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Material informatics driven design and experimental validation of lead titanate as an aqueous solar photocathode

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ABSTRACT

Materials informatics is a rapidly emerging data- and knowledge-driven approach for the identification of novel materials for a range of applications, including solar energy conversion. Despite significant experimental effort, the development of highly efficient, stable, and cost-effective photovoltaic materials remains a challenging scientific problem. The quest for precisely defined semiconductor properties revolves around an immensely broad landscape of structural parameters. Here, we have resolved this challenge by applying material informatics to design a novel photocathode material for dye-sensitized solar cells (DSSCs). By conducting a virtual screening of 50,000 known inorganic compounds, we have identified lead titanate ($PbTiO_3$), a perovskite, as the most promising photocathode material. Notably, lead titanate is significantly different from the traditional base elements or crystal structures used for photocathodes. The fabricated $PbTiO_3$ DSSC devices exhibited the best performance in aqueous solution, showing remarkably high fill factors compared to typical photocathode systems. The results highlight the pivotal role materials informatics can play in streamlining the experimental development of materials with the desired properties.

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1. Introduction

The discovery of new materials with previously unknown or unexpected properties often leads to paradigm shifts in well-established research fields. Examples include the discovery of high-temperature superconductivity, organo-lead halide perovskite solar cells, and antibiotics [1–3]. Historically, such discoveries have often been serendipitous, involving a combination of intuition and the Edisonian approach, because of difficulties with predicting a material's properties or reverse engineering materials with desired properties. The Edisonian approach is inefficient for exploring a broad landscape of cross-correlated parameters, and therefore, it fails to spot many unexplored materials with potentially unique properties [4]. Resent studies estimate number of chemically feasible quaternary inorganic compounds between 10^{10} and 10^{13} [5,6]. Given the large number of candidate materials, it is impossible to synthesize and evaluate all possible experimental conditions or device configurations. An analogous problem is faced by the drug industry, where computational medicinal chemistry and cheminformatics approaches rely on virtual screening of chemical libraries for rational discovery of novel bioactive compounds [7–11]. Thus, emerging materials-informatics approaches offer an opportunity to leverage available databases and transform the serendipitous discovery process into a data- and knowledge-driven rational design and synthesis strategy, which can accelerate the identification of materials with desired properties [12–22].

The development of solar-energy materials is an example of a challenge where a particularly stringent set of materials properties must be satisfied to achieve high solar-to-electric power-conversion efficiencies (PCEs). For the dye sensitized solar cell (DSSC) research community, a long-standing problem has been the low performance of the p-type solar cells and photocathode materials, which are needed for tandem devices [23,24]. Typically p-type DSSCs use NiO as the photocathode, but there has not been a sustained rise in PCE above 4% since the initial report in 1999 in part due to the consistently low fill factor [25–30]. Newer photocathodes have focused on a relatively small subset of potential semiconductors chosen primarily by their similarity in crystal

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structure (*e.g.* delafossites) or elemental composition (*e.g.* Cu(I)) [31–36]. Despite significant effort to replace NiO, thus far no candidate material has surpassed NiO in overall performance metrics.

Here, we have applied materials informatics to this problem and employed a virtual screening of ~50,000 materials representing a majority of known stoichiometric inorganic compounds. This unique computational exercise was enabled by the use of novel materials descriptors reported in our recent publication [37]. We have identified $PbTiO_3$, which does not contain any typical features characteristic of a p-type DSSC, as the top computational "hit" material [37]. Experimentally fabricated p-type DSSC devices demonstrated record fill factors when $PbTiO_3$ was used with an aqueous electrolyte. The success of this proof of concept study opens the door for the expansive use of materials informatics, relying on materials descriptors, for designing novel compounds with improved physical chemical properties for a wide range of applications [37].

2. Experimental methods

2.1. Material fingerprints

Using our recent approach, we encoded the electronic structure diagram for each material as band structure fingerprints [37]. Along every special *k*-point along the wave-vector, the energy diagram was discretized into 32 bins serving as our fingerprint array. It is worth emphasizing that each of the 14 different Bravais lattices has a unique set of *k*-points. The comparison of a set of *k*-points belonging to a single Bravais lattice type will be considered a symmetry-dependent band structure fingerprint. To name a few examples, the Brillouin zone path of a Cubic Lattice (Γ–*X*–*M*–Γ–*R*–*X*|*M*–*R*) will be encoded with just four points (Γ, *M*, *R*, *X*) giving rise to a fingerprint array of length 128. Body-centered orthorhombic lattice is much more complex (Γ–*X*–*L*–*T*–*W*–*R*–X_1–*Z*–Γ–*Y*–*S*–*W*|L_1–*Y*|Y_1–*Z*) and represented by 13 points (Γ, L, L_1, L_2, R, S, T, W, X, X_1, Y, Y_1, Z) or fingerprint array of length 416. Conversely, the comparison of identical *k*-points not specifically belonging to any Bravais lattice is always possible when considering the Γ point as it is common to all lattice types. In present work, we limited our models only to the Γ point of the band structure fingerprint.

2.2. Similarity search

Among many chemical similarity metrics used in cheminformatics, Tanimoto similarity coefficient, T_c, between chemicals A and B is the most widely used [38]. It is calculated as shown in Eq. (1),

$$T_c = \frac{\sum_{j=1}^{n} x_{jA} x_{jB}}{\left(\sum_{j=1}^{n} (x_{jA})^2 + \sum_{j=1}^{n} (x_{jB})^2 - \sum_{j=1}^{n} x_{jA} x_{jB}\right)} \quad (1)$$

where x_j is the value of the *j*th descriptor and *n* is the total number of descriptors. Tanimoto similarities range from 0.0 (no similarity between chemicals A and B) to 1.0 (A and B are identical). The known p-type photocathodes NiO, Co_3O_4, Cu_2O, CuI, $CuAlO_2$, $CuGaO_2$, $NiCo_2O_4$, and $ZnCo_2O_4$ were used as reference query materials.

2.3. Data sources

We extracted band structure data for 46,936 materials from the AFLOWLIB repository, which represents approximately 60% of known stoichiometric inorganic materials listed in the Inorganic Crystal Structure Database (ICSD) [15,39]. All referenced DFT calculations were performed with the generalized gradient approximation (GGA) PBE exchange-correlation (XC) functional and projector-augmented wavefunction (PAW) potential according to the AFLOW standard for high-throughput computing [40]. This standard ensures reproducibility of the data, as well as provides visibility and reasoning for any parameter set used in calculations, such as accuracy thresholds, calculation pathways, and mesh dimensions.

2.4. $PbTiO_3$ film fabrication

All reagents were purchased from Fisher Scientific and used without further purification unless noted otherwise. In an inert, dry atmosphere lead acetate anhydrous (0.075 g) was combined with glacial acetic acid (2 mL). Then, titanium isopropoxide (0.05 mL) was combined with anhydrous ethanol (8 mL). The two solutions were combined and the resulting sol was capped and removed from the glove box. To create structured films, 10 wt.% ethyl cellulose (22 cp) was added to the precursor sol solution. A drop of the sol gel solution was wiped across fluorine doped tin oxide (FTO) glass substrates (12–14 Ω/cm^2) purchased from Hartford glass (TEC15) and cleaned with typical organic solvents and sonication. To make thin films, 5 layers of the sol solution were spun onto FTO glass substrate, with a 1 min 150 °C heat treatment between each layer. All films were then annealed in air for 70 min at 600 °C then cooled to room temperature.

2.5. DSSC assembly

Processed films were submerged in a 0.5 mM P1 (Dynamo) in acetonitrile solution for 20 min. Pt counter electrodes were fabricated by drop-casting H_2PtCl_6 in isopropanol solution onto an FTO glass slide, which included a powder-blasted pinhole, and annealing for 30 min at 380 °C. The DSSC was fabricated by sandwiching a 20 μm Surlyn polymer gasket (Solaronix) between the working P1 dyed $PbTiO_3$ electrode and the Pt counter electrode using a custom-built heating apparatus. The devices were backfilled with electrolyte using a custom-built vacuum chamber. The electrolyte consists of 1.0 M LiI and both the solvent and the amount of I_2 was varied as noted in text. The pinhole was sealed with a microscope coverslip and additional Surlyn polymer.

2.6. Analytical methods

Film morphology, thickness, and crystal structure were measured with a Hitachi S-4700 cold-cathode field-emission scanning electron microscope (SEM), a Bruker DektakXT profilometer, and a Rigaku Multiflex x-ray diffractometer with Cu source, respectively. X-ray diffraction spectra were collected from 20° to 60° 2θ at a scan rate of 2° 2θ/min. Transmission electron microscopy (TEM) and scanning TEM (STEM) images were obtained with an FEI Tecnai Osiris operating at 200 kV with a sub-nanometer probe and equipped with a Super-X EDS system. This system was used to acquire drift-corrected energy dispersive x-ray spectroscopy (EDS) elemental maps of $PbTiO_3$ particles using the Bruker Esprit software, and standardless Cliff-Lorimer analysis was performed on the deconvoluted EDS spectra for elemental quantification. Optical properties of each film were measured in a Cary 5000 UV–Vis–NIR from Agilent with integrating sphere attachment. Dye desorption measurements and the absorption profile of dye-loaded films in solution were taken using a Hewlett-Packard UV-Vis-NIR absorption spectrometer HP 8453. Macroscale pictures were taken using an iPhone 5c. Fluorescence measurements were performed on dye-loaded films using a 532 nm laser coupled to a Nikon D-Eclipse C1 SI microscope with a CCD detector. EIS and Mott-Schottky analysis were done using a Gamry Reference 600 electrochemical impedance spectrometer. EIS was performed on DSSCs under 1-sun

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