SCALEUP
Large scale molecular simulation of perovskite solar cells

Project duration: from 01.2020 to 01.2023
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Publishable Summary

Metal halide perovskites (MHP) have emerged as one of the most studied semiconductors due to their excellent optoelectronic properties. This is evidenced by the rapid development of perovskite solar cells (PSCs) with a record certified photoconversion efficiency nowadays of 26.1% for single junction devices, similar to those of silicon cells and 33.7% for tandems (https://www.nrel.gov/pv/cell-efficiency.html). Nonetheless, industrial application of PSCs is critically hampered by instability issues, including intrinsic, environmental, and operational factors. Instability is attributed to several chemical and dynamical processes that occur at very distinct time scales, like slow ionic rearrangements and physical and chemical interactions in the bulk and at interfaces with contact layers. These phenomena cause IV hysteresis, low frequency signals difficult to interpret and ultimately, device degradation.

In this project, we have adopted a multidisciplinary methodology to comprehensively understand perovskite materials and perovskite solar cells at a fundamental level. This approach encompasses atomistic simulations, as well as electronic and spectroscopic investigations of both perovskite materials and their interfaces. It further involves the creation and examination of experimental test devices, followed by a meticulous evaluation of device performance and stability, all rooted in a fundamental viewpoint.

The underlying principle driving this project is to traverse from the microscopic scale, characterized by short size and time scales, to the macroscopic domain.

As the main results of the project implementation, we can highlight the following:

1) Development of a standard and versatile first-principles atomistic description of perovskite materials, involving the testing of DFT functionals and the finding of a set of reliable and transferable reactive force fields for Classical Molecular Dynamics (CMD) simulations. This is important because it allows to describe and quantify the slow ionic phenomena that determine the short-term stability behaviour of perovskites. We also developed new protocols based on ReaxFF and tight-binding methods, as well as a strategy to find force fields parameters from DFT calculations using a genetic algorithm.

2) Fundamental studies of the role of the interface between perovskites and the selective contacts were accomplished. DFT functionals were used to elucidate complex degradation mechanisms observed in materials such as MoO3, NiOx, and ZnO. Through the lens of DFT, these intricate degradation processes found theoretical explanations, shedding light on the underlying molecular interactions. In addition, passivation strategies were conceived and meticulously expounded upon for both MoO3 and NiOx. These strategies, born from theoretical insights, held the promise of enhancing material stability and performance. The

theoretical foundation allowed for a systematic approach to passivation, opening doors to a new realm of material engineering.

3) Alternative device architectures were fabricated and tested, including NiOx-based solar cells and modification of the interfaces by intercalation of hydrophobic polymers. Thorough analysis via current-voltage and impedance spectroscopy measurements were carried out for these devices. This made it possible to link the results to the insights of the atomistic studies and allowed the implementation of numerical simulations of the full devices. These combined studies cast light of the complex interplay of all factors affecting the performance and stability of perovskite solar cells.

4) New features of the commercial test equipment aimed at studying the stability of perovskite solar cells have been developed within the project. This includes spectrophotometric measurements during aging test and atmosphere control. A new version of the software for device numerical simulation has also been released and successfully tested to describe the devices fabricated by the consortium partners.

Building upon the progress achieved within SCALEUP and drawing from the key findings outlined above, we are currently pursuing the following next steps:

1. In the theory and modelling aspects, we are enhancing the optimization process for parameter fitting derived from DFT calculations by incorporating Machine Learning techniques such as linear regression and artificial neural networks.
2. On the experimental front, we are focusing on the passivation and encapsulation of perovskite interfaces with organic polymers in order to enhance the stability of fully operational devices at relevant environments.

The avenues for exploitation of the expertise acquired through the SCALEUP project extend to the final development and commercial utilization of instruments designed to assess the stability of perovskite-based fully functional devices in real-time. This technology has the potential for broader application in addressing similar stability issues in other related technologies.

Project consortium

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Participating countries and financing:

Country	Number of organisations involved	Project costs in EUR	Public funding in EUR
SPAIN	1	172 500	122 915
GERMANY	1	264 353	264 353
SWITZERLAND	1	421 913	168 765
ISRAEL	1	153 795	153 795
THE NETHERLANDS	1	12 931	-
Total	*5*	1 025 492	709 828

Funding agencies involved and contracts

Funding Agency	Contract N° and Title
Ministry of Science (AEI)	National project PCI2019-111839-2
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Bundesamt für Energie (BFE)	SI/501958-01, Model Validation and Stability Characterization Platform with Atmosphere Control for Perovskite Solar Cells
Ministry of Energy (MoE)	Multiscale molecular simulation of perovskite solar cells
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