Exploring high-performance Al-O based thermoelectric materials working at high temperatues using first-principles calculations in combination with machine learning

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Thermoelectric (TE) conversion is one of the waste-heat utilization technologies. Recently, although thermoelectric devices working at room temperature have been attracting attention for plant monitoring and wearable devices, those working at high temperatures, which are effective for reducing CO_2 emissions and saving energy, have not been investigated extensively. We have focused on aluminum oxide (Al-O) based materials, because they have excellent heat-resistance and are easy to perform stable substitutional doping even around 900 K. However, since 3,447 Al-O based materials are recorded in the crystal structure database named "Materials Project [1] (MP)", it is not realistic to calculate all of them using first-principles calculations. In this study, we aim to efficiently search for high-performance Al-O based thermoelectric materials working at 900 K in combination with machine learning.

To prepare a training data set used for a neural network model (NNM), we selected 109 samples among 3,447 crystal structures in MP. We performed VASP [2] and BoltzTraP [3] calculations to evaluate their electric conductivity (σ), Seebeck coefficient (S), and electron-mediated thermal conductivity (κ_e). The lattice thermal conductivity (κ_l) was estimated using the formula proposed by Yan *et al* [4]. Eventually, we obtained the figure of merit [$ZT = \sigma S^2 T/(k_e+k_l)$] for each p-type TE compound. Then we optimized and used a NNM to predict the ZT and thermoelectric conversion efficiency (h) of residual 3,338 Al-O based materials. As shown in Table 1, we efficiently explored four high-TE performance candidates with a melting point exceeding 2,000 K that is enough higher than 900 K [5].

We will present details of NNM optimization procedures and discuss the reason behind the high ZT by examining the structrue–property correlations in our talk.

Table 1. Four high-TE performance candidates explored among 3,447 Al-O-based materials using the optimized NNM.

Materials ID	Formula	ZT	η [%]
mp-4051	$AlPO_4$ ($C222_1$)	0.766	13.19
mp-7848	$AlPO_4$ ($I\bar{4}$)	0.742	12.90
mp-5724	$AlPO_4$ ($P2_1$)	0.481	9.32
mp-7849	$AlAsO_4$ ($I\bar{4}$)	0.465	9.08

References

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