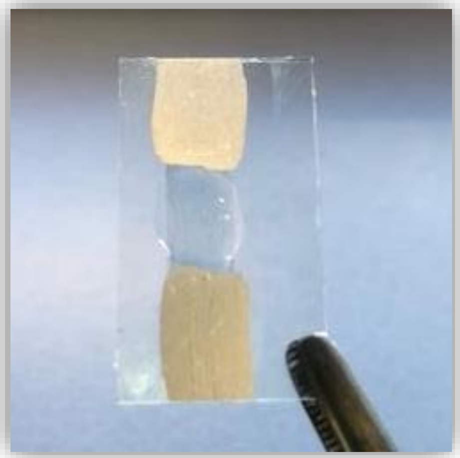
More than 4 times power factor improvement in a nanostructured Sb-doped SnO_2 film produced by the presence of a solid electrolyte



Mauricio Solis-de la Fuente*, Sergio Castro-Ruiz, Pauline Rullière, Sébastien Fantini, Jorge García-Cañadas

Universitat Jaume I, TESLab (http://teslab.uji.es), Spain

Solvionic (https://en.solvionic.com) France

E-mail: msolis@uji.es

OUTLINE

1. Introduction

2. Experimental part

3. Results

4. Summary

5. Acknowledgements

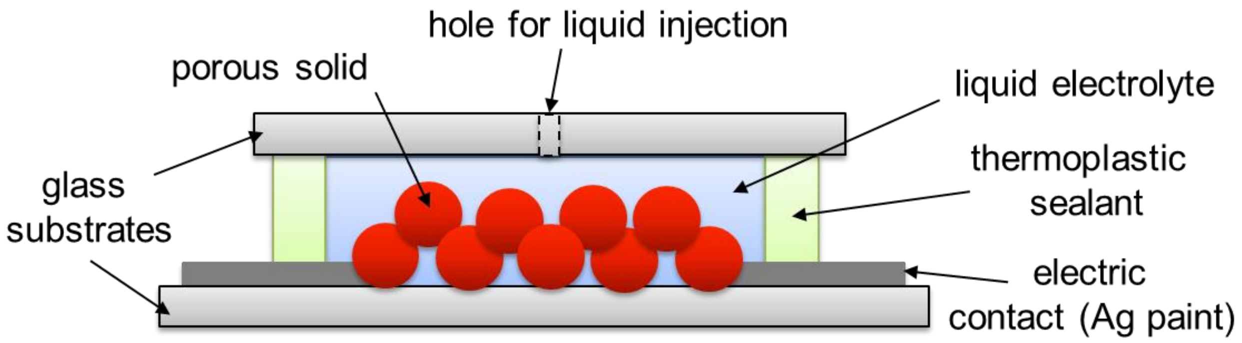
1. Introduction

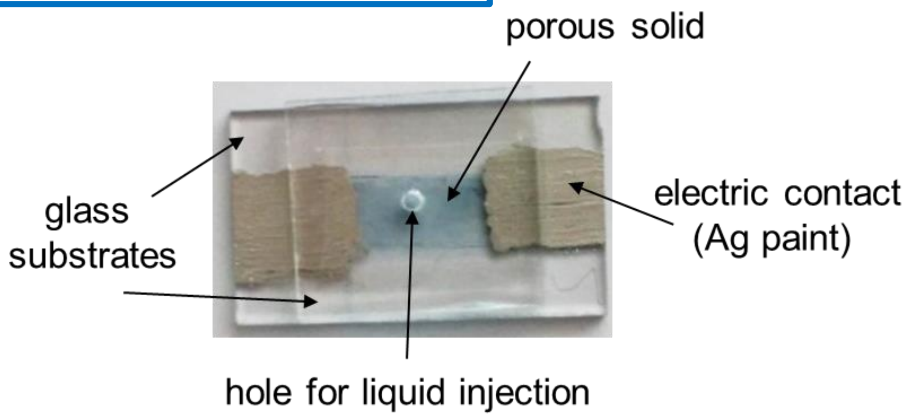
We reported in 2018 a **new thermoelectric (TE) concept**: **TE solid** which is **permeated by a liquid electrolyte** capable to **improve the power factor more than 3 times**.

ACS APPLIED ENERGY MATERIALS
Letter
Cite This: *ACS Appl. Energy Mater.* 2018, 1, 254–259
www.acsaem.org

Large Power Factor Improvement in a Novel Solid–Liquid Thermoelectric Hybrid Device

Lourdes Márquez-García,[†] Braulio Beltrán-Pitarch,[†] Damian Powell,[‡] Gao Min,[‡] and Jorge García-Cañadas*[,†]



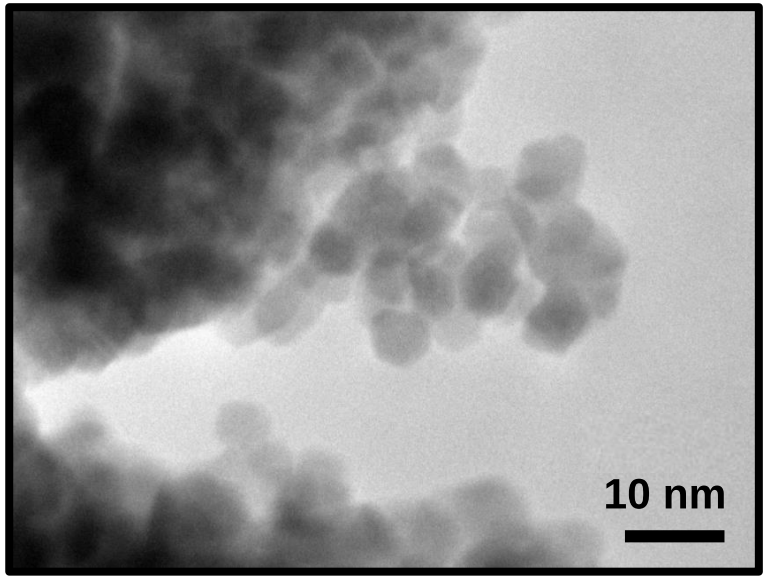


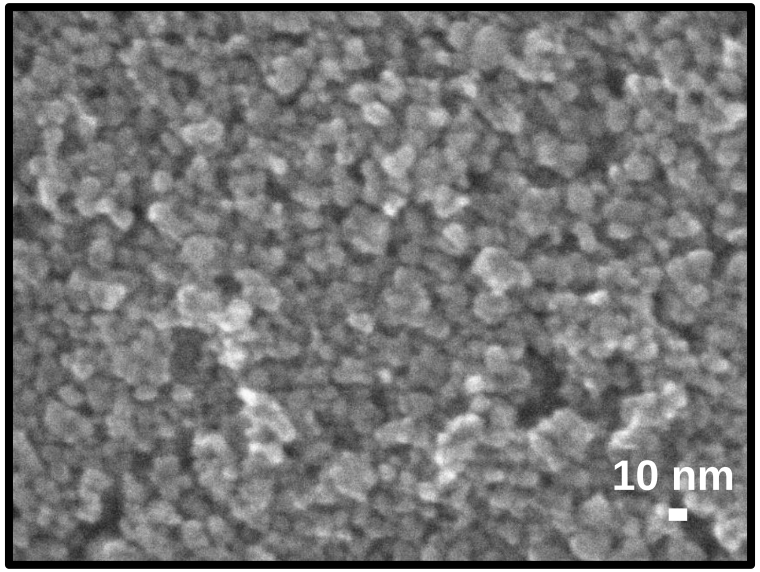
In this study, we **replace** the **liquid** electrolyte by a **solid electrolyte** to fabricate an **all-solid-state device**

2. Experimental part

Nanostructured Sb:SnO_2

Films were prepared from a commercial **colloidal water dispersion** directly deposited by **spin coated** (5 layers) on a glass substrate and **annealed at 550 °C** for 45 min.





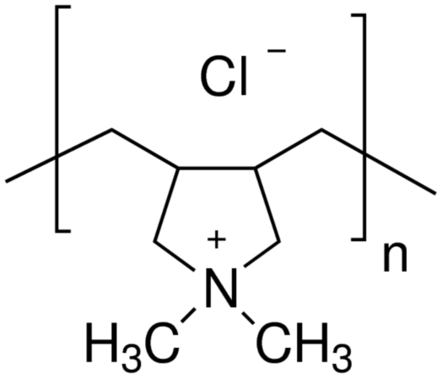
Nanostructured films of **nanoparticles** of around **4 to 10 nm** diameter. Film thickness is ≈1.0 μm. **Pores** in the **2-50 nm range** are present.

2. Experimental part

Solid electrolyte

Polyelectrolytes are **macromolecules with ionizable groups**, which dissociate in polar media into polyions and counterions with opposite charges.

Polydiallyl-dimethylammonium chloride (PDADMAC)



Deposition at room temperature.

Water as solvent.

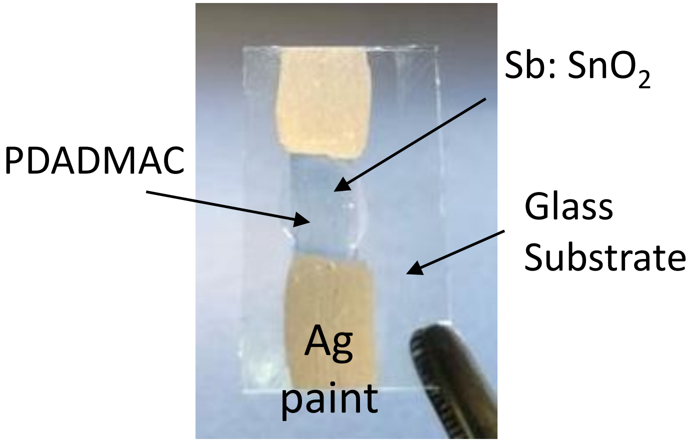
Large S values ∼19 mV/K [1].

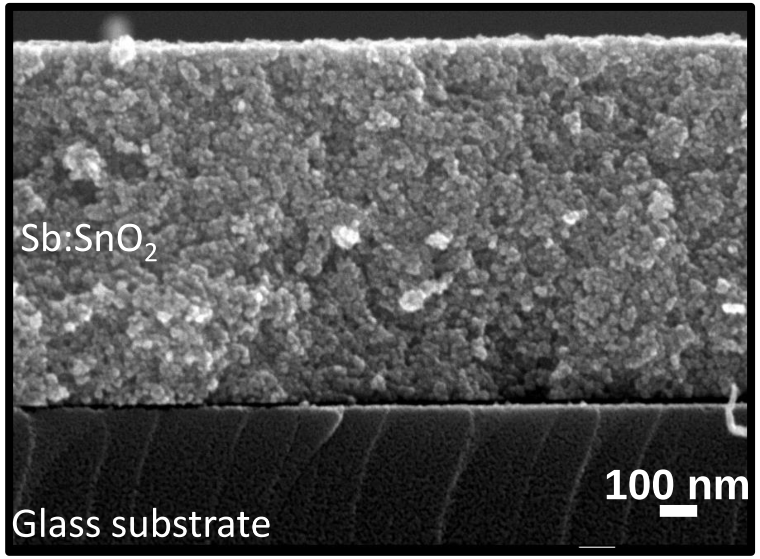
[1] S. L. Kima , J. H. Hsub , C. Yu. Thermoelectric effects in solid-state polyelectrolytes. *Organic Electronics* 54 (**2018**) 231–236.

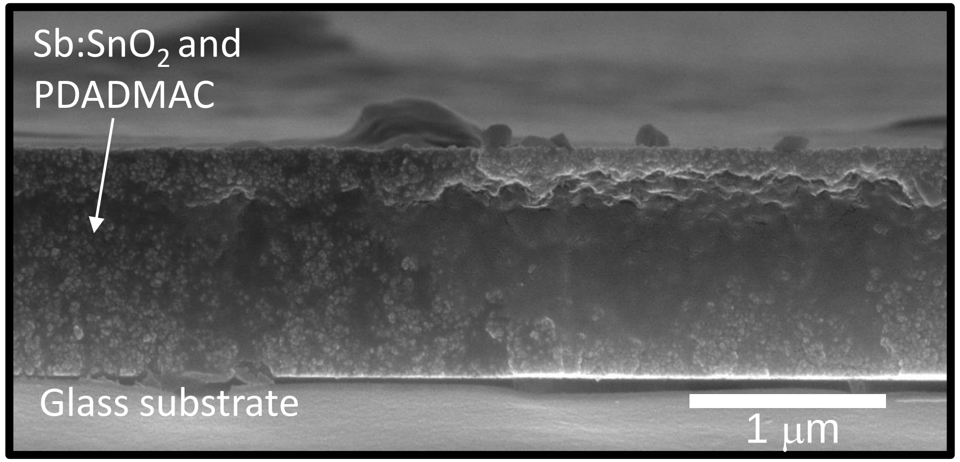
2. Experimental part

Device preparation

PDADMAC (20 %wt solution in H_2O) **Solvionic prepared** was deposited by **drop casting** on top of the **Sb:SnO$_2$ film contacted by Ag paint contacts**. It was allowed to **dry at room** temperature for at least **2 h**.

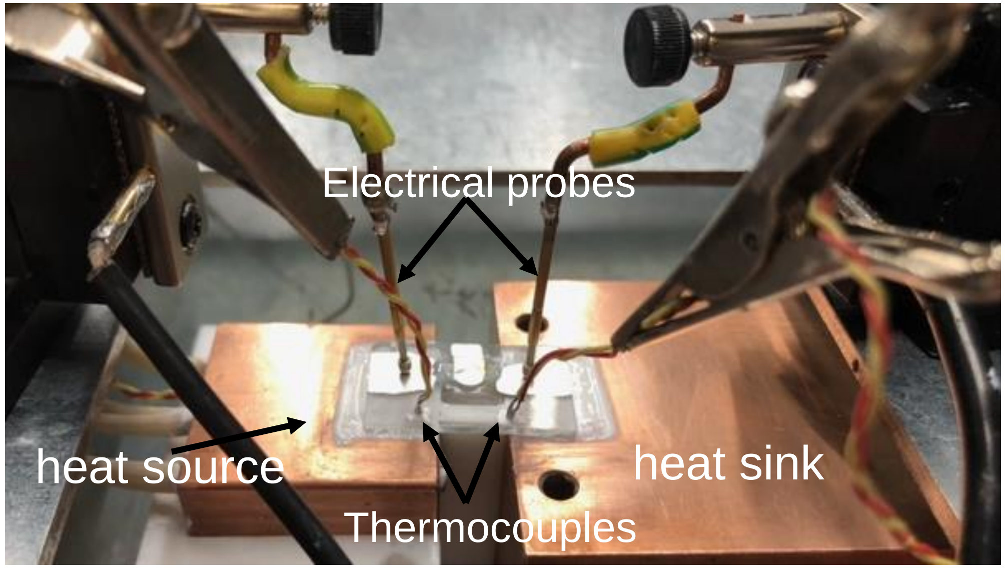






PDADMAC polyelectrolyte **penetrates into the Sb:SnO$_2$** nanostructured film.

Thermoelectric measurement setup



Seebeck coefficient (S)

Obtained from the slope of the open-circuit voltage (Voc) vs temperature difference (ΔT) plot.

Nanovoltmeter Keithley 2182
Thermocouples type K and thermometer RS 1316
Temperature controller Watlow EZ-zone

Device electric resistance (R)

Obtained from the slope of the I–V curve at 5 K temperature difference.

Source Meter Keithley 2450

3. Results

Seebeck coefficient and device resistance measurements

Three similar **samples** were measured **before and after** the deposition of the polyelectrolyte:

Sample	Seebeck coefficient (μV/K)			Device electric resistance (kΩ)			$PF_{with}/PF_{without}$
	Without electrolyte	With electrolyte	S variation (%)	Without electrolyte	With electrolyte	R variation (%)	
S1	**-39.31**	**-35.76**	-9.03	**11.86**	**2.43**	-79.47	**4.06**
S2	**-47.57**	**-42.82**	-9.98	**15.83**	**2.68**	-83.07	**4.86**
S3	**-43.38**	**-39.66**	-8.57	**18.95**	**2.78**	-85.33	**5.72**

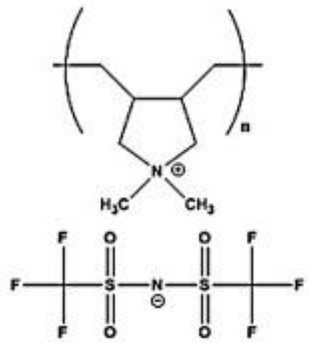
- **Nearly no variation (9.2% reduction)** in the absolute value of the **Seebeck coefficient.**
- **83% decrease in the electrical resistivity of Sb:SnO_2.**

A **remarkable** average **improvement** in the power factor (**PF**) of **4.9 times** was obtained.

The resistance of a **device with only the polyelectrolyte** (no Sb:SnO_2) is **1.5 MΩ**, so **no** electrical **conduction** occurs **through the polyelectrolyte**.

3. Results

PDADMATFSI



Why the improvement is produced?

To identify how the PF improvement is produced we evaluated **an additional polyelectrolyte** formed by the same polycation but **varying the anion,** prepared by **Solvionic.** Instead of Cl^-, **$TFSI^-$**, 20 % wt **acetonitrile** solutions were tested.

Sample	Seebeck coefficient (µV/K)			Electric resistance (kΩ)			$PF_{with}/PF_{without}$
	Without electrolyte	With electrolyte	S variation (%)	Without electrolyte	With electrolyte	R variation (%)	
S4	**-43.55**	**-41.90**	-3.78	**7.18**	**6.49**	-9.50	**1.04**
S5	**-61.95**	**-60.09**	-2.99	**18.65**	**15.08**	-19.15	**1.16**
S6	**-60.10**	**-59.17**	-1.55	**18.16**	**16.33**	-10.06	**1.08**

No PF improvements were found, so the presence of **Cl^- (or H_2O) produces the significant drop in resistance.**

3. Results

Solid electrolyte: solvent effect

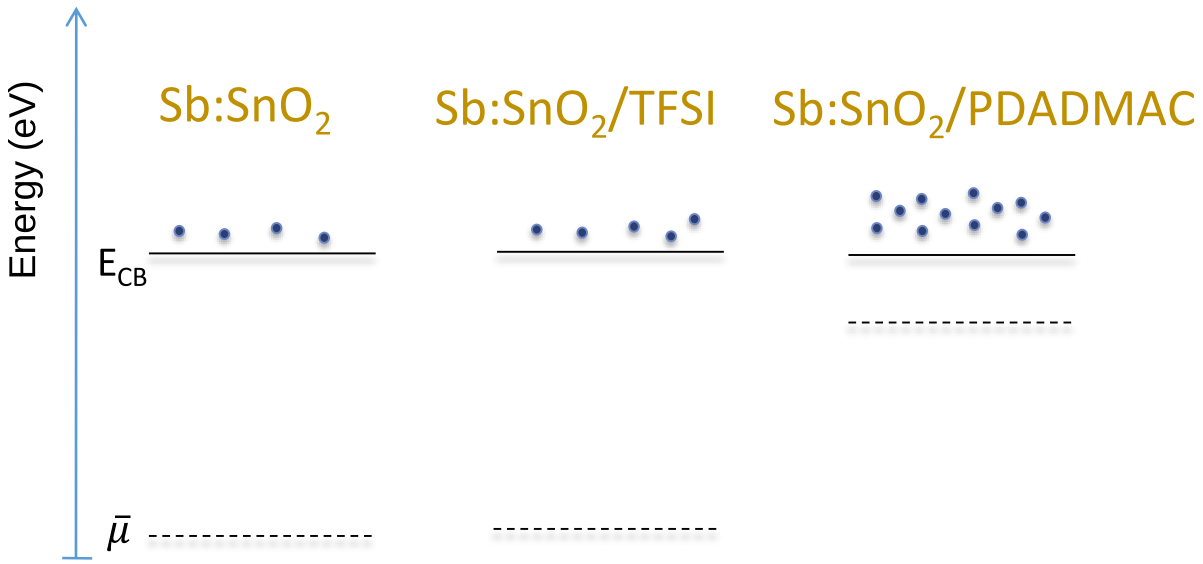
Since the solvent of the polyelectrolytes that do not produce PF improvement was acetonitrile (ACN), we evaluated if the use of water solvent played a role, so **water was added** (10 %wt) **to the acetonitrile** solutions.

Sample	Seebeck coefficient (μV/K)			Electric resistance (kΩ)			$PF_{with}/PF_{without}$
	Without electrolyte	With electrolyte	S variation (%)	Without electrolyte	With electrolyte	R variation (%)	
S7 (TFSI+ 10% H_2O)	**-42.27**	**-43.35**	2.57	**13.64**	**15.19**	11.32	**0.93**

Again, **no PF improvements were found**, so the presence of **Cl^- ions produces the significant drop in resistance**.

Solid electrolyte effect (possible explanation)

An **increase in the electrical conductivity** (electrical resistance drops) of a nanostructured $Sb{:}SnO_2$ film and a **decrease in the Seebeck coefficient** was found when the film was in contact with PDADMAC, reaching **4.9 times improvement in the PF**.



- For PDADMAC, **Cl^- ions exchange electrons with $Sb{:}SnO_2$**, which sets the $\bar{\mu}$ **closer to the E_{CB}**.
- Electron concentration ***n* increases as $\bar{\mu}$ is closer to the E_{CB}**.
- Since $\sigma = qn\mu$, **↑*n* produces ↑*σ***.

4. Summary

- After polyelectrolyte PDADMAC deposition on Sb:SnO_2 nanostructured films, a significant improvement in the **power factor (PF) of 4.9 times** was obtained.
- Further investigations showed that the **Cl^- ions produce substantial drops in electrical resistance R.**
- The water addition **did not produce power factor improvements in the Sb:SnO_2/TFSI system.**

The use of this **polyelectrolyte** will be also **tested using state-of-the-art thermoelectric materials** (e.g. Bi_2Te_3) within the EU **UncorrelaTEd Project**.

4. Acknowledgements

This project has received funding from the European Union’s Horizon 2020 research and innovation programme under **grant agreement No 863222 (UncorrelaTEd project),** and from the **Spanish Agencia Estatal de Investigación (MAT2017-91015-EXP)**.

Disclaimer This presentation reflects only the author's view and that the Agency is not responsible for any use that may be made of the information it contains.

UNIVERSITAT JAUME I

TESLab

http://teslab.uji.es

E-mail: msolis@uji.es

UncorrelaTEd

http://uncorrelated.uji.es/