Investigation for Thermoelectric Properties of the MoS$_2$ Monolayer–Graphene Heterostructure: Density Functional Theory Calculations and Electrical Transport Measurements

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ABSTRACT: We investigated the thermoelectric (TE) properties of the MoS$_2$ monolayer–graphene heterostructure which consists of the MoS$_2$ monolayer and graphene. The electronic structures of the MoS$_2$ monolayer–graphene heterostructure are mainly contributed from graphene and the MoS$_2$ monolayer for the valence band maximum and conduction band minimum, respectively. The change in the electronic structures near the Fermi level is responsible for the fact that the calculated Seebeck coefficients $S$ and electrical conductivity $\sigma/\tau$ of MoS$_2$ monolayer–graphene are largely affected from those of graphene and the MoS$_2$ monolayer. Its power factor $S^2\sigma/\tau$ is increased compared to those of graphene and the MoS$_2$ monolayer at an electron concentration of $10^{11}$ to $10^{12}$ cm$^{-2}$, which corresponds to a three-dimensional concentration of $3 \times 10^{18}$ to $3 \times 10^{19}$ cm$^{-3}$. We also demonstrated that the MoS$_2$ monolayer shows the p-type TE behavior, while the MoS$_2$ monolayer–graphene heterostructure is given to the n-type TE material. The current study provides a strategy to improve TE properties of the MoS$_2$ monolayer through the formation of the MoS$_2$ monolayer–graphene heterostructure.

1. INTRODUCTION

Thermoelectric (TE) materials have been in spotlight as a new potential source of renewable energy. They enable the conversion of thermal and electrical energy, which can be used for both power generation and refrigeration. The TE efficiency is described by the dimensionless figure of merit, $zT = (S^2\sigma T)/\kappa$, where $S$, $\sigma$, $T$, and $\kappa$ represent the Seebeck coefficients, electrical conductivity, temperature, and thermal conductivity, respectively. The method to improve the $zT$ is increasing the power factor (PF) $S^2\sigma$ and reducing the thermal conductivity. Because the thermal conductivity of the potential TE materials are approaching to the minimum theoretical limit on the journey to higher $zT$ values, a paradigm shift is necessary toward the enhancement of TE PF, which is affected by the carrier (electron or hole) concentration and the electronic structures.

Two-dimensional (2D) transition metal dichalcogenides (TMDCs) such as MoS$_2$ are reported to have great potential in flexible electronics, optoelectronics, energy storage, and power nanodevices because of their unique electronic properties such as a suitable direct band gap. TMDCs also have been studied as candidates for TE materials because of their characteristic geometric and electronic structures. TMDCs usually possess very low thermal conductivity $\kappa$ and high Seebeck coefficients $S$ along a direction perpendicular to the 2D lattice layers. Although MoS$_2$ has high Seebeck coefficients $S$ (700–900 $\mu$V/K), it has a poor electrical conductivity $\sigma$ which is too small to expect high PF.$^5,7$ Therefore, a major challenge for enhancing the TE performance of TMDCs is to improve their electrical conductivity. Previous density functional theory (DFT) studies showed that the TE properties can be improved in 2H-MoQ$_2$ (Q = S and Se) based on the layer mixing of compounds such as MoS$_2$/MoSe$_2$ heterostructures.$^8$ It is also experimentally investigated that the 2D TMDCs materials are ideal candidates for thermal management and TE applications.$^9$

Graphene has shown high electrical conductivity, high carrier mobility, and high mechanical properties.$^{10,11}$ One can speculate that the fabrication of TE materials with graphene could help to enhance the electrical conductivity. Recently, it has been reported that the PF can be greatly improved by applying graphene coating to TE materials with low electrical conductivity, where electrical conductivity is complemented by graphene with high electrical conductivity and mobility.$^{12,13}$ Also, it is reported that the PF on the Bi$_{24}$Sb$_{15}$Te$_{8}$Gr$_{0.08}$ (Gr: graphene) composite has been improved up to 3.7 mW/mK.$^{14}$ Agarwal’s group reported...
that electrical conductivity of the $\text{Bi}_2\text{Te}_3$ is strongly enhanced by constructing the nanocomposite with graphene, although the Seebeck coefficients of nanocomposite $\text{Bi}_2\text{Te}_3$–graphene are reduced compared to the pristine $\text{Bi}_2\text{Te}_3$. Therefore, the fabrication of low electrical conductivity materials with graphene would be a useful way to improve the PF of TE materials.

In this work, we focused on the TE properties of the MoS$_2$ monolayer–graphene heterostructure in comparison to those of the pristine MoS$_2$ monolayer and graphene. Ebnonnasir’s group suggested that the MoS$_2$ monolayer–graphene heterostructures are suitable for photovoltaic devices because of interfacial electronic transfer between the MoS$_2$ monolayer and graphene layer which should be related to the increase in electrical conductivity. We expect that the formation of the heterostructure with graphene would possibly enhance the PF of the MoS$_2$ monolayer by increasing electrical conductivity.

Using DFT calculations, we investigated the Seebeck coefficients $S$ and electrical conductivity $\sigma/\tau$ in the MoS$_2$ monolayer–graphene heterostructure. Our theoretical studies demonstrate that the electrical conductivity $\sigma/\tau$ and the PF $S^2\sigma/\tau$ of the MoS$_2$ monolayer–graphene heterostructure are considerably enhanced. With this result, we suggested that the heterostructurization of the MoS$_2$ monolayer with high electrical conductivity material graphene should be considered as a promising way to enhance the TE properties of the MoS$_2$ monolayer.

2. METHODS

In our DFT calculation, we employed the frozen-core projector-augmented-wave method, encoded in the Vienna Ab initio Simulation Package (VASP) code under the generalized gradient approximation of Perdew–Burke–Ernzerhof and the $k$-mesh of $30 \times 30 \times 1$ in the Brillouin zone. Figure 1 shows a hypothetically constructed system in which we consider the $4 \times 4$ supercell and $3 \times 3$ supercell of graphene and the MoS$_2$ monolayer, respectively.

We optimized the atomic positions of MoS$_2$ monolayer–graphene heterostructures on the basis of DFT calculations, while keeping the cell parameters under the self-consistent field convergence thresholds of $10^{-4}$ eV and 0.05 eV/Å for the total electronic energy and force, respectively. Note that we focus on how to enhance the TE properties in the MoS$_2$ monolayer, so as to better describe the change in electronic structure and TE properties of the MoS$_2$ monolayer, and the lattice constant of the MoS$_2$ monolayer–graphene heterostructure is used as the lattice constant of MoS$_2$ ($\text{The strain in graphene is } 3.23\%$.)

We also investigate the optimum interlayer distance by employing the vdW-DF scheme. The result reveals that the optimal interlayer distance in the MoS$_2$ monolayer–graphene heterostructure is $\sim 3.3$ Å, presented in Figure S1 of the Supporting Information, and this result is in agreement with previous theoretical studies.

3. RESULTS AND DISCUSSION

To quantitatively characterize the mechanical properties of the MoS$_2$ monolayer–graphene heterostructure shown in Figure 1, the binding energy ($E_b$) per atom between the MoS$_2$ monolayer and graphene layer is calculated as

$$E_b = \frac{E_{\text{MoS}_2,\text{graphene}} - (E_{\text{MoS}_2} + E_{\text{graphene}})}{N}$$

where $E_{\text{MoS}_2,\text{graphene}}$, $E_{\text{MoS}_2}$, $E_{\text{graphene}}$, and $N$ refer to the total energies of a MoS$_2$ monolayer–graphene heterostructure, a MoS$_2$ monolayer, an isolated graphene, and the number of atoms in the considered unit cell ($N = 59$), respectively. The calculated binding energy per atom is $-86$ meV. The negative binding energy denotes that the heterostructure configuration of graphene and MoS$_2$ is thermochemically stable.

The electronic structures calculated for the MoS$_2$ monolayer–graphene heterostructure are summarized in Figure 2, in which the red, black, and blue lines refer to total density of states (DOS), the projected DOS for the MoS$_2$ monolayer, graphene layer, and total DOS, respectively. The inset shows the calculated DOS for the pure MoS$_2$ monolayer (yellow line) and projected DOS for the MoS$_2$ monolayer in the heterostructure (black line).

Figure 1. Perspective view of the crystal structure of MoS$_2$ monolayer–graphene. The Mo, S, and C atoms are shown as purple, yellow, and blue balls, respectively. The dashed line indicates the surface unit cell. (a) Top view and (b) side view. The interlayer distance is 3.3 Å.

Figure 2. Electronic structure calculated for the MoS$_2$ monolayer–graphene heterostructure. The black, blue, and red solid lines refer to the projected DOS for the MoS$_2$ monolayer, graphene layer, and total DOS, respectively. The inset shows the calculated DOS for the pure MoS$_2$ monolayer (yellow line) and projected DOS for the MoS$_2$ monolayer in the heterostructure (black line).
Note that the Dirac point of graphene is placed at the conduction band minimum (CBM) of MoS$_2$, which results in the semimetallic band structures. The valence band maximum is mainly contributed from graphene, while the CBM is composed of graphene and the MoS$_2$ monolayer. Thus, the transport properties for the holes are governed by graphene, while those for electrons are influenced from both graphene and the MoS$_2$ monolayer. Interestingly, the DOS peak is generated at the edge of the CBM in the MoS$_2$ monolayer–graphene heterostructure, deducing the enhancement of TE properties, while there are no DOS peaks at the edge around the Fermi level for the pure MoS$_2$ monolayer and graphene.

TE properties were calculated using the BoltzTraP code, which is based on Boltzmann transport theory under rigid band approximation and constant relaxation time approximation. Figure 3 shows the calculated in-plane Seebeck coefficients $S$, electrical conductivity $\sigma/\tau$, and PF $S^2\sigma/\tau$ for the MoS$_2$ monolayer (black), graphene (blue), and the MoS$_2$ monolayer–graphene heterostructure (red) at 300 K are plotted depending on their carrier (hole and electron) concentration, in which $n_h$ and $n_e$ indicate the hole and electron carrier concentrations, respectively. The left column [(a,c,e)] is the TE property for the hole carrier p-type, while the right column [(b,d,f)] is that for the electron carrier n-type.

Even if the carrier type of the MoS$_2$ monolayer in experiments would be modulated by the substrate, according to the theoretical study, the MoS$_2$ monolayer was reported as a p-type TE material. Because the effect of the substrate is not included in the theoretical study, we considered the intrinsic carrier type of the MoS$_2$ monolayer as the p-type. Indeed, our calculated Seebeck coefficients $S$ for the MoS$_2$ monolayer are given as $\sim 700 \mu$V/K at $n_h = 1.0 \times 10^{11}$ cm$^{-2}$ and $-510 \mu$V/K at $n_e = 1.0 \times 10^{11}$ cm$^{-2}$ (corresponding to $\sim 3 \times 10^{13}$ cm$^{-3}$ in a three-dimensional concentration) at 300 K. The measured Seebeck coefficients $S$ for graphene are $\sim 80 \mu$V/K at $n_e = 5 \times 10^{11}$ cm$^{-2}$ at 300 K. The Seebeck coefficients $S$ of the MoS$_2$ monolayer–graphene heterostructure calculated as a function of the carrier concentration at 300 K are shown in Figure 3a,b, which exhibits that the calculated maximum Seebeck coefficients are $\sim 112 \mu$V/K at $n_h = 3 \times 10^{11}$ cm$^{-2}$ and $-169 \mu$V/K at $n_e = 3 \times 10^{11}$ cm$^{-2}$. The calculated Seebeck coefficients $S$ of the MoS$_2$ monolayer–graphene heterostructure are much smaller than that of the MoS$_2$ monolayer in which large Seebeck coefficients $S$ of the MoS$_2$ monolayer are caused from the wide band gap, implying poor electrical conductivity. However, it is slightly bigger than that of pure graphene for the electron carrier. In the MoS$_2$ monolayer–graphene heterostructure, the Seebeck coefficients $S$ for the electron carrier are larger than the Seebeck coefficients for the hole carrier, indicating that the MoS$_2$ monolayer–graphene heterostructure should possess n-type TE properties.

The dependence of $\sigma/\tau$ on the electron carrier concentration and that on the hole carrier concentrations for the MoS$_2$ monolayer, graphene, and the MoS$_2$ monolayer–graphene heterostructure are presented in Figure 3c,d, which reveals that the calculated $\sigma/\tau$ for the MoS$_2$ monolayer is much smaller than that of others, and the $\sigma/\tau$ of MoS$_2$ monolayer–graphene heterostructure is slightly higher than that of graphene for the electron and hole carriers. As the reason for the increase of $\sigma/\tau$ in the MoS$_2$ monolayer–graphene heterostructure, two observations are important. One is the generation of the DOS peak at the edge of the CBM which is possible to predict the increase in carrier concentration, and it would play an important role in enhancing $\sigma/\tau$ for the electron carrier. The second observation is a charge transfer between layers. To elucidate the charge transfer between layers, we integrated the projected DOS up to the Fermi level for the MoS$_2$ monolayer and graphene layers in the MoS$_2$ monolayer–graphene heterostructure. For comparison, the integral of DOS from the valence-band bottom to the Fermi level was also carried out for the pure MoS$_2$ monolayer and graphene. Note that the integrated DOS gives the number of valence electrons in the unit cell. The ratio of the integrated DOS of the pure MoS$_2$ monolayer to that of pure graphene is about 1.27, while the ratio of the integrated projected DOS of the MoS$_2$ monolayer to that of graphene in the MoS$_2$ monolayer–graphene heterostructure is 1.92. The ratio of the integrated DOS of the MoS$_2$ monolayer to that of graphene in the MoS$_2$ monolayer–graphene heterostructure is greater than the ratio in the pure MoS$_2$ monolayer and graphene. This reveals that the charge transfer occurs from the graphene layer to the MoS$_2$ monolayer in the MoS$_2$ monolayer–graphene heterostructure, resulting in increased carrier concentration and carrier mobility. This led to significantly improved electrical conductivity $\sigma$ for the hole carrier in the MoS$_2$ monolayer–graphene heterostructure.
Under the assumption that the relaxation time $\tau$ is energy-independent, the calculated PF $S^2\sigma/\tau$ is presented as a function of the carrier concentration in Figure 3 e,f. The carrier concentration dependence of the PF/\tau calculated for the MoS$_2$ monolayer−graphene heterostructure shows two peaks at hole carrier concentration $n_h = \sim 4 \times 10^{10}$ cm$^{-2}$ and electron carrier concentration $n_e = \sim 3 \times 10^{11}$ cm$^{-2}$. The PF/\tau for the electron carrier is considerably greater than that for the hole carrier (e.g., the n-type PF/\tau is larger than the p-type PF/\tau by a factor of $\sim 3$). We note that the electronic states are crucial for determining the TE properties in the MoS$_2$ monolayer−graphene heterostructure. Because of the increase in carrier concentration and electrical conductivity, the MoS$_2$ monolayer−graphene heterostructure has strongly enhanced TE properties.

Figure 4a,b shows the synthesized sample of MoS$_2$ with the reduced graphene oxide (RGO) hybrid. Figure 4a shows a bright-field transmission electron microscopy (TEM) micrograph of exfoliated MoS$_2$ and (b) a high-resolution TEM image of MoS$_2$ with the RGO hybrid composite with an inset showing the sintered body. The experimental transport properties such as electrical conductivity of MoS$_2$ monolayer−graphene heterostructure were characterized using a TE property measurement system (TPMS, ZEM-3, ULVAC-RIKO), and the properties were measured along the direction perpendicular to the pressing axis of the spark plasma sintering process.

Figure 4c shows the measured electrical conductivity for samples which exhibited semiconducting behavior in their electrical conductivity. The measured electrical conductivity of MoS$_2$ without RGO does not have a large increase with temperature, and the measured electrical conductivity dramatically increases with the increasing RGO content. This demonstrates that graphene coating improved the electrical conductivity of MoS$_2$ and is a useful way in enhancing the electrical conductivity of MoS$_2$. Figure 4d shows the calculated electrical conductivity of the MoS$_2$ monolayer and the MoS$_2$ monolayer−graphene heterostructure at $n_e = 10^{12}$ cm$^{-2}$. The electrical conductivity for the MoS$_2$ monolayer does not change much with the change in temperature and the electrical conductivity of the MoS$_2$ monolayer−graphene heterostructure increases with increasing temperature, which are consistent with experiment.

4. CONCLUSIONS

Using DFT, we investigated the electronic and TE properties such as $S$, $\sigma/\tau$, and $S^2\sigma/\tau$ of the MoS$_2$ monolayer, graphene, and the MoS$_2$ monolayer−graphene heterostructure depending on the carrier concentration and temperature. Our study shows that the electrical conductivity in the MoS$_2$ monolayer−graphene heterostructure is strongly enhanced for both hole and electron carriers, which is not only caused by the charge transfer between layers but also by the generation of the DOS peak at the edge of the CBM. Because of the increased electrical conductivity, the MoS$_2$ monolayer−graphene heterostructure is expected to have strongly enhanced TE properties. Thus, we suggest that the heterostructurization of MoS$_2$ with high electrical conductivity material graphene should be
considered as a promising way to enhance the TE properties of MoS₂.

■ ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c04488.

Relative energy as a function of distance between MoS₂ and the graphene layer in the MoS₂–graphene heterostructure (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education (2017R1D1A1B03036257 and 2020R1A5A1019141), the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (no. 2016K1A4A4A01922028, 2020R1F1A1052898, and 2020M3H4A2084418), and Supercomputing Center/Korea Institute of Science and Technology Information with supercomputing resources including technical support (KSC-2017C-0002).

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