

## Article

# Surface Modification of Bi<sub>2</sub>Te<sub>3</sub> Nanoplates Deposited with Tin, Palladium, and Tin/Palladium Using Electroless Deposition

Kaito Kohashi <sup>1</sup>, Yutaro Okano <sup>1</sup>, Daiki Tanisawa <sup>1</sup>, Keisuke Kaneko <sup>1</sup>, Shugo Miyake <sup>2</sup> and Masayuki Takashiri <sup>1,\*</sup> 

<sup>1</sup> Department of Materials Science, Tokai University, 4-1-1, Kitakaname, Hiratsuka 259-1292, Japan; 3cajm021@mail.u-tokai.ac.jp (K.K.); 3cajm010@mail.u-tokai.ac.jp (Y.O.); 2cajm022@mail.u-tokai.ac.jp (D.T.); 0cer1115@mail.u-tokai.ac.jp (K.K.)

<sup>2</sup> Department of Mechanical Engineering, Kobe City College of Technology, 8-3, Gakuen-Higashimachi, Nishi-ku, Kobe 651-2194, Japan; miyake@kobe-kosen.ac.jp

\* Correspondence: takashiri@tokai-u.jp

**Abstract:** Surface-modified nanoplate-shaped thermoelectric materials can achieve good thermoelectric performance. Herein, single-crystalline Bi<sub>2</sub>Te<sub>3</sub> nanoplates with regular hexagonal shapes were prepared via solvothermal techniques. Surface modification was performed to deposit different metals onto the nanoplates using electroless deposition. Nanoparticle-shaped tin (Sn) and layer-shaped palladium (Pd) formed on the Bi<sub>2</sub>Te<sub>3</sub> nanoplates via electroless deposition. For the sequential deposition of Sn and Pd, the surface morphology was mostly the same as that of the Sn-Bi<sub>2</sub>Te<sub>3</sub> nanoplates. To assess the thermoelectric properties of the nanoplates as closely as possible, they were compressed into thin bulk shapes at 300 K. The Sn-Bi<sub>2</sub>Te<sub>3</sub> and Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates exhibited the lowest lattice thermal conductivity of 1.1 W/(m·K), indicating that nanoparticle-shaped Sn facilitated the scattering of phonons. By contrast, the Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates exhibited the highest electrical conductivity. Thus, the highest power factor (15 μW/(m·K<sup>2</sup>)) and dimensionless ZT (32 × 10<sup>−3</sup>) were obtained for the Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates. These thermoelectric properties were not as high as those of the sintered Bi<sub>2</sub>Te<sub>3</sub> samples; however, this study revealed the effect of different metal depositions on Bi<sub>2</sub>Te<sub>3</sub> nanoplates for improving thermoelectric performance. These findings offer venues for improving thermoelectric performance by sintering nanoplates deposited with appropriate metals.

**Keywords:** solvothermal technique; electroless deposition; thermoelectrics; nanoplate



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## 1. Introduction

With the development of wireless sensor networks for the Internet of Things (IoT), securing a power supply for sensors has become a major issue [1]. Therefore, energy harvesting has received considerable attention [2]. These methods convert environmental energy, including light [3,4], vibration [5,6], heat [7,8], and electromagnetic waves [9,10], into electrical energy. Among the different types of environmental energy, thermal energy is the most accessible, owing to its ubiquity; electrical energy can be obtained from it using a thermoelectric generation system. The performance of thermoelectric generation depends directly on the properties of the thermoelectric materials. The main thermoelectric properties are the power factor,  $PF$ , and the dimensionless figure of merit,  $ZT$ , which are expressed as  $PF = \sigma S^2$  and  $ZT = \sigma S^2 T / \kappa$ , respectively, where  $\sigma$ ,  $S$ ,  $T$ , and  $\kappa$  are the electrical conductivity, Seebeck coefficient, absolute temperature, and thermal conductivity, respectively.

Theoretical and experimental studies of thermoelectricity began in earnest in the 1950s [11], and various structures of thermoelectric generators have since been proposed [12–16]. Various thermoelectric materials have been considered, including inorganic and organic materials [17–23]. However, despite these efforts, the best-performing materials at temperatures of approximately 300 K have been bismuth-telluride-based alloys developed in the 1950s [24–30]. These alloys have hexagonal unit cells, and the lattice constant along the  $c$ -axis is almost seven times greater than that along the  $a$ - or  $b$ -axis, contributing to

the significant anisotropic thermoelectric properties of the material. These alloys exhibit properties of an indirect-transition-type semiconductor with a band gap ranging from 0.1 to 0.2 eV. The typical single-crystalline  $\text{Bi}_2\text{Te}_3$  bulk exhibited a  $ZT$  of approximately 0.8 [31]. Therefore, when thermoelectric generators are used to power wireless sensors, bismuth-telluride-based alloys are the most practical. However, their material performance is not sufficient to power wireless sensors, requiring further performance improvements. One approach to improving material performance is nanostructuring, which can lower thermal conductivity owing to phonon scattering and high Seebeck coefficient, which are due to lower dimensionality [32]. To this end, various nanostructured bismuth-telluride-based alloys have been developed, including superlattices [33,34], nanocrystals [35–38], nanoparticles [39–41], and nanoplates [42–44]. In superlattices, Venkatasubramanian et al. fabricated the p-type  $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$  superlattices using metal–organic chemical vapor deposition and achieved a  $ZT$  of 2.4 at 300 K [33]. In nanocrystals, Poudel et al. fabricated bismuth antimony telluride bulk alloys made by hot-pressing nanoparticles and achieved a  $ZT$  of 1.4 at 373 K [35]. Wu et al. fabricated nanoporous (001)-oriented  $\text{Bi}_2\text{Te}_3$  nanoplates using a facile process combining chemical and physical methods, including annealing and a reaction-engineered solution method, and the estimated  $ZT$  was larger than 1.2 at 300 K [44]. In particular, nanoplates have excellent geometries that can be both nanostructured and low-dimensional. Because nanoplates are extremely thin (tens of nanometers) compared with their diameter, they can be considered two-dimensional structures. Therefore, the synthesis of bismuth-telluride-based alloy nanoplates and the evaluation of their thermoelectric properties have been actively investigated [45–49]. Nanocomposites of nanoplates with other materials have also been developed [50–54].

Recently, studies have been conducted to further improve the performance of nanoplates. One approach is the surface modification of nanoplates with different materials [55,56]. Zheng et al. fabricated  $\text{Au-Sb}_2\text{Te}_3$  nanoplates with a beneficial interface barrier and high phonon scattering, resulting in synergistically optimal electrical and thermal properties [57]. In our previous study, tin nanoparticles were deposited on the surface of a  $\text{Bi}_2\text{Te}_3$  nanoplate using electroless deposition, which reduced the lattice thermal conductivity [58]. When different materials were deposited on the surfaces of nanoplates, the crystal growth modes differed depending on the lattice matching and interfacial free energy [59]. Therefore, the morphology of the nanoplate surface varies depending on the type of material deposited on its surface, affecting its thermoelectric performance.

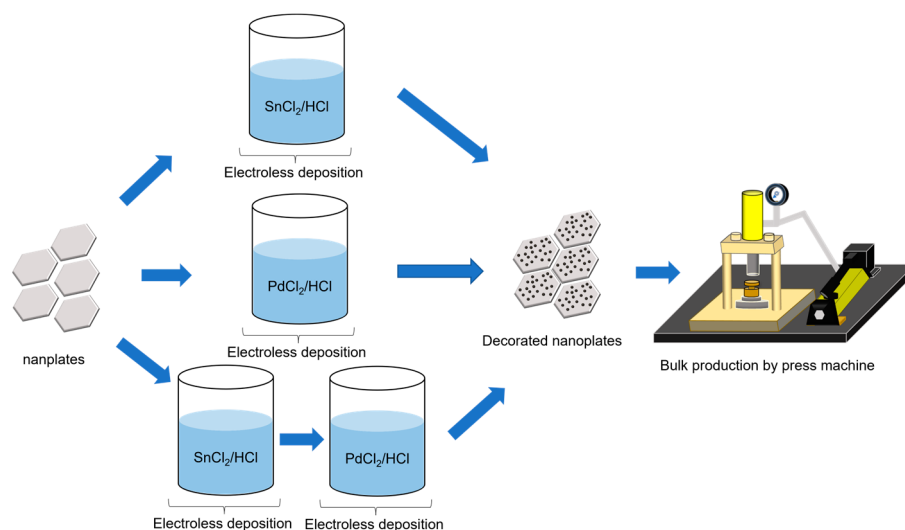
In this study, we prepared  $\text{Bi}_2\text{Te}_3$  nanoplates via solvothermal techniques and deposited different metals (tin (Sn), palladium (Pd), and Sn/Pd) on the nanoplate surface using electroless deposition. Sn and Pd have been typically used in electroless deposition, exhibiting different deposition modes [60]. In addition, the magnitude of diffusion in  $\text{Bi}_2\text{Te}_3$  is different for Sn and Pd. Sn tends to diffuse into  $\text{Bi}_2\text{Te}_3$ , while Pd tends to remain on the  $\text{Bi}_2\text{Te}_3$  surface [61]. Therefore, different  $\text{Bi}_2\text{Te}_3$  surface structures can be fabricated by using Sn and Pd, and the difference can be reflected in the thermoelectric performance. Their structural and thermoelectric properties were examined using various analytical techniques. The surface morphology of the nanoplates depended on the deposited metal. To measure the thermoelectric properties of the nanoplates, the nanoplate assemblies were compressed into thin bulk shapes. The thermoelectric properties of the bulk samples varied depending on the metal type and surface morphology of the nanoplates. Finally, we discuss the different modes of crystal growth on the nanoplates depending on the metal type and their effect on the thermoelectric properties.

## 2. Materials and Methods

The studied  $\text{Bi}_2\text{Te}_3$  nanoplates were fabricated via a solvothermal technique. The detailed fabrication procedure is described in previous studies [62–64]. In brief,  $\text{Bi}_2\text{Te}_3$  nanoplates were fabricated as follows: 0.4 g of polyvinylpyrrolidone (PVP) (99.9% purity, K30, Ms ~40,000, Fujifilm Wako Co., Osaka, Japan) was dissolved in 18 mL of  $\text{C}_2\text{H}_6\text{O}_2$  (99.5% purity, Fujifilm Wako Co., Osaka, Japan). Subsequently, 20 mM of  $\text{Bi}_2\text{O}_3$  (99.9%

purity, Fujifilm Wako Co., Osaka, Japan), 70 mM of  $\text{TeO}_2$  (99.9% purity, Kojundo Chemical Lab., Sakado, Japan), and 2 mL of NaOH solution (0.5 M) (purity > 97.0%, Fujifilm Wako Co., Osaka, Japan) were added. The resultant solution was placed in an autoclave and heated to 200 °C for 4 h under continuous stirring. The precipitated products were collected using centrifugation and washed several times with distilled water and ethanol. The collected products were dried under vacuum at 60 °C for 24 h. The total weight of dried  $\text{Bi}_2\text{Te}_3$  nanoplates was 0.71 g. The production efficiency, which is defined as the total weight of the dried  $\text{Bi}_2\text{Te}_3$  nanoplates in the precursor solution divided by the weight of Bi and Te in the initial  $\text{Bi}_2\text{O}_3$  and  $\text{TeO}_2$  powders, was 54%.

The process of metal deposition on the studied  $\text{Bi}_2\text{Te}_3$  nanoplates is shown in Figure 1. A similar electroless deposition process has been described in our previous study [58]. To deposit the metals on the  $\text{Bi}_2\text{Te}_3$  nanoplates, a displacement deposition method was used for electroless deposition [65]. Three types of metals were deposited on the  $\text{Bi}_2\text{Te}_3$  nanoplates: Sn, Pd, and the lamination of Sn and Pd (Sn/Pd). For the Sn deposition, a precursor solution comprising 60 mM of  $\text{SnCl}_2$  (purity > 97.0%, Fujifilm Wako Co., Osaka, Japan) and 0.66 M of HCl (concentration of 35–37%, Fujifilm Wako Co., Osaka, Japan) was used. For the Pd deposition, a precursor solution comprising 1.1 mM of  $\text{PdCl}_2$  (purity > 99.0%, Fujifilm Wako Co., Osaka, Japan) and 0.66 M of HCl (concentration of 35–37%, Fujifilm Wako Co., Osaka, Japan) was used. The  $\text{Bi}_2\text{Te}_3$  nanoplates (0.65 g) were immersed in each solution for 10 min. For the Sn/Pd deposition, the same conditions were used for the two solutions, as mentioned earlier. First,  $\text{Bi}_2\text{Te}_3$  nanoplates (0.65 g) were immersed in the Sn-containing solution for 10 min. After rinsing with deionized water, the Sn-deposited  $\text{Bi}_2\text{Te}_3$  nanoplates were immersed in the Pd-containing solution for 10 min. Thereafter, the three types of metal-deposited  $\text{Bi}_2\text{Te}_3$  nanoplates were corrected using filter paper and rinsed with deionized water. In the end, the nanoplates were dried under vacuum at 60 °C for 24 h. In order to evaluate the thermoelectric properties of the prepared nanoplates by representing their properties as close as possible to those of pristine nanoplates, but not the melting structure, 0.38 g of nanoplates were pressed at approximately 300 K under uniaxial pressure (0.45 GPa). The samples were 22 mm long, 5 mm wide, and 0.6 mm thick.



**Figure 1.** Schematic of fabrication processes for metal deposition on  $\text{Bi}_2\text{Te}_3$  nanoplates, followed by compression into thin bulk shapes.

The surface morphologies of the prepared nanoplates were examined using field-emission scanning electron microscopy (FE-SEM; S-4800, Hitachi, Tokyo, Japan), transmission electron microscopy (TEM; JEM-ARM200F, JEOL, Tokyo, Japan), and selected-area electron diffraction (SAED). The atomic composition of the nanoplates was determined using an electron probe microanalyzer (EPMA; EPMA-8050G, Shimadzu, Kyoto, Japan). The phase purity and crystal structure of the nanoplates were characterized using X-ray

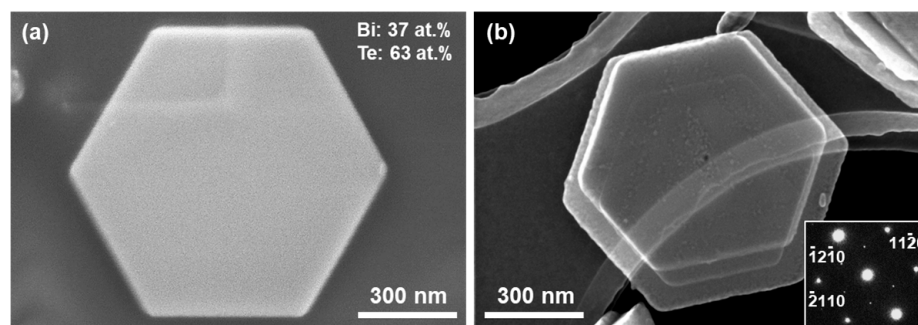
diffraction (XRD; MiniFlex 600, Rigaku, Tokyo, Japan) with Cu-K $\alpha$  radiation ( $\lambda = 1.54 \text{ \AA}$  with  $2\theta$  ranging from  $10^\circ$  to  $80^\circ$ ).

Thermal conductivity  $\kappa$  was calculated using the equation  $\kappa = \alpha\rho C$ , where  $\alpha$ ,  $\rho$ , and  $C$  are thermal diffusivity, density, and specific heat, respectively. The thermal diffusivity  $\alpha$  was measured at 300 K using a thermophysical-property-measuring instrument (Thermowave Analyzer, Bethel, Ishioka, Japan) based on spot-periodic heating radiation thermometry with an accuracy of  $\pm 5\%$  [66]. This is a non-contact method that independently measures both in-plane and cross-plane thermal conductivity. Density was determined by measuring the size and mass. Literature values were used for the specific heat [14,67]. The electrical conductivity and Seebeck coefficient were measured at 300 K using the ZEM-3 method (Advance Riko, Yokohama, Japan) in a helium atmosphere with an accuracy of  $\pm 5\%$ . Finally, the power factor and the dimensionless figure of merit were calculated.

### 3. Results and Discussion

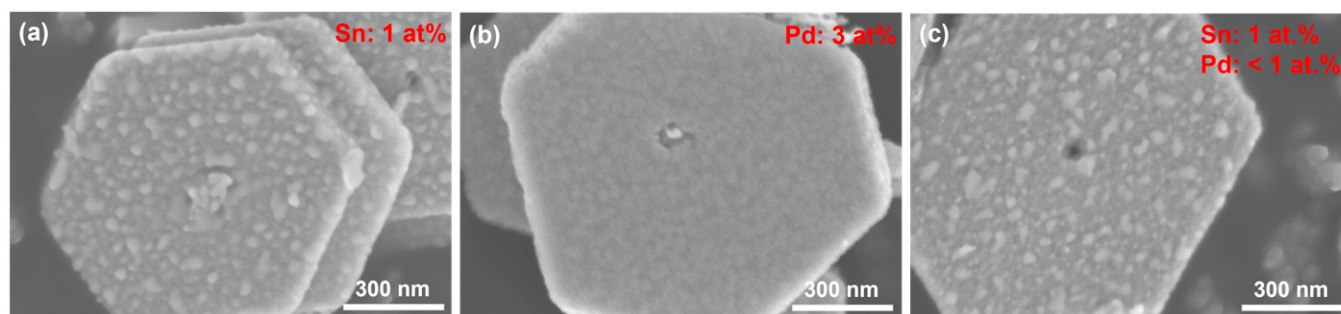
#### 3.1. Structural Characteristics of $\text{Bi}_2\text{Te}_3$ Nanoplates Deposited with Different Metals

Figure 2 shows the structural characteristics of pristine  $\text{Bi}_2\text{Te}_3$  nanoplates. The SEM image in Figure 2a shows that the  $\text{Bi}_2\text{Te}_3$  nanoplates exhibited a significantly flat surface and a regular hexagonal shape with an approximate lateral size of  $1 \text{ }\mu\text{m}$ . The atomic composition of the plates (measured using EPMA) was 37 at% Bi and 63 at% Te, differing by 3% from the stoichiometric ratio (Bi:Te = 40:60). A TEM image of a typical  $\text{Bi}_2\text{Te}_3$  nanoplate is shown in Figure 2b. The nanoplates were considerably thin (less than 50 nm) because of the overlap between the nanoplates and the mesh behind them. The SAED pattern shown in the inset of Figure 2b was indexed to the (00 $l$ ) zone axis of rhombohedral  $\text{Bi}_2\text{Te}_3$ , which indicated that this nanoplate was single-crystalline. In brief, the mechanism of crystal growth of  $\text{Bi}_2\text{Te}_3$  nanoplates is as follows [68]. After the chemical reactions, the  $\text{Bi}_2\text{Te}_3$  nuclei were formed and subsequently aggregated.  $\text{Bi}_2\text{Te}_3$  nanoparticles were generated when the radius of the aggregated nuclei exceeded their critical nucleus size. The nanoplate's formation was attributed to its inherent crystal structure. Due to the high surface energy of the nuclei, the aggregated particles of  $\text{Bi}_2\text{Te}_3$  were not in thermodynamic equilibrium and were metastable. As a result,  $\text{Bi}_2\text{Te}_3$  nanoplates with better crystallinity became thermodynamically preferred. After  $\text{Bi}_2\text{Te}_3$  nanoplates were formed, the Ostwald ripening process took place. The smaller nanoparticles adsorbed around the plates were consumed and grew gradually, leading to larger nanoplates along the top–bottom crystalline plane. This phenomenon happened because the rhombohedral  $\text{Bi}_2\text{Te}_3$  can be described as a stack of infinite layers extending along the top–bottom crystalline plane connected by van der Waals bonds. From a thermodynamic perspective, the free energy of a broken covalent bond is higher than that of a dangling van der Waals bond. This suggests that  $\text{Bi}_2\text{Te}_3$  crystal growth is faster along the top–bottom crystalline plane than along the  $c$ -axis, which is perpendicular to the top–bottom planes. This is because crystalline facets tend to develop on the low-index plane.

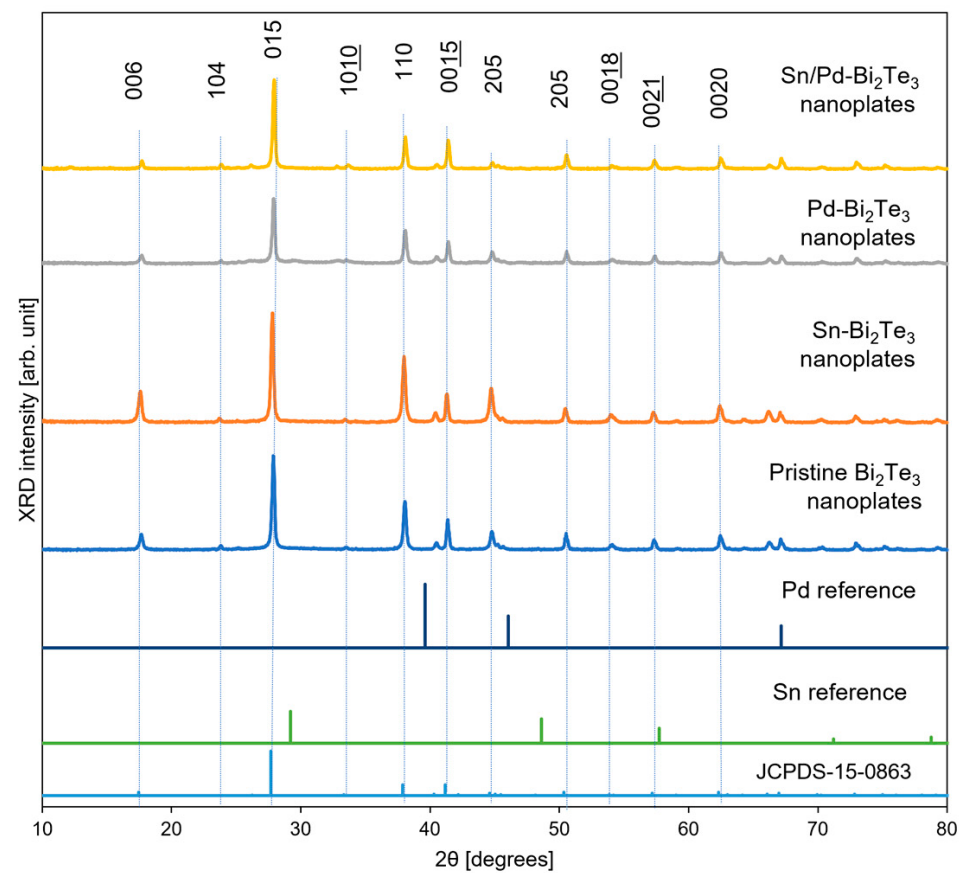


**Figure 2.** (a) SEM image of a  $\text{Bi}_2\text{Te}_3$  nanoplate with atomic composition determined by EPMA and (b) TEM image of a  $\text{Bi}_2\text{Te}_3$  nanoplate and the corresponding SAED pattern.

Figure 3 shows the SEM images of the  $\text{Bi}_2\text{Te}_3$  nanoplates after electroless deposition of different metals. Notably, the SEM image of the Sn- $\text{Bi}_2\text{Te}_3$  nanoplates is referred to from our previous report [58]. As shown in Figure 3a, when Sn was deposited on the  $\text{Bi}_2\text{Te}_3$  nanoplates, nanoparticles with a size of several tens of nanometers were dispersed on their surface. The concentration of Sn, as measured using EPMA, was 1 at%, suggesting that nanoparticle-shaped Sn was deposited on the prepared  $\text{Bi}_2\text{Te}_3$  nanoplates. The surface morphology of the Pd-deposited  $\text{Bi}_2\text{Te}_3$  nanoplates was different from that of the Sn-deposited nanoplates, as shown in Figure 3b. The nanoplate surface was covered with a thin, speckled Pd layer with a Pd concentration of 3 at%. Therefore, the shape and amount of deposition on the prepared  $\text{Bi}_2\text{Te}_3$  nanoplates varied depending on the type of metal. This difference was attributed to the different liquid–solid interfacial free energies of Sn- $\text{Bi}_2\text{Te}_3$  and Pd- $\text{Bi}_2\text{Te}_3$  [59]. When the interfacial free energy was higher (as in the case of Sn- $\text{Bi}_2\text{Te}_3$ ), the crystals grew into island shapes. By contrast, when the interfacial free energy was lower (as in the case of Pd- $\text{Bi}_2\text{Te}_3$ ), the crystals grew in a layered shape. As shown in Figure 3c, when Sn and Pd were sequentially deposited on the nanoplate surface, the surface morphology was similar to that of the Sn-deposited nanoplates. The concentration of Sn was 1 at%, which was the same as for the Sn-deposited nanoplates; however, the concentration of Pd was less than 1 at%, which was lower than for the Pd-deposited nanoplates. These results suggest that Pd readily adhered to the  $\text{Bi}_2\text{Te}_3$  surface but struggled to adhere to the Sn surface. The phase purity and crystal structure of the  $\text{Bi}_2\text{Te}_3$  nanoplates with the electroless deposition of different metals were examined via XRD analysis, as shown in Figure 4.  $\text{Bi}_2\text{Te}_3$  showed rhombohedral tetradymite-type crystal structures with the space group  $D_{3d}^5(R\bar{3}m)$  and hexagonal unit cells. The lattice constant along the  $c$ -axis was 30.3 Å, while that along the  $a$ - or  $b$ -axis was 4.35 Å. The unit cell consists of five covalently bonded monatomic sheets arranged along the  $c$ -axis in the sequence  $-\text{Te}^{(1)}-\text{Bi}-\text{Te}^{(2)}-\text{Bi}-\text{Te}^{(1)}-$ . The superscripts (1) and (2) indicate two distinct chemical states of the anions. The bonds between Te(1) and Bi include both covalent and ionic bonds, while Te(2) and Bi are purely covalently bonded. There is a very weak van der Waals attraction between adjacent Te(1) layers. Most of the peaks observed in the XRD patterns of the nanoplates without metal deposition were indexed to the standard diffraction pattern of  $\text{Bi}_2\text{Te}_3$  (JCPDS 15-0863). The main peaks were observed in the  $c$ -axis-oriented (00 $l$ ), (015), and (10 $\bar{1}0$ ) planes, and several smaller peaks were also observed. Although electroless deposition was performed to deposit different metals on the nanoplate surface, the XRD patterns did not exhibit any peaks corresponding to those attributed to the deposited metals. This was due to the low crystallinity, i.e., extremely small crystallite size or high defect density, of the electrodeposited metals. Similar phenomena have been reported for the electrodeposited layers of bismuth-telluride-based alloys [58,69,70].



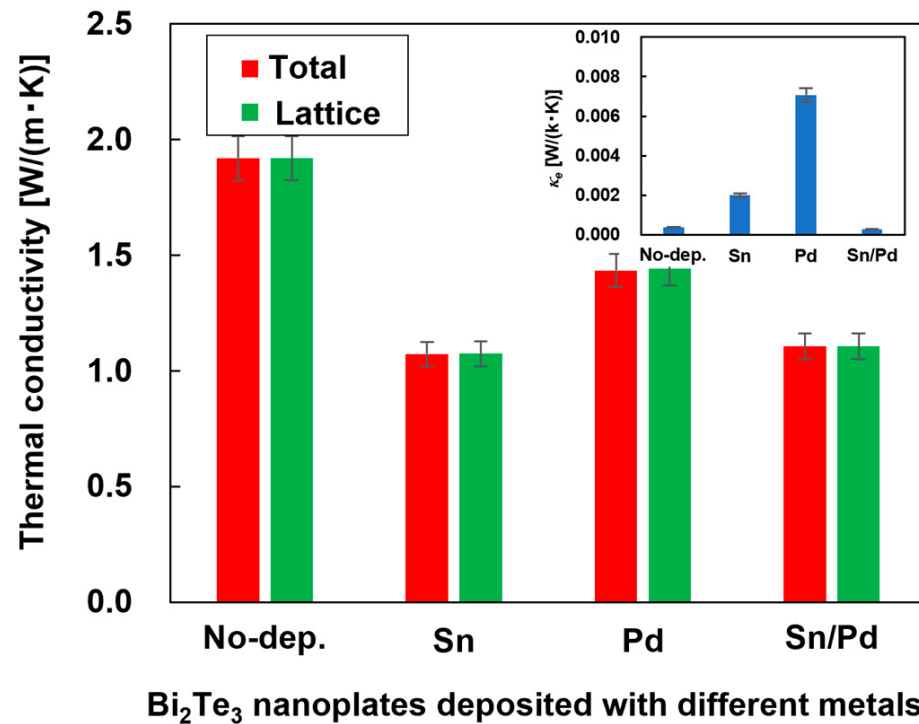
**Figure 3.** SEM images of  $\text{Bi}_2\text{Te}_3$  nanoplates fabricated using electroless deposition with (a) Sn [58], (b) Pd, and (c) Sn/Pd.



**Figure 4.** X-ray diffraction patterns of  $\text{Bi}_2\text{Te}_3$  nanoplates deposited with different metals using electroless deposition.

### 3.2. Thermal Conductivity of $\text{Bi}_2\text{Te}_3$ Nanoplates Deposited with Different Metals

Figure 5 shows the in-plane thermal conductivities of the bulk  $\text{Bi}_2\text{Te}_3$  nanoplates with different metal depositions. The total thermal conductivity,  $\kappa_{\text{total}}$ , was estimated based on the measured thermal diffusivities, densities, and specific heats. The electronic thermal conductivity,  $\kappa_e$ , described in the inset was calculated from the experimentally measured electrical conductivities and using the Wiedemann–Franz law, given by  $\kappa_e = L\sigma T$ . Here, the Lorenz number,  $L$ , was  $2.44 \times 10^{-8} \text{ W}\Omega/\text{K}^2$  for the classical free-electron model. The lattice thermal conductivity,  $\kappa_{\text{lattice}}$ , was obtained by subtracting the electronic thermal conductivity. The lattice thermal conductivities of all nanoplates were mostly the same as the total thermal conductivities, owing to their low electronic thermal conductivities, as shown in the inset. Notably, the thermal conductivities of the pristine  $\text{Bi}_2\text{Te}_3$  nanoplates and the  $\text{Sn-Bi}_2\text{Te}_3$  nanoplates are referred to from our previous report [58]. When electroless deposition was not performed on the  $\text{Bi}_2\text{Te}_3$  nanoplates, the lattice thermal conductivity was  $1.9 \text{ W}/(\text{m}\cdot\text{K})$ , corresponding to the lattice thermal conductivity of single-crystalline  $\text{Bi}_2\text{Te}_3$  in the  $a$ - or  $b$ -axis directions ( $1.3$ – $2.3 \text{ W}/(\text{m}\cdot\text{K})$ ) [71]. Following the metal deposition on the  $\text{Bi}_2\text{Te}_3$  nanoplates, the lattice thermal conductivities decreased. In particular, the  $\text{Sn-Bi}_2\text{Te}_3$  and  $\text{Sn/Pd-Bi}_2\text{Te}_3$  nanoplates exhibited the lowest value of  $1.1 \text{ W}/(\text{m}\cdot\text{K})$ , followed by the  $\text{Sn/Pd-Bi}_2\text{Te}_3$  nanoplates with the value of  $1.4 \text{ W}/(\text{m}\cdot\text{K})$ . This phenomenon indicates that the dispersion of metal nanoparticles on the nanoplate surface caused more phonons to scatter compared with a structure with a metal layer on the nanoplate surface, reducing the lattice thermal conductivity. This phenomenon was consistent with a previous report [72].



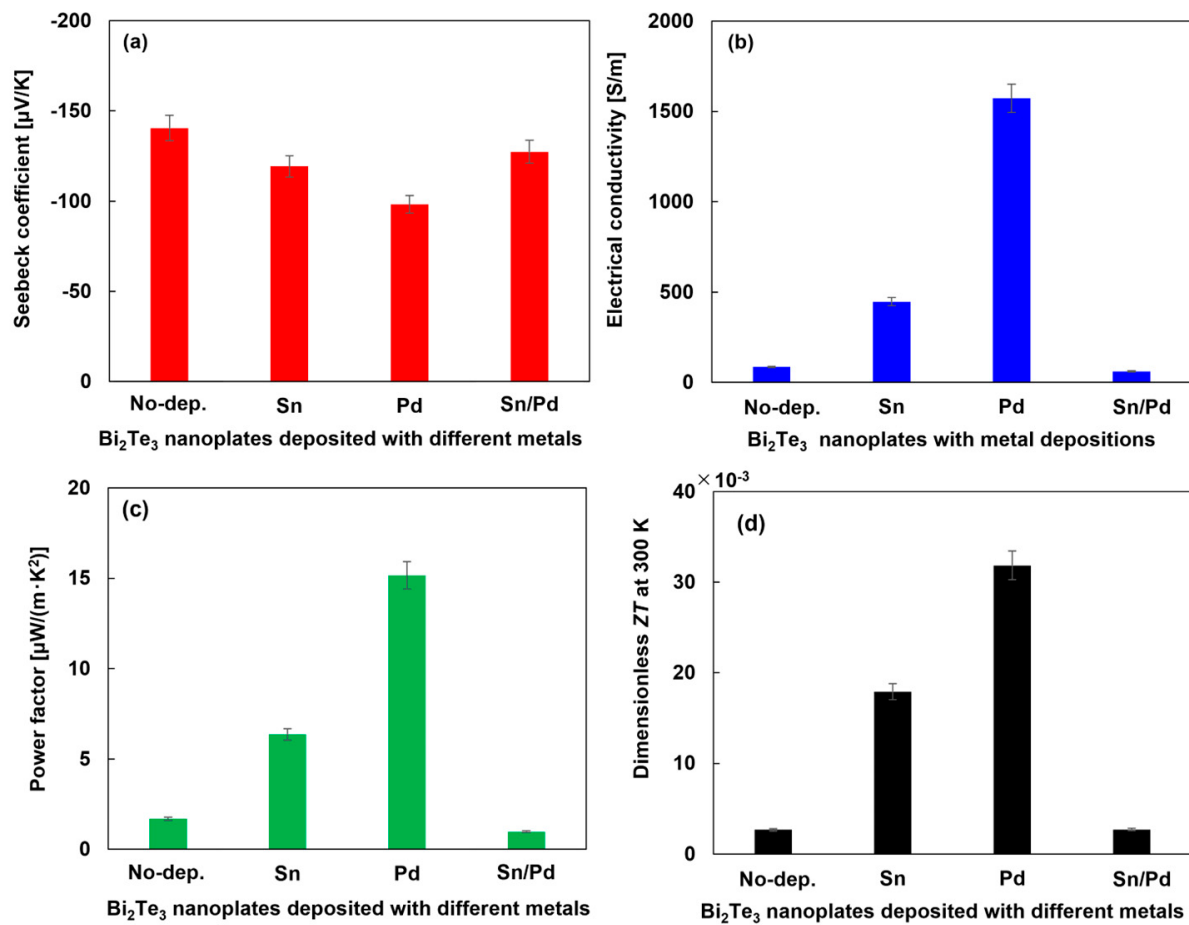
**Figure 5.** In-plane thermal conductivities of bulk-formed Bi<sub>2</sub>Te<sub>3</sub> nanoplates deposited with different metals using electroless deposition measured at 300 K. The inset describes the electronic thermal conductivity of the nanoplates determined using the Wiedemann–Franz law.

### 3.3. Thermoelectric Properties of Bi<sub>2</sub>Te<sub>3</sub> Nanoplates Deposited with Different Metals

Figure 6 shows the in-plane thermoelectric properties of bulk-formed Bi<sub>2</sub>Te<sub>3</sub> nanoplates deposited with different metals. In Figure 6a, the most negative Seebeck coefficient of  $-141 \mu\text{V/K}$  was observed in the Bi<sub>2</sub>Te<sub>3</sub> nanoplates without metal deposition. The deposition of Sn and Pd on the Bi<sub>2</sub>Te<sub>3</sub> nanoplates, respectively, decreased the negative Seebeck coefficient, which was strongly related to the carrier concentration. The relationship between the Seebeck coefficient  $S$  and carrier concentration  $n$  is given by Equation (1) [73,74], as follows:

$$S = \frac{8\pi^2 k_B^2 T}{3eh^2} m^* \left( \frac{\pi}{3n} \right)^{\frac{2}{3}}, \quad (1)$$

where  $m^*$  denotes effective mass. According to the above equation, the Seebeck coefficient scales inversely with the carrier concentration. Therefore, the negative Seebeck coefficient decreased because the carrier concentration corresponding to the metal concentration increased, owing to the metal deposition. In particular, the Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates exhibited the lowest negative Seebeck coefficient of  $-98 \mu\text{V/K}$ , owing to the high carrier concentration as determined by EPMA. By contrast, the Seebeck coefficient of the Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates negatively increased to  $-127 \mu\text{V/K}$ , corresponding to the value located between that for the Bi<sub>2</sub>Te<sub>3</sub> nanoplates without metal deposition and that for the plates with Sn deposition. This was because the Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates had higher carrier concentrations compared to that of the Bi<sub>2</sub>Te<sub>3</sub> nanoplates without metal deposition and lower carrier concentrations compared to that of Sn-Bi<sub>2</sub>Te<sub>3</sub> and Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates. The mechanism for the lower carrier concentration when Sn and Pd were deposited simultaneously compared to when each was deposited alone is unclear. However, a possible explanation is that the carriers generated by the deposition of Sn and Pd could be offset by the coexistence of these two metals [75].



**Figure 6.** In-plane thermoelectric properties of bulk-formed Bi<sub>2</sub>Te<sub>3</sub> nanoplates deposited with different metals using electroless deposition measured at 300 K. (a) Seebeck coefficient, (b) electrical conductivity, (c) power factor, and (d) dimensionless figure of merit.

In Figure 6b, the electrical conductivity of the Bi<sub>2</sub>Te<sub>3</sub> nanoplates without metal deposition is shown to be 86 S/m. The highest electrical conductivity of 1572 S/m was observed for the Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates, followed by 447 S/m for the Sn-Bi<sub>2</sub>Te<sub>3</sub> nanoplates, owing to the increase in the carrier concentration compared to the Bi<sub>2</sub>Te<sub>3</sub> nanoplates without metal deposition. By contrast, the Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates exhibited the lowest electrical conductivity of 60 S/m, which was lower than that of the pristine Bi<sub>2</sub>Te<sub>3</sub> nanoplates. This phenomenon cannot be explained by the magnitude of the carrier concentration alone. Electrical conductivity  $\sigma$  is strongly related to the carrier concentration  $n$  as well as mobility  $\mu$ , as expressed by Equation (2):

$$\sigma = ne\mu. \quad (2)$$

Therefore, the Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates had a higher carrier concentration but lower mobility than the pristine Bi<sub>2</sub>Te<sub>3</sub> nanoplates. The low mobility of Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates is due to electron scattering at the interfaces between the Sn and Pd layers.

The power factor of the Bi<sub>2</sub>Te<sub>3</sub> nanoplates with different metal depositions is shown in Figure 6c. The power factor showed a trend similar to that of electrical conductivity because the inter-sample variation of the electrical conductivity was greater than the inter-sample variation of the Seebeck coefficient. The power factors of the Sn-Bi<sub>2</sub>Te<sub>3</sub> and Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates were 6.4 and 15.2  $\mu\text{W}/(\text{m}\cdot\text{K}^2)$ , respectively, which are higher than that of the pristine Bi<sub>2</sub>Te<sub>3</sub> nanoplates at 1.7  $\mu\text{W}/(\text{m}\cdot\text{K}^2)$ . Therefore, the power factor was improved by single-metal deposition on the nanoplatelets, and the power factor depended on the type of metals. However, the Sn/Pd-Bi<sub>2</sub>Te<sub>3</sub> nanoplates exhibited the lowest power factor of 1.0  $\mu\text{W}/(\text{m}\cdot\text{K}^2)$ , which was approximately 60% lower than that of pristine Bi<sub>2</sub>Te<sub>3</sub>.

nanoplates, owing to the lowest electrical conductivity. Therefore, in this study, the simultaneous metal deposition did not enhance the power factor of the nanoplates.

The dimensionless  $ZT$  of the  $\text{Bi}_2\text{Te}_3$  nanoplates with different metal depositions is shown in Figure 6d. The Pd- $\text{Bi}_2\text{Te}_3$  nanoplates exhibited the highest  $ZT$  of  $32 \times 10^{-3}$ . This is because they had extremely high electrical conductivity among the four samples even though they did not have the lowest thermal conductivity or the highest negative Seebeck coefficient. The second highest  $ZT$  of  $18 \times 10^{-3}$  was exhibited by the Sn- $\text{Bi}_2\text{Te}_3$  nanoplates. Due to the lowest thermal conductivity of the Sn- $\text{Bi}_2\text{Te}_3$  nanoplates, the difference in  $ZT$  between the Pb- $\text{Bi}_2\text{Te}_3$  and Sn- $\text{Bi}_2\text{Te}_3$  nanoplates decreased compared to the difference in power factor between them. The  $ZT$  of the Sn/Pd- $\text{Bi}_2\text{Te}_3$  nanoplates was  $2.7 \times 10^{-3}$ , which was the same value as the pristine  $\text{Bi}_2\text{Te}_3$  nanoplates owing to the low thermal conductivity of the Sn/Pd- $\text{Bi}_2\text{Te}_3$  nanoplates. Therefore, all cases of the metal deposition on the  $\text{Bi}_2\text{Te}_3$  nanoplates did not decrease the  $ZT$ . However, the highest  $ZT$  in this study was lower than that for the sintered  $\text{Bi}_2\text{Te}_3$  bulk samples ( $ZT = 0.8$  at 300 K) [76]. This was because the samples in this study underwent no sintering (e.g., spark plasma sintering) to assess the thermoelectric properties of the nanoplates by representing their properties as close as possible to those of the pristine nanoplates without melting. This study reveals the effects of different metal depositions on  $\text{Bi}_2\text{Te}_3$  nanoplates, with implications regarding their thermoelectric performance. In addition, we consider there is room to improve thermoelectric performance by optimizing the type of metal and the amount of metal deposited. In the future, thermoelectric performance will be further improved by sintering nanoplates deposited with appropriate metals.

#### 4. Conclusions

The studied  $\text{Bi}_2\text{Te}_3$  nanoplates were deposited with Sn, Pd, and Sn/Pd, to modify their surface morphologies and improve their thermoelectric properties. The studied  $\text{Bi}_2\text{Te}_3$  nanoplates were prepared using solvothermal techniques and metal deposition was performed using electroless deposition. Sn nanoparticles at a concentration of approximately 1 at% were deposited on the prepared  $\text{Bi}_2\text{Te}_3$  nanoplates. For Pd deposition, a Pd layer with a concentration of approximately 3 at% was deposited on the  $\text{Bi}_2\text{Te}_3$  nanoplates. When Sn and Pd were sequentially deposited on the nanoplate surface, the surface morphology was similar to that of the Sn-deposited nanoplates where the Sn and Pd concentrations were 1 at% and less than 1 at%, respectively. In order to assess the thermoelectric properties of the nanoplates by representing their properties as close as possible to those of the inherent nanoplate but not the melting structures, the nanoplate assemblies were compressed into thin bulk shapes at 300 K. The lattice thermal conductivity was affected by the surface morphology of the nanoplates. The Sn- $\text{Bi}_2\text{Te}_3$  and Sn/Pd- $\text{Bi}_2\text{Te}_3$  nanoplates exhibited the lowest value of  $1.1 \text{ W}/(\text{m}\cdot\text{K})$ , indicating that the dispersion of metal nanoparticles on the nanoplate surface caused more phonon scattering. However, the Pd- $\text{Bi}_2\text{Te}_3$  nanoplates exhibited the highest power factor and dimensionless  $ZT$  owing to their significantly high electrical conductivities.

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