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論文概要

Thermoelectric Properties of Molecular Solids
(有機低分子結晶の熱電特性)

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This thesis is entitled “Thermoelectric Properties of Molecular Solids”, written in English and constructed by six chapters.

Chapter 1 starts from history and recent progress of thermoelectric materials. To reveal strategy to obtain good thermoelectric materials, general expression of physical parameters associated with thermoelectric efficiency is discussed.

Introduction to common chemicals and general experimental methods are given in Chapter 2.

In Chapter 3, Seebeck coefficient of β-$(CH_3)_4N[Pd(dmit)_2]_2$ (dmit: 1,3-dithiole-2-thione-4,5-dithiolate) has been measured and calculated using the Boltzmann equation. Although the highest occupied molecular orbital (HOMO) of the title compound makes the conduction band, the obtained Seebeck coefficient is negative, which agrees with the theoretical prediction. Since the compound has 1− charge on the dimer, theoretical calculation including the Coulomb repulsion has been conducted. The calculation reproduces the experimental result, which supports validity of the calculation even in such an anomalous metal state.

In Chapter 4, thermoelectric generators (TEGs) using high-conducting charge-transfer complexes have been demonstrated. These TEGs show ordinary operation where the power output behaves as a square of the current. In particular, a TEG using $(TMTSF)_2PF_6$ (TMTSF: tetramethyltetraselenafulvalene) and $Cu(DMDCNQI)_2$ (DMDCNQI: 2,5-dimethyl *N*,*N*’-dicyanoquinonediimine) achieves a maximum power output of 36 μW cm^{-2} with a temperature difference of 10 K. This value is quite high among organic TEGs. The power output decreases with lowering the temperature due to influence of the contact resistance. This reveals that energy-level matching between the thermoelectric material and electrode is important.

In Chapter 5, field-effect modulated Seebeck coefficient has been measured in rubrene single crystals. A novel measurement system using laser heating which enables fast and accurate Seebeck coefficient measurement in a field-effect transistor structure has been developed. To obtain a comprehensive relation between electrical conductivity and Seebeck coefficient, thermoelectric properties at various carrier concentration have been measured down to low temperature. The hopping-type transport at low temperatures is discussed in view of the quantity of the trap states.

These results provide meaningful information of thermoelectric properties of molecular solids, which contributes to the development of thermoelectrics.