

Article

Enhanced Thermoelectric Performance of β -Ag₂Se/RGO Composites Synthesized by Cold Sintering Process for Ambient Energy Harvesting

Dulyawich Palaporn ¹, Ikhwan Darmawan ^{1,2} , Piyawat Piyasin ^{1,3} and Supree Pinitsoontorn ^{1,4,*} 

¹ Department of Physics, Faculty of Science, Khon Kaen University, Khon Kaen 40002, Thailand; dulaawit.p@kku.ac.th (D.P.)

² Graduate School of Pure and Applied Sciences, University of Tsukuba, Tsukuba 305-8571, Ibaraki, Japan

³ Global Zero Emission Research Center, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8569, Ibaraki, Japan

⁴ Institution of Nanomaterials Research and Innovation for Energy, Faculty of Science, Khon Kaen University, Khon Kaen 40002, Thailand

* Correspondence: psupree@kku.ac.th

Abstract

Silver selenide (Ag₂Se) is a promising *n*-type thermoelectric material for near-room-temperature energy harvesting due to its high electrical conductivity and low lattice thermal conductivity. In this study, Ag₂Se-based composites were synthesized using a cold sintering process (CSP), enabling densification at low temperature under applied pressure. Reduced graphene oxide (RGO) was incorporated into the Ag₂Se matrix in small amounts (0.25–1.0 wt.%) to enhance thermoelectric performance. Structural analysis confirmed phase-pure β -Ag₂Se, while SEM and TEM revealed homogeneous RGO dispersion and strong interfacial adhesion. RGO addition led to a reduced carrier concentration due to carrier trapping by oxygen-bearing functional groups, resulting in decreased electrical conductivity. However, the absolute Seebeck coefficient increased with RGO content, maintaining a balanced power factor. Simultaneously, RGO suppressed thermal conductivity to below 0.75 W m^{−1} K^{−1} at room temperature. The optimal composition, 0.75 wt.% RGO, exhibited the highest average *zT* of 0.98 across the temperature range from room temperature to 383 K. These results demonstrate that combining the CSP with RGO incorporation offers a scalable and cost-effective strategy for enhancing the thermoelectric performance of Ag₂Se-based materials.

Keywords: silver selenide; graphene; thermoelectric material; cold sintering process



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1. Introduction

In recent years, green energy technologies have gained significant attention as a means to support global sustainable development [1]. Traditional reliance on fossil fuels faces growing challenges due to resource depletion and environmental concerns. As a result, various strategies have emerged to generate clean power. Among them, thermoelectric technology enables direct conversion of heat into electrical energy through solid-state devices [2]. Thermoelectric generators can operate when a temperature gradient is established across a material, making them attractive for low-grade heat recovery and decentralized power generation. The efficiency of thermoelectric materials is quantified by the dimensionless figure-of-merit (*zT*), defined as:

$$zT = \frac{S^2\sigma}{\kappa}T \quad (1)$$

where S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the thermal conductivity [3]. The product $S^2\sigma$ is referred to as the power factor (PF), which reflects the material's ability to convert thermal energy into electrical power. Enhancing zT requires a delicate balance between these interdependent parameters.

Over the past decades, Bi_2Te_3 -based thermoelectric materials have been widely recognized as the most efficient candidates for near-room-temperature applications [4]. However, tellurium (Te) is an extremely scarce element and its price is expected to rise sharply if Te-based materials reach mass-market adoption [5]. As a result, Te-free alternatives are being actively explored.

Silver selenide (Ag_2Se) has emerged as a promising substitute for Bi_2Te_3 due to its high electrical conductivity, low lattice thermal conductivity, and narrow bandgap ($\sim 0.07\text{--}0.20$ eV) [6–8]. Below 407 K, Ag_2Se adopts an orthorhombic β -phase, which exhibits excellent thermoelectric properties. To further enhance its thermoelectric performance, various strategies have been employed, including precise stoichiometric control [9], doping [10], and nanostructuring [11].

Incorporating carbon-based materials has shown considerable potential for improving thermoelectric properties. Various carbon-based fillers, such as carbon nanotubes (CNTs) [12], multi-walled carbon nanotubes (MWCNTs) [13], and carbon black (CB) [14], have been explored to enhance electrical conductivity and phonon scattering. Among these, reduced graphene oxide (RGO) stands out due to its large surface area, which promotes uniform dispersion within the thermoelectric matrix and facilitates carrier concentration manipulation. Additionally, the high interface density introduced by RGO increases phonon scattering, effectively reducing thermal conductivity and improving zT [15,16]. For example, the integration of RGO into BiSbTe raised its zT from 1.09 to 1.29 at 300 K [17], while Thanh et al. reported a ~ 6.5 -fold enhancement in zT for La/Nb-doped SrTiO_3 upon RGO incorporation [18].

A recent study reported that incorporating 30 wt.% RGO into Ag_2Se via hydrothermal synthesis followed by hot-pressing significantly reduced the thermal conductivity to ~ 0.492 W/mK at 333 K, achieving a zT of ~ 0.39 [15]. However, such a high RGO content compromises the mechanical integrity of the composite, as excessive filler disrupts the connectivity between Ag_2Se grains, leading to reduced structural robustness. Moreover, the literature suggests that excessive RGO may suppress zT due to its tendency to attract charge carriers, thereby diminishing carrier mobility [19,20]. To address this, several studies have explored the use of low RGO concentrations to enhance the thermoelectric performance [21,22]. A small amount of RGO can effectively reduce thermal conductivity [23] while still maintaining sufficient carrier transport pathways [24,25]. This balance is crucial for optimizing zT without sacrificing mechanical or electrical integrity.

To consolidate thermoelectric materials, conventional sintering techniques such as spark plasma sintering (SPS) [26,27] and hot-pressing (HP) [28,29] are commonly employed. However, these methods require high sintering temperatures, which can induce grain growth, cause atomic diffusion, and degrade thermoelectric performance [30,31]. To overcome this limitation, the cold sintering process (CSP) has gained significant interest as a low-temperature densification technique [32]. The CSP utilizes a solvent to partially dissolve the target material, enabling particle rearrangement and densification at substantially reduced sintering temperatures. The solvent-assisted densification mechanism involves void filling between particles. Upon solvent evaporation, the dissolved material precipitates into these spaces, forming a dense microstructure [33,34]. A few thermoelectric materials have been consolidated using the CSP, for instance, $\text{Ca}_3\text{Co}_4\text{O}_9$ [35], Bi_2Te_3 [36],

and Cu_2Se [37]. Moreover, the CSP can be modified to incorporate dopants or composite materials by introducing them in liquid form—such as solutions or colloids—during sintering [8,38]. In a previous study, AgNO_3 solution was used as a Ag source to dope extra Ag^+ into a Ag_2Se system via the CSP, yielding highly dense samples with homogeneously distributed Ag dopants [39]. These findings demonstrate that the CSP offers a versatile and energy-efficient route for tailoring microstructure and thermoelectric properties of the materials by selecting appropriate solvents and processing conditions [40].

In this work, we leverage the advantages of the CSP to improve the dispersion of RGO within the Ag_2Se matrix, enabling a substantial reduction in the required RGO content while maintaining high thermoelectric performance. The use of a liquid-phase RGO colloid ensures uniform distribution throughout the matrix, minimizing agglomeration and maximizing interfacial phonon scattering. Furthermore, the low sintering temperature inherent to the CSP preserves the nanostructure of Ag_2Se and prevents RGO decomposition, eliminating the need for an inert atmosphere during processing. Consequently, a minimal RGO addition of just 0.75 wt.% was sufficient to significantly improve the thermoelectric properties of Ag_2Se , enhancing electrical performance without compromising structural integrity. These findings highlight the potential of this method for future energy-harvesting applications, particularly in low-temperature environments.

2. Materials and Methods

2.1. Materials

Silver selenide (Ag_2Se) powder was synthesized from elemental silver powder (Ag, 99.8% purity, Sigma-Aldrich, Saint Louis, MO, USA) and selenium powder (Se, 99.5% purity, Sigma-Aldrich, Saint Louis, MO, USA) without further purification. Reduced graphene oxide (RGO) was prepared according to our previous protocol [41].

2.2. Synthesis

Ag_2Se powder was fabricated via a wet ball-milling method. Ag and Se powders were mixed in a stoichiometric ratio corresponding to Ag_2Se . Yttria-stabilized zirconia (YSZ) balls with a diameter of 2 mm were loaded in a polypropylene vessel at a ball-to-powder mass ratio of 20:1, along with 15 mL of n-heptane as a milling medium. The mixture was milled at 250 rpm for 1 h. The resulting wet powder was then dried in an oven at 353 K for 24 h to obtain dry Ag_2Se powder.

To prepare composite pellets, Ag_2Se and RGO powder were manually mixed in a mortar, with RGO contents of 0.25, 0.50, 0.75, and 1.0 wt.%. Additionally, 0.1 mL of ethanol was added to facilitate mixing, and the blend was hand-grounded for 15 min. The mixture was then loaded into a stainless-steel mold and subjected to the cold sintering process (CSP) at 473 K under a uniaxial pressure of 500 MPa for 1 h in ambient atmosphere. A schematic illustration of the pellet fabrication process is shown in Figure 1, and the sample designations are summarized in Table 1.

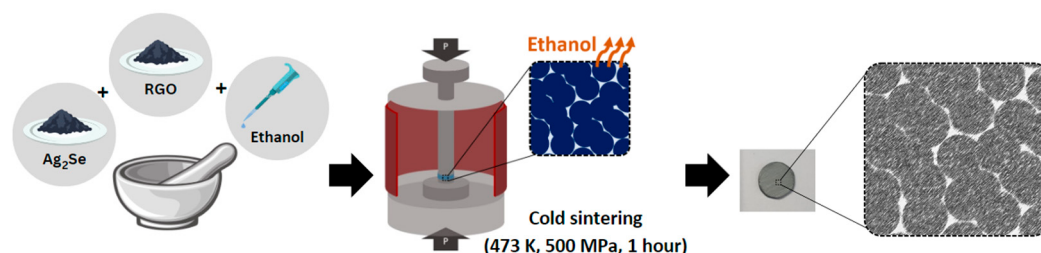


Figure 1. Schematic illustration for the fabrication of Ag_2Se /RGO composites.

Table 1. Sample designation for the Ag₂Se and Ag₂Se/RGO samples.

Sample	RGO wt.%	Phase
Ag ₂ Se-powder	-	Powder
Ag ₂ Se	-	Pellet
Ag ₂ Se-0.25%	0.25	Pellet
Ag ₂ Se-0.5%	0.5	Pellet
Ag ₂ Se-0.75%	0.75	Pellet
Ag ₂ Se-1%	1.0	Pellet

2.3. Characterization

The sample density (d) was determined using the Archimedes principle. Crystal structure analysis was performed via X-ray diffraction (XRD, PANalytical, EMPYREAN, Almelo, Netherlands) using Cu K α radiation ($\lambda = 1.54 \text{ \AA}$). Microstructural features and elemental composition were examined using scanning electron microscopy (SEM, FEI, Helios NanoLab G3 CX, Waltham, MA, USA) coupled with energy dispersive X-ray spectroscopy (EDS). Transmission electron microscopy (TEM, Thermo Scientific, Talos F200X G2, Waltham, MA, USA) was used to investigate detailed nanostructure lattice planes in the samples. Thermoelectric properties including Seebeck coefficient and electrical conductivity were measured using a 4-point probe method (LSR-3, Linseis, Selb, Germany) over the temperature range from room temperature to 383 K under He atmosphere. Thermal diffusivity (α) was measured using a laser flash method (Linseis, LFA-500, Selb, Germany) under Ar atmosphere across the same temperature range. Thermal conductivity (κ) was calculated using the relation: $\kappa = \alpha C_p d$, where C_p is the specific heat capacity, measured by differential scanning calorimetry (DSC, Rigaku, Thermo plus evo2, Tokyo, Japan). All thermoelectric measurements have been repeated three times at each temperature point to ensure reproducibility and accuracy.

3. Results and Discussion

3.1. Physical and Microstructural Analysis

The relative density of the samples decreased progressively with increasing RGO content, as shown in Figure 2a. The pristine Ag₂Se sample exhibited the highest density (98.8%), while the sample containing 1 wt.% RGO (Ag₂Se-1%) showed the lowest density (88.7%). This trend is consistent with previous reports involving the incorporation of graphene-based materials into thermoelectric materials, such as Bi₂Te₃ [16], Cu₂SnSe₃ [42], and CoVSn [43], where increasing graphene content led to a reduction in bulk density. The observed decrease in density can be attributed to the significantly lower intrinsic density of RGO compared to Ag₂Se. Furthermore, RGO flakes may adhere between Ag₂Se grains, impeding particle compaction and introducing voids between grains, as schematically illustrated in Figure 2b. These microstructural disruptions contribute to the overall reduction in sample density and confirm the successful incorporation of RGO into the Ag₂Se matrix.

The crystallographic structure of the samples was examined using XRD, as shown in Figure 3. The diffraction patterns match well with the ICDD reference (00-024-1041), confirming the presence of β -Ag₂Se with an orthorhombic structure and space group $p2_12_12_1$. Rietveld refinement results, summarized in Table S1 yielded lattice parameters of $a = 4.34 \text{ \AA}$, $b = 7.07 \text{ \AA}$, and $c = 7.77 \text{ \AA}$ —characteristic of the orthorhombic β -Ag₂Se phase. These values are consistent with previous reports [44,45]. Importantly, the orthorhombic crystal structure of β -Ag₂Se remained stable upon RGO incorporation, indicating that the compositing process did not significantly alter the crystallographic parameters. However, no distinct diffraction peaks corresponding to RGO were observed in any of the composite

samples. This absence is attributed to the relatively low-crystalline RGO compared to the highly crystalline Ag_2Se matrix, which dominates the diffraction signal and masks the presence of RGO. Therefore, to confirm the successful incorporation of RGO within the Ag_2Se matrix, morphological analysis was employed as a complementary approach.

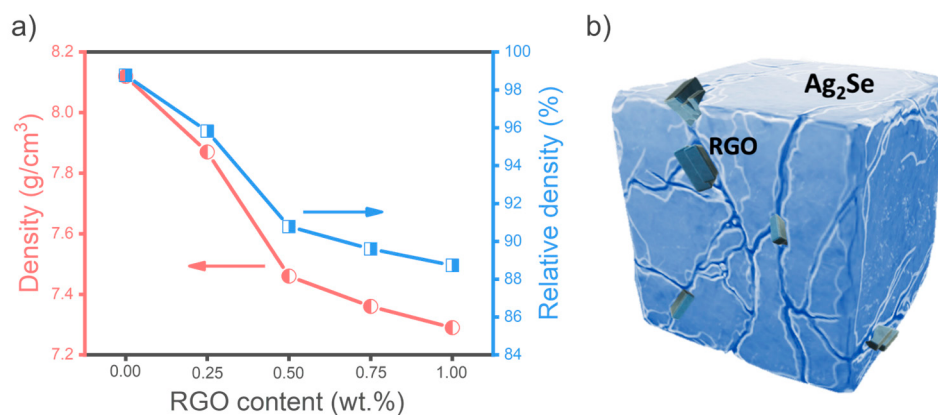


Figure 2. (a) Densities of Ag_2Se /RGO samples with varying RGO contents; (b) Schematic illustration of RGO flakes dispersed in the Ag_2Se -based matrix.

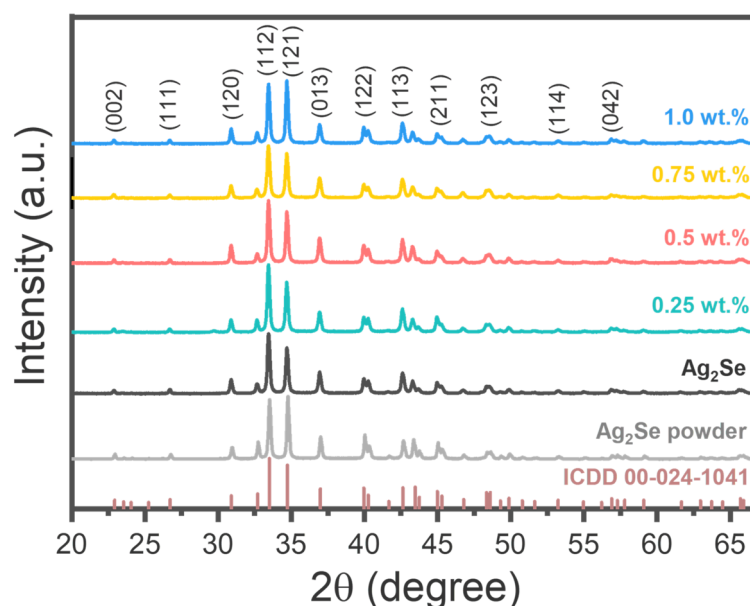


Figure 3. XRD patterns of Ag_2Se samples and Ag_2Se /RGO composites along with the $\beta\text{-Ag}_2\text{Se}$ reference pattern.

SEM analysis was conducted to study the surface morphology of the Ag_2Se -based samples, as shown in Figure 4. In Figure 4a, the pristine Ag_2Se powder exhibits particle sizes ranging from approximately 3 to 10 μm in low-magnification images. Higher-magnification observations reveal that these micro-sized particles are composed of smaller, agglomerated primary particles. In addition, Figure 4b–f display the microstructure of samples consolidated via the CSP with varying RGO contents (0–1.0 wt.%). The pristine sample (Figure 4b) shows a densely packed morphology, consistent with its highest relative density of 98.8%. In contrast, the adherence of RGO to the Ag_2Se matrix is clearly observed in Figure 4c–f. Low-magnification images reveal a progressive increase in RGO dispersion with rising nominal composition, indicating that the CSP effectively facilitates homogeneous integration of RGO within the matrix. At higher magnification, the RGO sheets are observed directly attached to Ag_2Se grains. However, at elevated RGO concentrations (1.0 wt.%), the

presence of RGO appears to disrupt particle packing, leading to microstructural discontinuities and a more fractured matrix (Figure 4f). These morphological changes further support the observed reduction in sample density with increasing RGO content.

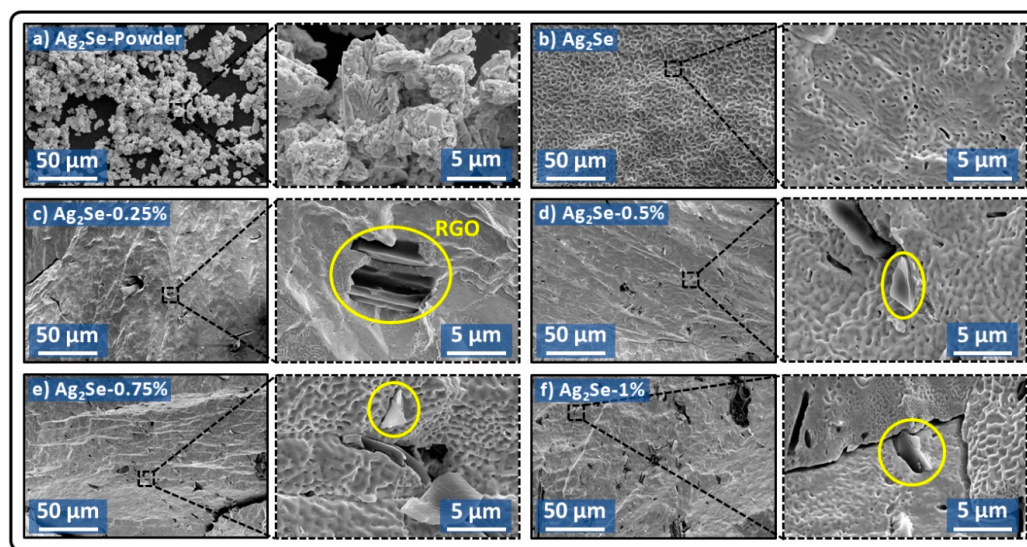


Figure 4. SEM images at low and high magnifications of (a) Ag_2Se powder; (b) pristine Ag_2Se sample, (c–f) Ag_2Se composites with 0.25–1.0 wt.% RGO. The yellow circles highlight the RGO flakes adhered to Ag_2Se grains.

To complement the morphological analysis, EDS was performed on all samples, with the results presented in Figure S1. The atomic ratio of silver to selenium remains close to the stoichiometric 2:1 ratio, confirming the chemical integrity of Ag_2Se . In addition, carbon and oxygen signals were detected, with increasing intensity correlating with higher RGO content. This is consistent with the composition of RGO, which primarily consists of carbon and contains oxygen-bearing functional groups. These findings confirm the successful incorporation of RGO into the Ag_2Se matrix via the CSP.

To investigate the nanostructure of the prepared samples in greater detail, TEM analysis was performed on both pristine and RGO-composited Ag_2Se samples. As shown in Figure 5a, the low-magnification image of the pristine Ag_2Se sample reveals a densely consolidated microstructure, confirming that the CSP is effective in densifying the powders into a compact bulk sample. Furthermore, no residual liquid phase originating from the CSP was observed, indicating that the consolidation was successfully achieved without leaving unwanted secondary phases. The red square highlights an area selected for high-resolution analysis. The selected area electron diffraction (SAED) pattern (top inset) displays sharp diffraction spots of the orthorhombic β - Ag_2Se phase, indicating a highly ordered crystal structure. Fast Fourier transform (FFT) analysis of the high-resolution TEM image (bottom inset) confirms the presence of the (112) and (101) planes, with interplanar spacings of 0.261 nm and 0.375 nm, respectively, consistent with the XRD results. In contrast, Figure 5b presents the TEM image of an RGO-composited Ag_2Se sample. At low magnification, RGO sheets are clearly seen adhering to the surface of Ag_2Se particles, confirming successful compositing via the CSP. Upon closer examination, lattice fringes corresponding to the Ag_2Se phase remain visible, although the presence of RGO introduces localized irregularities in the lattice. The inset (yellow square) identifies the (120) and (102) planes of Ag_2Se with interplanar spacings of 0.282 nm and 0.294 nm, respectively. Meanwhile, the RGO region exhibits partial crystallinity, but with poor orientation and less distinct lattice ordering.

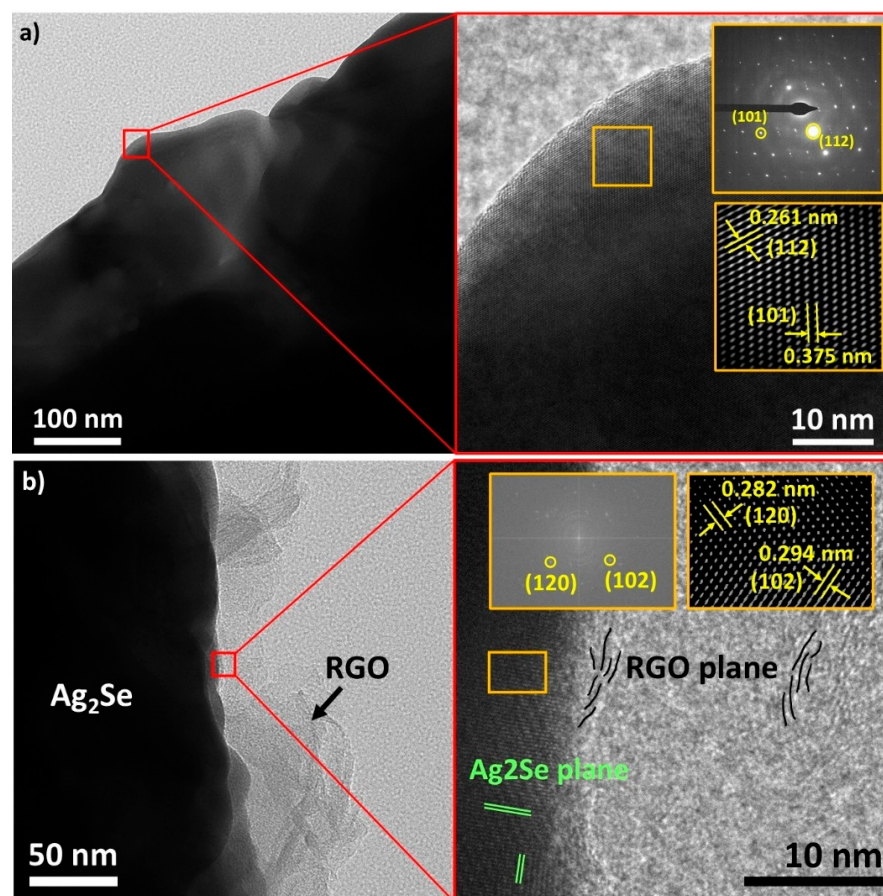


Figure 5. TEM images at low and high magnifications of (a) pristine Ag_2Se sample; (b) $\text{Ag}_2\text{Se}/\text{RGO}$ composites.

3.2. Transport and Thermoelectric Properties

Carrier concentration (n_H) and electron mobility (μ_H) were measured by using the Hall effect technique, as shown in Figure 6a. The pristine Ag_2Se sample exhibited the highest n_H ($\sim 7.68 \times 10^{18} \text{ cm}^{-3}$), which gradually decreased to $7.08 \times 10^{18} \text{ cm}^{-3}$ at 0.25 wt.% RGO. Beyond 0.5 wt.% RGO, n_H further declined and stabilized around $4.50 \times 10^{18} \text{ cm}^{-3}$. On the other hand, μ_H displayed an inverse trend. The pristine sample showed a mobility of $775 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, which increased with RGO content, peaking at $1280 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for the 0.75 wt.% sample. However, at 1.0 wt.% RGO, μ_H dropped sharply to $1001 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

The reduction in n_H is attributed to the presence of oxygen-bearing functional groups on RGO, which act as carrier traps by attracting free electrons—effectively lowering the carrier concentration. This carrier-trapping mechanism has been previously reported in thermoelectric systems containing RGO, for instance, in the Bi-Sb-Te/RGO [46] and SnSe/RGO [47] systems. However, at higher RGO loadings (>0.5 wt.%), the tendency of RGO to agglomerate into larger clusters reduces its interfacial contact with the Ag_2Se matrix, thereby limiting further carrier trapping. Meanwhile, the extended surface area of RGO facilitates efficient charge transport by minimizing grain boundary scattering, which enhances μ_H [48,49]. Nevertheless, the abrupt decline in mobility at 1.0 wt.% RGO is likely due to structural fractures observed in the sample (Figure 4f), which disrupt carrier pathways. This highlights that excessive filler content can compromise structural integrity and hinder charge transport.

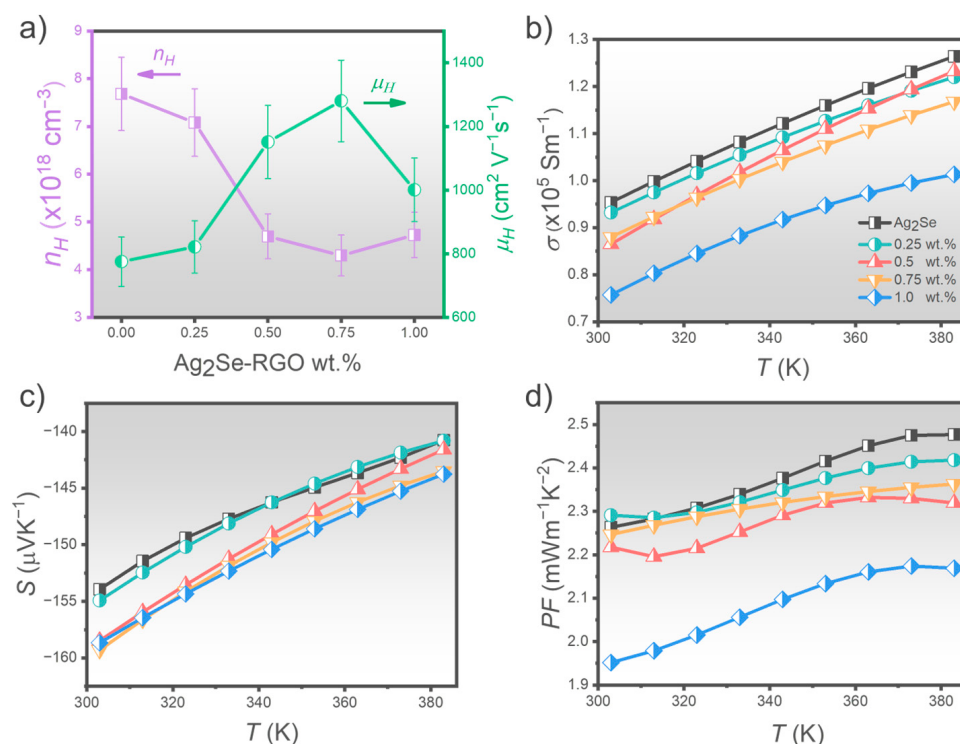


Figure 6. (a) Carrier concentration and mobility of the Ag₂Se-based samples with varying RGO from 0 to 1.0 wt.%, Temperature-dependent; (b) electrical conductivity (σ); (c) Seebeck coefficient; and (d) power factor (PF) of the Ag₂Se-based samples with varying RGO contents.

Figure 6b illustrates the temperature-dependent electrical conductivity (σ) of Ag₂Se samples with varying RGO content. A slight reduction in σ is observed with initial RGO addition, followed by a more pronounced decline at higher concentrations. Specifically, electrical conductivity decreases from $9.5 \times 10^4 \text{ S/m}$ for pristine Ag₂Se to 9.3, 8.6, 8.8, and $7.6 \times 10^4 \text{ S/m}$ for samples containing 0.25, 0.5, 0.75 and 1.0 wt.% RGO, respectively. This trend indicates that the reduction in σ for 0.25–0.75 wt.% RGO samples primarily stems from a decrease in n_H , which is directly proportional to electrical conductivity according to the relation:

$$\sigma = n_H e \mu_H \quad (2)$$

where e is the electron charge ($1.602 \times 10^{-19} \text{ C}$). Although μ_H increases with RGO content, the dominant influence of decreasing n_H results in an overall decline in σ . At 1.0 wt.% RGO, n_H stabilizes relative to 0.5 and 0.75 wt.% samples. Nevertheless, electrical conductivity continues to decrease. This behavior is attributed to the fractured microstructure of the Ag₂Se matrix, as shown in Figure 4f, which disrupts electron transport pathway. The corresponding reduction in μ_H , discussed earlier, further contributes to the significant drop in electrical conductivity to its lowest observed value.

The Seebeck coefficient (S) values are plotted in Figure 6c. The negative sign confirms that Ag₂Se and its composites exhibit n -type thermoelectric behavior. Moreover, the absolute Seebeck coefficient ($|S|$) decreases with increasing temperature, indicating semiconductor-like behavior. For Ag₂Se–0.25%, the Seebeck coefficient remains comparable to the pristine sample ($|S| \sim 154 \mu\text{V K}^{-1}$). However, with RGO content above 0.5 wt.%, $|S|$ increases slightly to $\sim 159 \mu\text{V K}^{-1}$ and then saturates. This enhancement in $|S|$ upon RGO addition can be interpreted using the following relation [3]:

$$S = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left[\frac{\pi}{3n} \right]^{\frac{2}{3}} \quad (3)$$

where k_B , m^* , and h are the Boltzmann constant, the effective mass of charge carriers, and the Planck's constant, respectively. As shown in Figure 6a, n_H decreases with increasing RGO content up to 0.5 wt.%, which reasonably explains the gradual increase in $|S|$ due to its inverse dependence on n_H . Beyond 0.5 wt.%, n_H tends to stabilize, resulting in a corresponding saturation of the Seebeck coefficient.

Figure 6d presents the power factor (PF) of the Ag_2Se samples. Although the Seebeck coefficient improves with increasing RGO content, the enhancement is insufficient to offset the significant reduction in electrical conductivity. Consequently, PF slightly decreases from 2.4 to 2.3 $mW\ m^{-1}\ K^{-2}$ as the RGO content increases from 0.25 to 0.75 wt.% at 383 K. A more pronounced drop in PF is observed for the 1.0 wt.% RGO sample, reaching 2.1 $mW\ m^{-1}\ K^{-2}$. These results indicate that RGO incorporation affects the n_H , leading to reduced electrical conductivity. While the increase in absolute Seebeck coefficient helps maintain a relatively balanced PF in the 0.25–0.75 wt.% range, further RGO addition to 1.0 wt.% adversely impacts the microstructure and charge transport, ultimately compromising the power factor of the composite.

Figure 7a depicts the total thermal conductivity (κ_{total}) of Ag_2Se samples with varying RGO content. The pristine sample exhibits maximum κ_{total} of 0.93 $W\ m^{-1}\ K^{-1}$ at 353 K. With increasing RGO content from 0.25 to 1.0 wt.%, κ_{total} gradually decreases, with minimum values below 0.75 $W\ m^{-1}\ K^{-1}$ at 303 K, indicating that RGO incorporation effectively suppresses thermal transport. This behavior can be interpreted by decomposing κ_{total} into two components: electronic thermal conductivity (κ_e) and lattice thermal conductivity (κ_l), as shown in Figure S2. The electronic contribution is calculated using the Wiedemann–Franz law: $\kappa_e = L\sigma T$, where L is the Lorentz number ($1.77 \times 10^{-8}\ W\Omega\ K^{-2}$) [8]. As electrical conductivity decreases with increasing RGO content, κ_e also declines (Figure S2a). In contrast, κ_l exhibits a more complex trend. It slightly increases with the addition of 0.25 wt.% RGO (Figure S2b), likely due to RGO filling voids between Ag_2Se particles and facilitating phonon transport. However, with further RGO addition (0.5–0.75 wt.%), κ_l decreases, as excess RGO introduces additional interfacial boundaries, enhancing phonon scattering. Interestingly, the sample with 1.0 wt.% RGO shows a rise in κ_l , attributed to RGO agglomeration forming continuous pathways for phonon transport. Given RGO possessing inherently high thermal conductivity, these regions reduce interfacial scattering and elevate κ_l .

Figure 7b presents the temperature-dependent zT . Overall, zT improves with increasing RGO content up to 0.75 wt.%. The 0.25 wt.% RGO sample shows a modest enhancement over pristine Ag_2Se , while the 0.5 and 0.75 wt.% samples exhibit more substantial improvement due to reduced thermal conductivity and relatively stable PF . Notably, the 0.75 wt.% RGO sample maintains a high zT (0.9–1.0) across the entire measured temperature range, demonstrating superior thermoelectric performance. In contrast, the 1.0 wt.% RGO sample shows the lowest zT due to a sharp decline in PF , despite its low thermal conductivity.

Although the 0.25 wt.% sample achieves the highest zT at 383 K, its performance is limited to a narrow temperature window. In contrast, the 0.75 wt.% RGO sample, with an average zT of 0.98 from room temperature to 383 K (Figure 7c), offers the best overall thermoelectric performance across the measured range, making it the optimal composition. Figure 7d compares the average zT values of Ag_2Se -based materials from previous studies, showing that the zT achieved in this work surpasses previously reported values. These results demonstrate that the strategies employed in this study are effective, maintaining a reasonable PF while significantly reducing total thermal conductivity. Ultimately, the combination of the CSP technique and RGO incorporation into Ag_2Se proves to be a promising approach for enhancing thermoelectric performance, offering clear advantages in terms of cost reduction and scalability for practical applications.

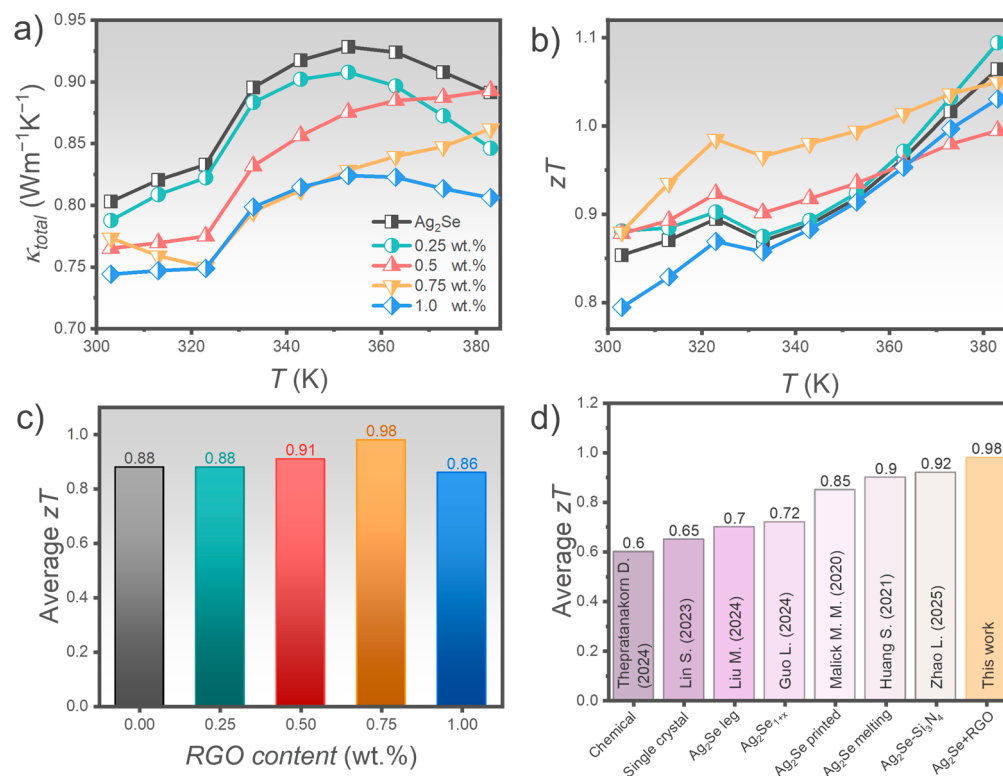


Figure 7. Thermoelectric performance of Ag₂Se samples with varying RGO content: (a) temperature-dependent total thermal conductivity (κ_{total}), (b) temperature-dependent zT values, (c) average zT values for each composition, (d) comparison of average zT with previously reported Ag₂Se-based materials (Data from [9,39,50–54]).

4. Conclusions

This work demonstrates that the CSP is an effective method for fabricating dense Ag₂Se-based thermoelectric composites at low temperature under applied pressure. The incorporation of RGO into the Ag₂Se matrix successfully enhances thermoelectric performance by reducing lattice thermal conductivity and increasing the Seebeck coefficient, which reached up to $-159 \mu\text{V K}^{-1}$ for the 0.75 wt.% RGO sample. Although RGO addition decreases electrical conductivity from $9.5 \times 10^4 \text{ S m}^{-1}$ for pristine Ag₂Se to $7.6 \times 10^4 \text{ S m}^{-1}$ at 1.0 wt.% RGO due to carrier trapping and microstructural disruption, the overall *PF* remains reasonable, ranging from 2.4 to $2.1 \text{ mW m}^{-1} \text{ K}^{-2}$ at 383 K across compositions. The optimal formulation, containing 0.75 wt.% RGO, achieves the highest average zT of 0.98 across the measured temperature range (303–383 K). Compared to previous strategies, this approach offers a practical and cost-efficient alternative for improving *n*-type thermoelectric materials. The synergy between the CSP and RGO incorporation provides a promising route for scalable energy-harvesting applications near room temperature.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/nano15211631/s1>, Figure S1: SEM images with corresponding EDS mapping for (a) Ag₂Se powder, (b) pristine Ag₂Se sample, (c–f) Ag₂Se composites with 0.25–1.0 wt.% RGO; Figure S2: (a) electronic thermal conductivity (κ_e) and (b) lattice thermal conductivity (κ_l) of pristine Ag₂Se and Ag₂Se/RGO composite samples; Table S1: The crystal structural parameters of Ag₂Se and Ag₂Se/RGO composites, determined by Rietveld refinement of the XRD patterns.

Author Contributions: Conceptualization, D.P., I.D., P.P., S.P.; Methodology, D.P., I.D., P.P.; Validation, D.P., S.P.; Formal analysis, D.P., I.D.; Investigation, D.P., I.D.; Resources, S.P.; Data Curation, D.P., I.D.; Writing—Original Draft Preparation, D.P., I.D.; Writing—Review and Editing, S.P.; Visualization, D.P.,

I.D.; Supervision, S.P.; Project administration, S.P.; Funding acquisition, S.P. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author(s).

Conflicts of Interest: The authors declare no conflicts of interest.

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