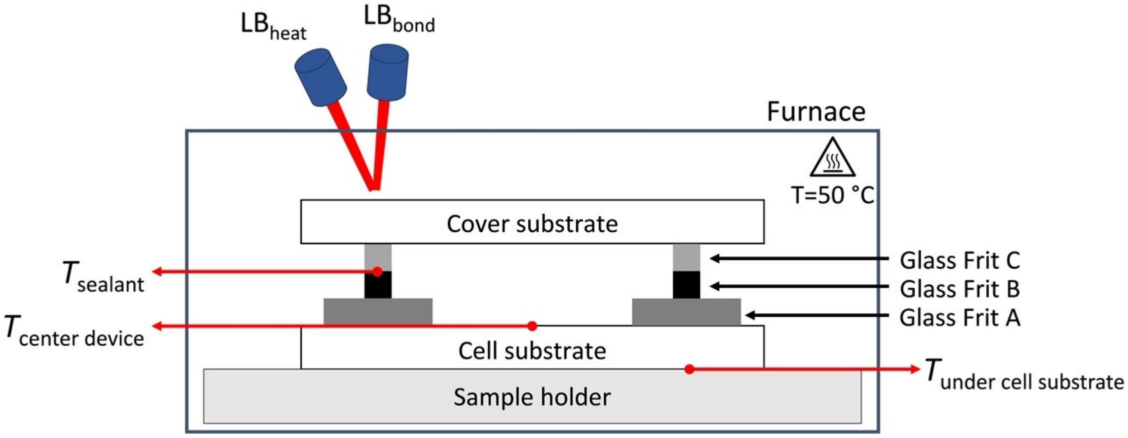


Figure S5 – Thermocouples positions for temperature measurement during sealing process.

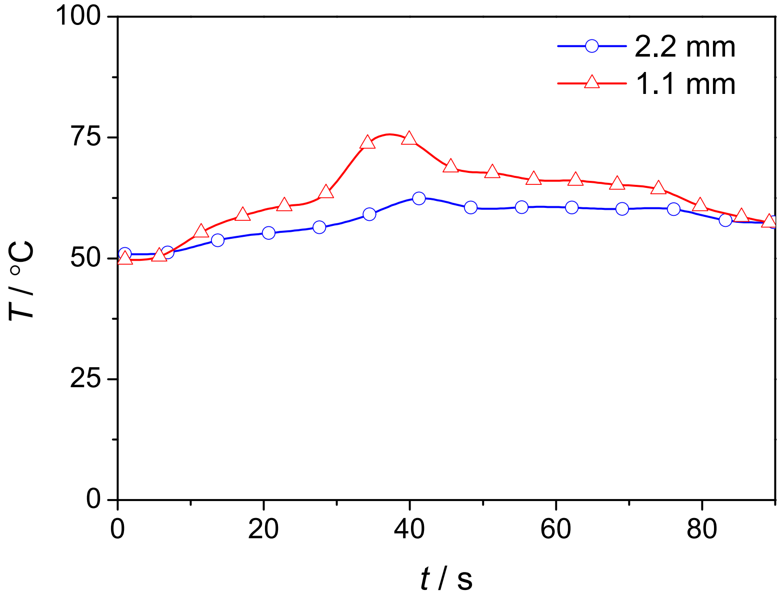


Figure S6 – Temperature history at the rear side of the glass (*i.e.* under cell substrate) for 2.2 mm and 1.1 mm thick glass substrates.

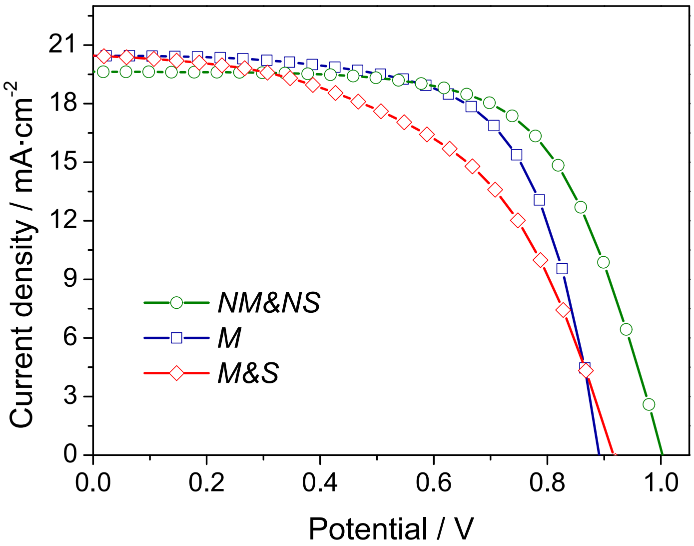


Figure S7– Current density *vs*. potential curves of *NS&NM*, *M* and *M&S* devices.

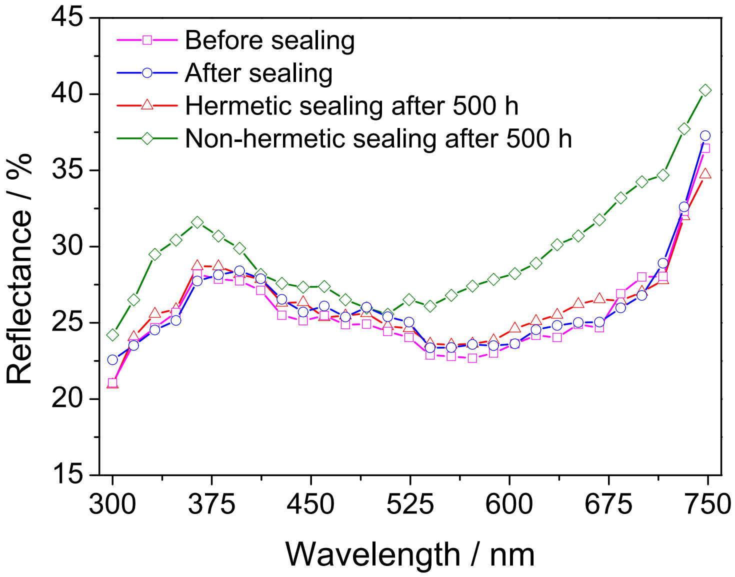


Figure S8 – Reflectance spectra of a PSC before and after sealing process; and for a hermetically and a non-hermetically encapsulated devices, after 500 h of humid air exposure.

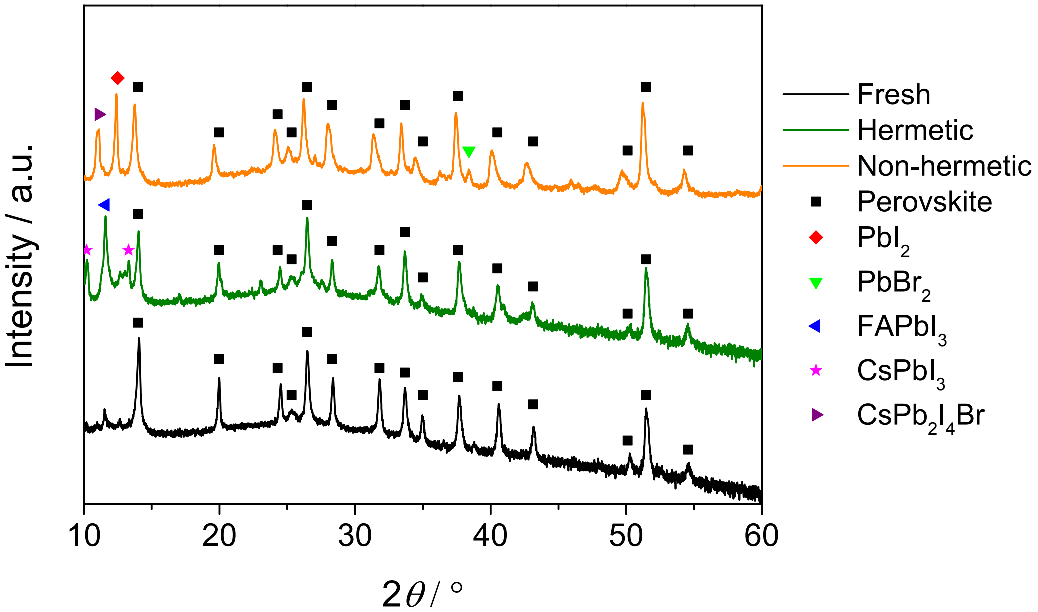


Figure S9 – XRD pattern for a fresh device and for hermetically and non-hermetically encapsulated devices, after the thermal cycling test between −40 °C to 85 °C.

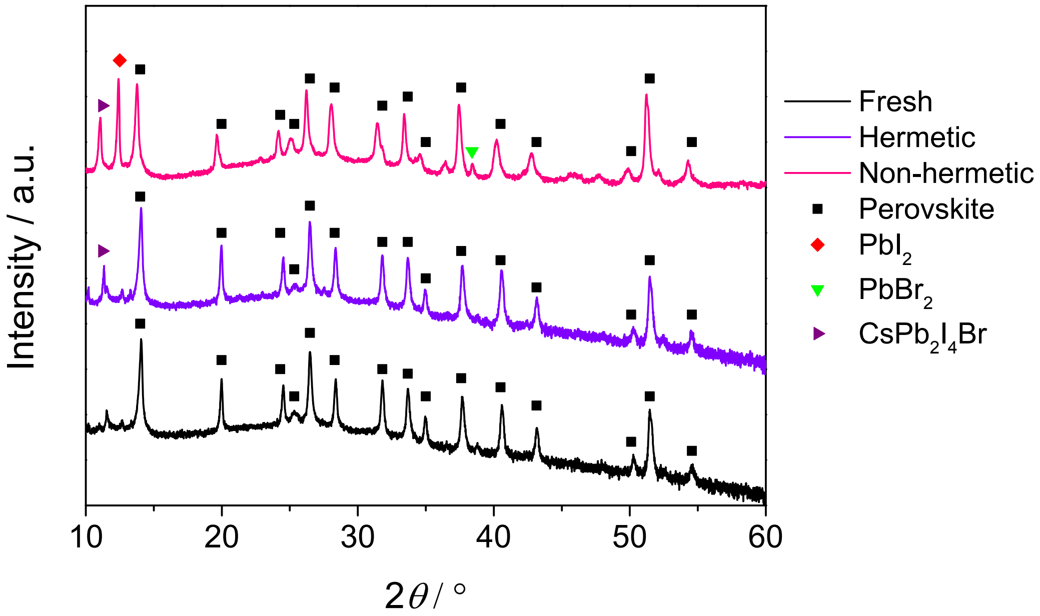


Figure S10 – XRD pattern for a fresh device and for hermetically and non-hermetically encapsulated devices, after the thermal cycling test between −40 °C to 65 °C.