Exploring Cs-Te Photocathode Materials via High-Throughput Density Functional Theory Calculations
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Vacuum

Surface

Point defects

Grain boundaries

1st Step: Photoexcitation

\( h_\nu \)

VBs

CBs

\( e^- \)

\( h^+ \)

2nd Step: Scattering

3rd Step: Overcoming Surface

Surface Roughness

External electric fields

Contaminations

Vacuum

Bulk

Cocchi and Saßnick, \textit{Micromachines} 2021, 12(9), 1002; doi: 10.3390/mi12091002

<table>
<thead>
<tr>
<th>Experimental Composition/Phase of Cs-Te</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Formula</strong></td>
</tr>
<tr>
<td><strong>Crystal system</strong></td>
</tr>
<tr>
<td><strong>Space group</strong></td>
</tr>
</tbody>
</table>
Theoretical Framework within DFT

☑ The Kohn-Sham equations in density functional theory map the many-body problem onto an "auxiliary" system of independent electrons:

\[
\begin{bmatrix}
-\frac{1}{2}\nabla^2 + v_s(r)
\end{bmatrix}
\varphi_i(r) = \epsilon_i^{KS} \varphi_i(r).
\]

☑ With the effective potential \(v_s\):

\[v_s(r) = v_{\text{ext}}(r) + v_H(r) + v_{xc}(r),\]

☑ The most crucial approximation in KS-DFT originates from the exchange-correlation potential \(v_{xc}\), here we use

☑ Meta-generalized gradient approximation (meta-GGA): SCAN

☑ DFT calculations have been performed with CP2K.


☑ The high-throughput workflow has been designed using AiiDA.


Cs-Te Materials

Stability:

<table>
<thead>
<tr>
<th>Formula</th>
<th>Space group</th>
<th>$\Delta E$ (eV/at.)</th>
<th>Band gap (eV)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cs$_8$Te$_4$</td>
<td>P1 [1]</td>
<td>0.000</td>
<td>2.26</td>
<td>OQMD</td>
</tr>
<tr>
<td>Cs$_8$Te$_4$</td>
<td>Pnma [62]</td>
<td>0.000</td>
<td>2.26</td>
<td>MP</td>
</tr>
<tr>
<td>Cs$_2$Te$_1$</td>
<td>Fm$\bar{3}$m [225]</td>
<td>0.002</td>
<td>2.14</td>
<td>OQMD</td>
</tr>
<tr>
<td>Cs$_{18}$Te$_9$</td>
<td>R3 [146]</td>
<td>0.004</td>
<td>1.96</td>
<td>OQMD m.</td>
</tr>
<tr>
<td>Cs$_4$Te$_2$</td>
<td>Fdd2 [43]</td>
<td>0.018</td>
<td>2.16</td>
<td>MP m.</td>
</tr>
<tr>
<td>Cs$_4$Te$_2$</td>
<td>Cmc$2_1$ [36]</td>
<td>0.018</td>
<td>2.16</td>
<td>MP m.</td>
</tr>
<tr>
<td>Cs$_4$Te$_2$</td>
<td>P$6_3$/mmc [194]</td>
<td>0.018</td>
<td>2.16</td>
<td>MP m.</td>
</tr>
</tbody>
</table>

The 2:1 composition:

Saßnick and Cocchi, in preparation, 2021
Electronic structure vs. stoichiometry

- \( \text{Te}_3 - P3_21 \) [154] (trigonal)
- \( \text{Cs}_4 \text{Te}_{10} - Cmcm \) [63] (orthorhombic)
- \( \text{Cs}_4 \text{Te}_4 - Pbam \) [55] (orthorhombic)
- \( \text{Cs}_8 \text{Te}_4 - Pnma \) [62] (orthorhombic)
- \( \text{Cs}_3 \text{Te}_1 - Pm\bar{3}m \) [221] (cubic)
- \( \text{Cs}_1 - Im\bar{3}m \) [229] (cubic)

0 eV is set to the valence band maximum (VBM).

Saßnick and Cocchi, in preparation, 2021
Summary and Conclusions

Summary

▶ First approximation of the stability of different phases.
▶ Analysis of relationship between compositional changes and electronic properties.
▶ Going beyond state-of-the-art by using an advanced approximation for the exchange-correlation functional.

Conclusions

▶ Additional (theoretically) stable structures found at experimentally relevant compositions.
▶ The largest band gap is reached at a composition of 2:1 due to fully occupied Te p-states for Cs-Te.

Open Positions

Student internship

High-throughput screening of binary alkali-based photocathode materials
https://www.daad.de/rise/en/rise-germany/find-an-internship/

Experimental Collaborators

Dr. Sven Lederer & Group

Sponsors

Project-id: nic00069