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Thin-film inorganic photovoltaics

The field of photovoltaics (PV) is extremely active, with research progressing rapidly in silicon, organic, inorganic, dye-sensitized and hybrid technologies. Thin-film PV uses inorganic compounds with direct bandgaps and high absorption coefficients for the absorber layer in a (typically) single-junction device. These layers are typically polycrystalline and very thin (1-2μm). Ideal materials have a bandgap of 1.1-1.5eV, enabling the optimal Shockley-Quasier efficiency limit of around 30% efficiency for a single-junction device. Already, copper indium gallium selenide (CIGS) cells have achieved laboratory efficiencies of ~20% and commercial efficiencies of ~15%. Cadmium telluride cells have slightly lower efficiencies but a substantial manufacturing base, and together CIGS and CdTe are giving us an explosion in low-cost, lightweight, flexible PV cells.

However, providing PV energy on the scale that it is truly needed will require something more ambitious, on the scale of square kilometers rather than square meters. At this level, supply issues are likely to set in for the rare elements In, Ga, Cd and Te. Copper zinc tin sulfide/selenide (CZTS) looks set to take over as supply stresses set in; record efficiencies are not far behind, having just broken the 10% threshold.

Roll-to-roll processing

Large-scale production of thin-film devices is a significant scale-up problem. While there is some interest in flexible polymeric substrates, building-sized devices need to be supported on more traditional engineering materials.

Specifically is a collaboration between academic and industrial groups including Tata Steel and NSG Pilkington. They are developing a range of roll-to-roll processes for functional coatings on steel and glass sheets—these may then be used as cladding for buildings.

CZTS and related materials are seen as strong candidates for photovoltaic coatings in the long term, helping to achieve their vision of “buildings as power plants”. Plausible unit operations include spray and inkjet deposition, near-infrared (NIR) heating and screen printing.

Ab initio thermodynamics

Thermodynamics is an essential tool for the prediction and understanding of phase equilibria, reaction energies and spontaneity. Of particular interest in chemistry is the Gibbs free energy G, which for a multicomponent system is approximately the sum of the chemical potentials μ of its components. In order to probe a range of reaction conditions, we need temperature and pressure-dependent chemical potentials for all the reaction components i. Disregarding any nuclear effects, these can be related to the electrostatic potential and vibrational energies:

\[ \mu_i(T, P) = E_{\text{pot}} + F_i + \int \mu_i \partial P \, dT + \int \mu_i \partial T \, dP + \int F_i \partial T \, dP + \int S \, dT \]

Energetics are calculated with density functional theory (DFT) calculations. Vibrational potentials may be available from experimental data, or can also be computed with DFT. Combining μ to obtain AG, we can assess the viability of a reaction and equilibria of competing reactions.

The preferred ab initio code for this project is FHI-aims, a modern package capable of scaling across thousands of cores. This has allowed us to make use of national-scale computing resources HECToR and Blue Joule, with large calculations using over 4,000 cores. The PBEsol functional is preferred; this is a relatively universal functional with a good balance of cost and accuracy for solid-state systems. The Phonopy package is used for lattice dynamics and hence heat capacity and IR absorption models. For modeling electronic properties including optical absorption spectra, more advanced electronic structure methods are called for: GW, or hybrid functionals such as B3LYP.

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• Aquila, the University of Bath cluster managed by BUCS.