High-throughput Screening and Machine Learning of Double Perovskite Chalcogenides for Photovoltaics and Light Emission

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Perovskite PV and Light Emission

Perovskites have a unique set of properties that makes them suitable for photovoltaics and light emission. These include strong solid absorption, low defect density, high carrier mobility and high photoluminescence efficiency. Moreover, perovskite-based solar cells can be fabricated at a much lower temperature than Silicon-based ones. However, like Silicon-based photovoltaic devices, they degrade when they are exposed to moisture or ultraviolet radiation.

Therefore, over the past few years, perovskite-based solar cells have experienced a steady and strong increase in power conversion efficiency and are now competing with Silicon-based devices.

The current record power conversion efficiency is at 22.1% for perovskite-based solar cells and at 25.6% for c-Si-based solar cells.

Compounds considered in the initial screening

\( A^{2+} = \{ \text{Mg, Ca, Mn, Fe, Co, Ni, Cu, Sr, Ba} \} \)

\( B^{3+} = \{ \text{Sc, Cr, Fe, Co, Ga, In, Sb, Bi} \} \)

\( X^- = \{ \text{S, Se, Te} \} \)

We initially considered 720 different double perovskite chalcogenides and their 63 different binary precursors \( (A \times B \times X) \).

Goals

1. Initial screening with DFT for double perovskite chalcogenides that have a negative formation energy w.r.t binary precursors and a band gap in the range for photovoltaics and light emission.
2. Machine Learning through formation energies to predict stable double perovskites that were not included in the initial screening.

Results of the initial screening

➢ We find 3 stable compounds: SrFeVS, BaFeVS, and BaFeSbSe.
   However, these compounds are found to be unstable when taking into account the secondary decomposition of FeSb into FeS and SbF.

➢ We can see that the most stable compounds all have either Barium or Strontium in site A. Moreover, our 16 most stable compounds (formation energy < 0.1 eV/atom) have a Goldschmidt tolerance factor \( t \in [0.92, 1.02] \); this is in good agreement with reported double perovskite oxides, for which \( t \in [0.85, 1.07] \).

➢ Most compounds are either metals or semimetals. This is also observed in the literature for simple perovskites, but not for reported double perovskite oxides, which are mostly semiconductors or insulators.

➢ Median formation energy: 0.846 eV/atom

Results of the Machine Learning predictions

➢ The median formation energy of our ML predictions is at 0.696 eV/atom, so supervised learning was able to outperform our chemical intuition.

➢ ML predicts 9 previously unreported stable compounds.

➢ More than half the ML predictions are double perovskite oxides, so we note that ML is able to extrapolate well.

➢ Eight out of nine stable compounds have Barium in site A, which confirms the tendency observed in the initial screening.

➢ We observe a net improvement of the ML prediction ability during our last (6th) round, therefore showing the supervised learning algorithms are indeed learning from the available data to improve their predicting capability.

Compounds included in the initial screening

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Workflow

High-throughput screening

Calculate formation energies w.r.t binary precursors and band gaps of 720 different double perovskite chalcogenides.

Supervised Learning

Train on available DFT data

Screen periodic table

Suggest new stable candidates

Density Functional Theory

Calculate formation energies and band gaps of Machine Learning candidates

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Summary & Conclusions

We found 9 previously unreported stable double perovskites, all predicted by Machine Learning.

\( \text{PbInSbO}_6 \), \( \text{BaBiAsO}_4 \), and \( \text{BaSbAsO}_4 \) were found to be stable semiconductors with a band gap of 2.92 eV, 2.69 eV and 1.77 eV, respectively.

All three have potential applications in photovoltaic and light-emitting devices. \( \text{BaBiPO}_4 \) was found to be a stable insulator with a band gap of 4.02 eV. \( \text{BaAuAsO}_4 \) was found to be a metal. \( \text{BaInPO}_4 \), \( \text{BaTiAsO}_4 \), \( \text{BaTiPO}_4 \) and \( \text{BaInAsO}_4 \) were found to be semimetals.

However, if the latter can be quantum confined, they could potentially be used as well for photovoltaics and light emission.

In conclusion, we have seen that most double perovskite chalcogenides considered here are unstable with respect to their binary precursors and have either metallic or semimetallic character. We observe that the most stable compounds all have either Barium or Strontium in site A and we suppose that the presence of the chalcogen anion instead of the oxygen anion in site X reduces the band gap of double perovskites.

We also conclude that Machine Learning has outperformed our chemical intuition. Indeed, Machine Learning was able to predict 9 stable compounds out of 80, whereas our initial high-throughput screening yielded 3 stable compounds out of 720. We expect that Density Functional Theory and Machine Learning will become more and more often used in synergy in order to accelerate materials discovery.