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Thermal and electrical conductivities of Cd–Zn alloys

B Saatçi[1], **M Ari, M Gündüz, F Meydaneri, M Bozoklu and S Durmuş**

Department of Physics, Faculty Arts and Sciences, Erciyes University, Talas Street, Kayseri, Turkey

E-mail: bayender@erciyes.edu.tr

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Abstract

The composition and temperature dependences of the thermal and electrical conductivities of three different Cd–Zn alloys have been investigated in the temperature range of 300–650 K. Thermal conductivities of the Cd–Zn alloys have been determined by using the radial heat flow method. It has been found that the thermal conductivity decreases slightly with increasing temperature and the data of thermal conductivity are shifting together to the higher values with increasing Cd composition. In addition, the electrical measurements were determined by using a standard DC four-point probe technique. The resistivity increases linearly and the electrical conductivity decreases exponentially with increasing temperature. The resistivity and electrical conductivity are independent of composition of Cd and Zn. Also, the temperature coefficient of Cd–Zn alloys has been determined, which is independent of composition of Cd and Zn. Finally, Lorenz number has been calculated using the thermal and electrical conductivity values at 373 and 533 K. The results satisfy the Wiedemann–Franz (WF) relation at $T < 373$ K, which suggests the dominant carriers of thermal conduction are mainly electrons. Above this temperature ($T > 373$ K), the WF relation could not hold and the phonon component contribution of thermal conductivity dominates the thermal conduction.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Alloying elements such as Cd and Zn group metals are usually employed for many applications, such as surface corrosion-protective coating and planting process materials. Cd–Zn alloys are usually used for soldering processes, since solders may contain Cd because of its low melting point. These alloying elements are also used for brazing processes; for higher temperatures we should use alloys containing large amounts of zinc. Cd and Zn also play an important role as alloying elements in many semiconductor and superconductor materials. Therefore, formation

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[1] Author to whom any correspondence should be addressed.

of this kind of alloys is very important, from an academic point of view and for technological applications.

The thermal and electrical conductivities are valuable tools for the study of transport mechanisms of alloys. The thermal and electrical conductivity values are very useful to clarify the properties of such alloys. The thermal conductivity, K, is one of the main fundamental properties of materials. Many attempts have been made to determine the thermal conductivity values of solid and liquid phases in various materials by using different methods [1]. One of the common techniques for measuring the thermal conductivity of solids is the radial heat flow method and it is based upon specimen geometry: i.e. cylindrical or spherical. This method was first used for measuring the thermal conductivity of solids for pure materials by Callendar and Nicolson [2], then this method was developed by Powell [3] and used by Niven [4] for measurements of K on woods and sands. The radial heat flow method was used by Angel [5] for measurements of thermal conductivity on nickel and aluminium. Also, a review of radial heat flow methods was presented by McElroy and Moore [6].

The electrical conductivity may be obtained to give better insight into the electrical transport of Cd–Zn alloys. The four-point probe method has proven to be a convenient tool for the measurement of electrical conductivities. The electrical conductivity measurements can be determined by a standard four-point probe measurement technique, which is one of the best methods to obtain electrical conductivity. A detailed description of the method has been given by Smits [7], who gives the functional relationship between the resistivity, ρ, voltage and current reading for different sample geometries.

Although the thermal and electrical conductivities of pure Cd and pure Zn materials have been obtained theoretically and experimentally, there is not enough information and data available about the thermal and electrical conductivities of Cd–Zn alloys.

The purpose of this study is to investigate the thermal conductivity, electrical conductivity and resistivity of Cd (Cd–5 at.% Zn) solid solution, Zn (Zn–1.3 at.% Cd) solid solution and eutectic phase (Cd–26.5 at.% Zn) for different temperatures. The phase diagram of Cd–Zn is given in figure 1 [8]. Firstly, the temperature and composition dependence of thermal conductivity are investigated. Secondly, resistivity and electrical conductivity were determined at different temperatures and compositions of the Cd–Zn alloys. Then, the Lorenz number was calculated and carrier types of thermal conductivity were determined. Finally, the temperature coefficient of the Cd–Zn alloys has been determined.

2. Experimental apparatus

Gündüz and Hunt [9, 10] designed a radial heat flow apparatus to measure the thermal conductivity of solid materials. Maraşlí and Hunt improved the experimental apparatus for higher temperature [11, 12]. A schematic drawing of the apparatus is shown in figure 2 and the details of the apparatus are given in [9–13].

The crucible consisted of three parts, a cylindrical bore (30 mm OD, 25 mm ID and 170 mm in length), and the top and bottom lids which were tightly fitted to the cylindrical part. There were usually three stationary thermocouples (inserted in 1.2 mm OD, 0.8 mm ID, alumina tube). Two thermocouples were placed 1.0–1.5 mm away from the central alumina tube; one of them was used for a control unit. The other one was used for measuring temperature together with the third stationary thermocouple which was placed about 12 mm away from the central alumina tube (for details see [9–13]). Another thermocouple which was moveable (inserted in 2 mm OD, 1 mm ID, alumina tube) was also placed 10 mm away from the centre and used for measuring the vertical temperature variation. The central heating element was a single Kanthal A_1 wire (typically 1.8 mm diameter and about 180 mm in length) placed inside a thin walled

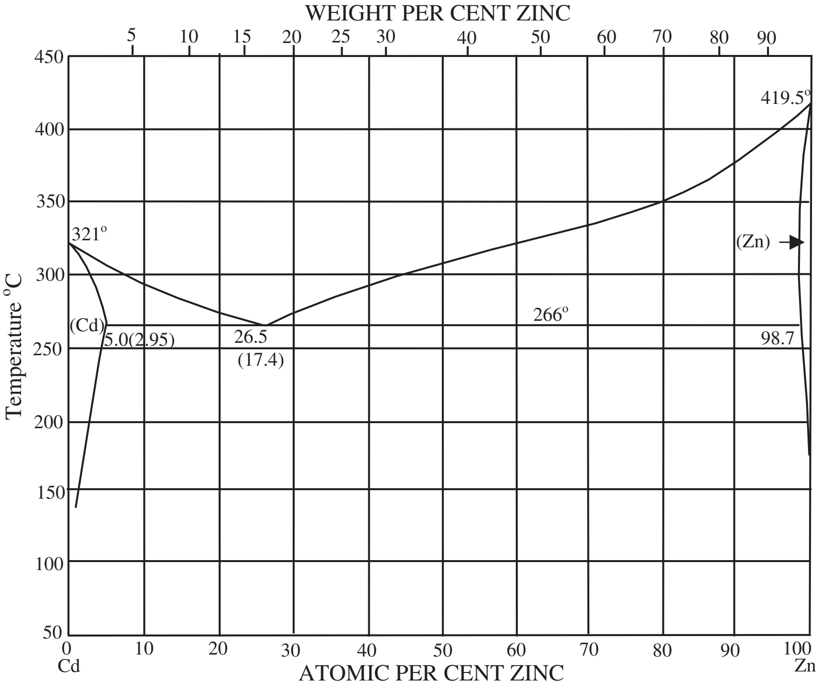


Figure 1. Cd–Zn phase diagram after reference [8].

alumina tube (3 mm OD, 2 mm ID). The end of the wire was threaded and screwed into 7 mm copper rods. The outside of the cylindrical crucible (sample) was kept cool with the water cooling jacket. In the apparatus, the water cooling jacket was very close to the outside of the specimen to obtain effective cooling. The jacket was placed in an alumina tube (100 mm OD, 85 mm ID and 600 mm in length) which has Kanthal A_1 resistance wire on it to give a hot zone of 300 mm. The potential difference between the ends of the central heating element and the known resistance was checked with Keithley 2000 and Hewlett Packard 34401 multimeters. Cd solid solution, eutectic phase and Zn solid solution were prepared in a vacuum melting furnace from pure 4N Cd and Zn. Firstly, sufficient pure materials were cleaned chemically, dried and then melted in the vacuum furnace. After several stirrings, the molten alloy was poured into the graphite crucible held in a hot filling furnace which was set at approximately 50 K above the eutectic melting temperature of the alloy. The alloy was then directionally solidified from bottom to top to ensure that the crucible was completely filled. The sample was then taken out of the hot filling furnace, all thermocouples and the inner heating element were placed in the sample, and the sample was inserted in the cooling jacket then the water cooling jacket was placed in the radial heat flow apparatus. The apparatus is capable of holding the sample to within ±0.05 K temperature range for a day and ±0.1 K temperature range for up to a week.

3. Thermal conductivity

The radial heat flow method has some unique theoretical and practical advantages that, through careful experimental work, can yield reliable results over a wide range of the thermal conductivity of solid phase K_S and temperature ranges. Firstly, a homogeneous sample must be obtained in order to measure the K_S value in alloys and then an experimental system providing radial heat flow must be used.

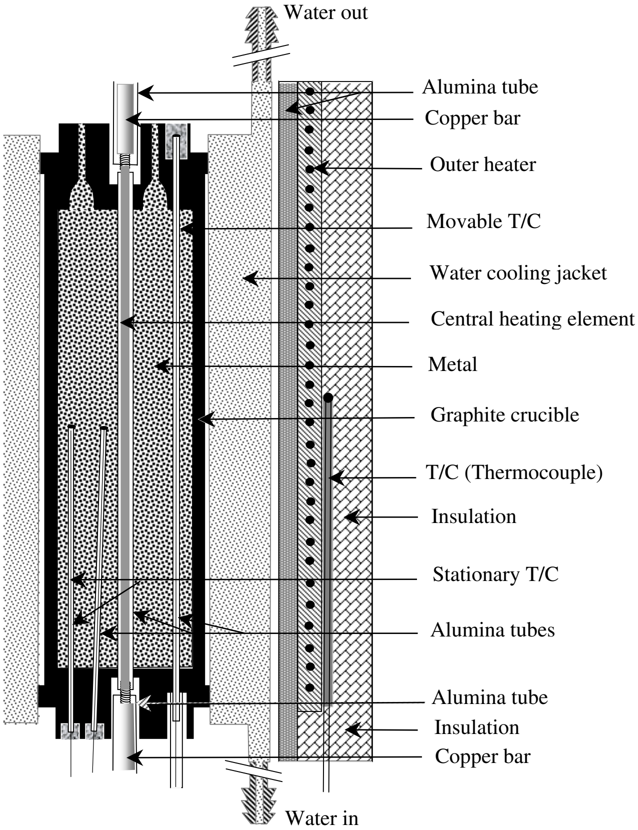


Figure 2. Schematic illustration of the apparatus [9–13].

For the radial heat flow the temperature gradients are given as

$$\left(\frac{\mathrm{d}T}{\mathrm{d}r}\right)_s = -\frac{Q}{2\pi r \ell K_s}. \tag{1}$$

The thermal conductivity of solid phase can be obtained by using an appropriate boundary condition with Fourier's law. Integration of equation (1) for a radial heat flow gives

$$K_\mathrm{S} = \frac{1}{2\pi \ell} \ln\left(\frac{r_2}{r_1}\right) \frac{Q}{T_1 - T_2}. \tag{2}$$

Q is the total input power, ℓ is the length of the heating element, K_S is the thermal conductivity of the solid phase, r_1 and r_2 are fixed distances from the centre of the sample ($r_2 > r_1$) and T_1 and T_2 are temperatures at the fixed positions r_1 and r_2 respectively. If Q, r_1, r_2, ℓ, T_1 and T_2 can be accurately measured for the well characterized sample then reliable K_s values can be evaluated. Equation (2) could be used to calculate the thermal conductivity of solid phase by measuring the difference in the temperatures between the fixed two points for a given power level provided that the vertical temperature variation is minimum or zero.

The Cd–Zn specimens were heated from the centre using a single heating wire in steps of 293–533 K (6 K below T_E). Firstly macroscopically parallel isotherms to the axial centre of the sample were obtained for desired temperature by moving the central heater up and down. The temperature of the specimen was controlled with a Euroterm type 9706 and it was stable

Table 1. Thermal conductivity, electrical conductivity, resistivity, Lorenz number and temperature coefficient of the Cd–Zn alloys at 373 and 533 K.

	Thermal conductivity K ($\mathrm{W\,m^{-1}\,K^{-1}}$)		Electrical conductivity σ ($10^{+6}\,\Omega^{-1}\,\mathrm{m}^{-1}$)		Resistivity ρ ($10^{-7}\,\Omega\,\mathrm{m}$)		Lorenz number L ($10^{-8}\,\mathrm{W}\Omega\,\mathrm{K}^{-2}$)		Temperature coefficient α ($\times 10^{-3}\,\mathrm{K}^{-1}$)
Samples/T	373 K	533 K	373 K	533 K	373 K	533 K	373 K	533 K	373–533 K
Cd–5 at.% Zn	96.62	85.28	8.73	2.65	1.15	3.77	2.97	6.04	−0.73
Cd–26.5 at.% Zn	91.00	81.53	8.86	1.94	1.13	5.15	3.6	6.14	−0.65
Zn–1.3 at.% Cd	86.20	76.94	6.34	2.40	1.58	4.17	2.7	7.73	−0.67

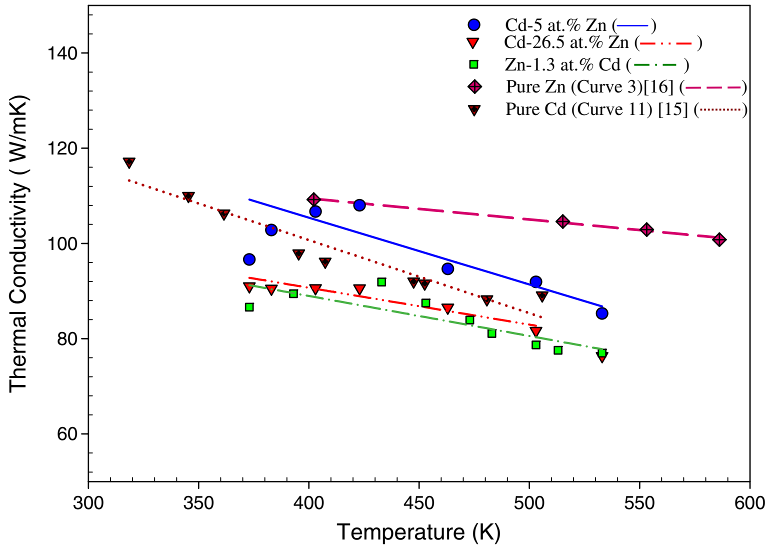


Figure 3. Variation of the thermal conductivities of the pure Cd [15], pure Zn [16] and Cd–Zn alloys with temperature.

to ±0.1 K and the samples were kept at steady state condition for at least 2 h; then total input power Q and T_1 and T_2 temperatures were measured at this condition with a multimeter and recorded with a Pico data-logger. When all the temperatures and Q measurements were completed the specimen was left to cool to room temperature then the sample was moved from the furnace and cut transversely near to the measurement points, after that the specimen was ground and polished for r_1 and r_2 measurements. The distance was measured with an Olympus HP2 optical microscope to an accuracy of ±0.01 mm. The transverse and the longitudinal sections of the sample were examined for porosity, cracks and casting defects to make sure that these would not introduce any errors to the measurements. The values of thermal conductivities are given in table 1 and shown in figure 3 for the Cd–Zn alloys. In this study, the experimental error of thermal conductivity measurements is about 5% [14].

4. Electrical conductivity

The four-point probe method is the most widely used technique for electrical profile measurement of materials. Two of the probes are used to source current and the other two probes are used to measure voltage; using four probes eliminates measurement errors due to the probe resistance, the spreading resistance under each probe, and the contact resistance between each metal probe and the material [7].

The electrical measurements of the samples were obtained by the DC four-point probe technique on circular-shape samples with 12.5 mm diameter. A Keithley 2400 Sourcemeter was used to provide constant current and the potential drop was measured by a Keithley 2700 multimeter through an interface card, which are controlled by a computer. Platinum wires with diameter of 0.5 mm were used as current and potential probes. The voltage drop was detected and the electrical resistivity and conductivity were determined using a standard conversion method. The measurements have been made in the temperature range 300–650 K. The temperature of the sample has been changed by a controllable Nabertherm type P 320 heater and the temperature of the sample was measured using a standard K type thermocouple which was placed near the samples. Some typical values of the electrical conductivities are also given in table 1 for Cd–Zn alloys.

5. Results and discussion

The microstructures of the samples were characterized using an LEO scanning electron microscope (SEM) equipped with energy dispersive x-ray (EDX) and a wavelength dispersive x-ray (WDX) spectrometer as well as a computer controlled image analyser. Figure 4 shows the SEM images of the samples which have no impurities, porosities or structural defects. The morphology and the grain structure of the samples were similar. The characteristics of the surface were also similar in the samples of higher Cd composition, but the sample which has higher Zn composition shows different surface characteristics. After the electrical measurements, the chemical compositions of the samples were determined by using the EDX spectrometer with possible error up to 1% and the results are shown in figure 5. According to the results of the EDX analysis, the peaks of Cd and Zn were clearly seen and percentage element compositions are similar to the compositions before the measurements.

The thermal conductivity of the alloys at T_E is obtained by extrapolating the thermal conductivity–temperature curves to the melting temperature of the alloys. The obtained values of thermal conductivities are given in figure 3, which shows that thermal conductivity decreases with temperature. Figure 3 also shows thermal conductivity results of pure Cd [15] and pure Zn [16]. The curves of the results show clear similarities. The composition dependence of thermal conductivity can also be seen in figure 3. It shows that the thermal conductivity increases with increasing Cd composition. The values of thermal conductivity slightly shifting together to the higher values with increasing Cd composition. The experimental results have been compared with the results of Touloukian *et al* [15, 16] and Saatçi *et al* [17]. Our experimental results are slightly lower than the pure Cd and pure Zn results; this might be due to the alloying effect. However, a reasonably good agreement has been obtained between the results.

Figure 6 shows the temperature dependence of resistivity of the Cd–Zn alloys in the temperature ranges of $T = 300$–650 K. The patterns of resistivity–temperature curves are obtained for three different compositions of Cd–Zn alloys. The electrical resistivities of samples are in the range of 5.98×10^{-8}–6.67×10^{-7} Ω m. From figure 6, it can be clearly seen that the resistivity linearly increases with the increasing temperature. The patterns of resistivity–temperature curves of all samples show similar characteristics. Hence, the Cd–Zn alloys with different compositions exhibit the same resistivity feature with increasing temperature. From the resistivity–temperature curves, we find that the resistivity of Cd–Zn alloys is slightly dependent on composition of Cd and Zn. Also, the temperature dependence of

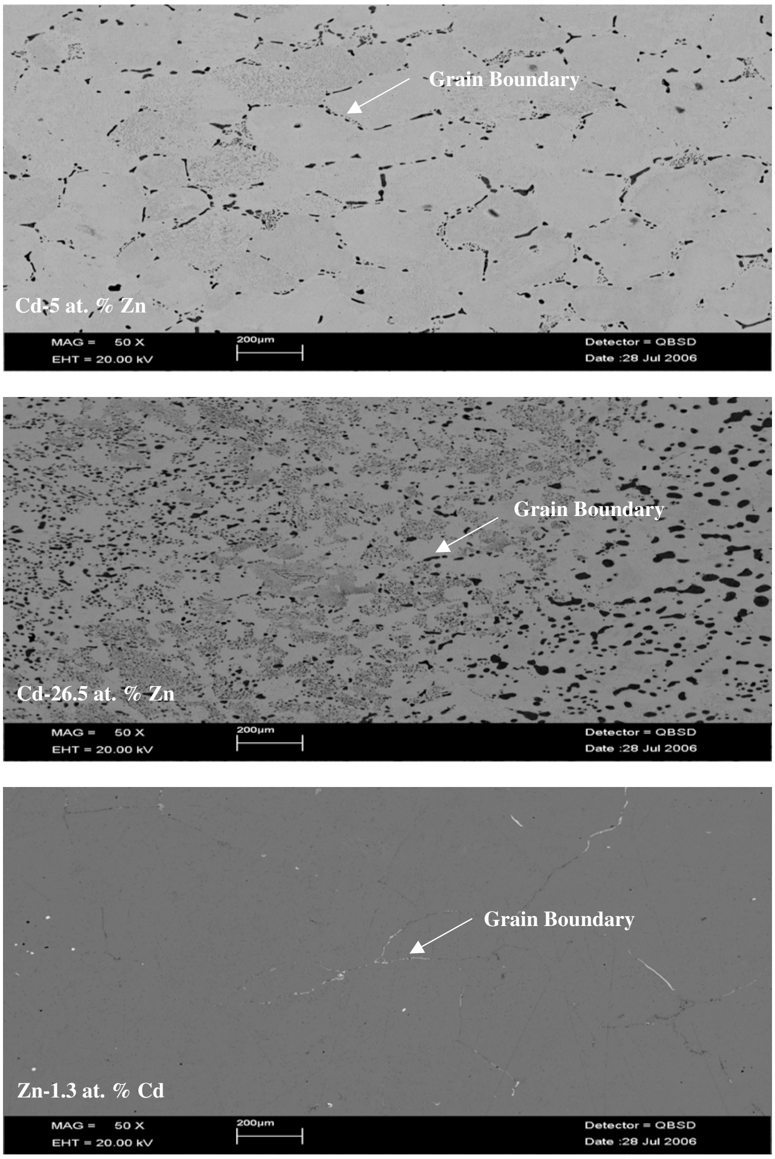
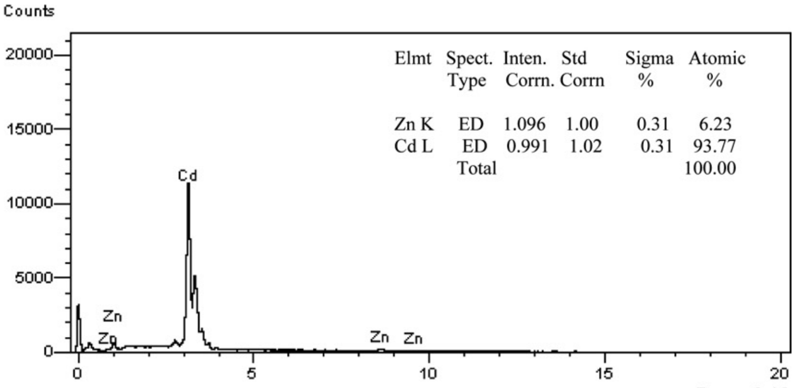


Figure 4. The scanning electron microscope photographs of Cd–Zn alloys.

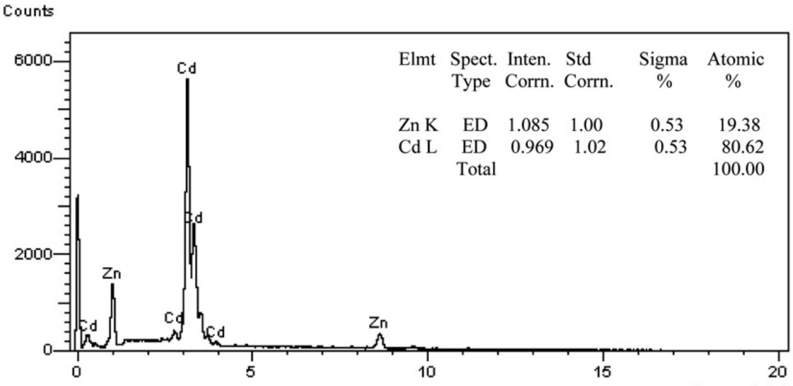
electrical conductivity has been obtained from the measured values of resistivity using standard equations. The characteristics of the electrical conductivity results of the Cd–Zn alloys can be seen in figure 7, which shows the electrical conductivity of samples versus temperature. The electrical conductivity of the Cd–Zn alloys decreases exponentially with the increasing temperature and nearly independent of the composition.

In addition, the Wiedemann–Franz relation was examined for the Cd–Zn alloys. The thermal conductivity of metallic materials is generally composed of an electronic component and a phonon component. The Wiedemann–Franz relation is a criterion for identifying the carrier of thermal conduction. When the electronic component contributes dominantly to



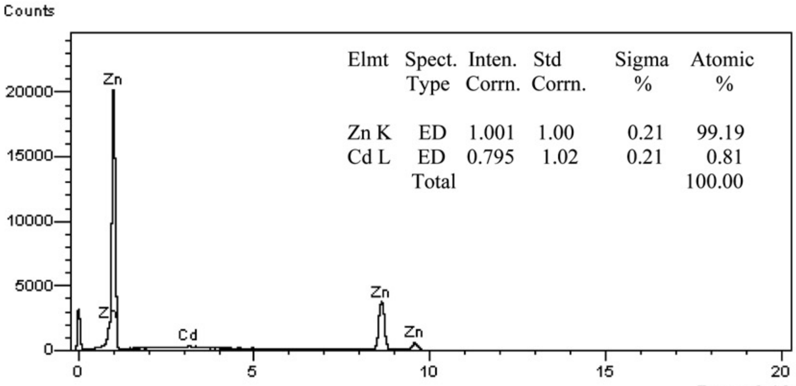
Elmt	Spect. Type	Inten. Corrn.	Std Corrn	Sigma %	Atomic %
Zn K	ED	1.096	1.00	0.31	6.23
Cd L	ED	0.991	1.02	0.31	93.77
	Total				100.00

Cd-5 at. % Zn Sample



Elmt	Spect. Type	Inten. Corrn.	Std Corrn.	Sigma %	Atomic %
Zn K	ED	1.085	1.00	0.53	19.38
Cd L	ED	0.969	1.02	0.53	80.62
	Total				100.00

Cd-26.5 at. % Zn Sample



Elmt	Spect. Type	Inten. Corrn.	Std Corrn.	Sigma %	Atomic %
Zn K	ED	1.001	1.00	0.21	99.19
Cd L	ED	0.795	1.02	0.21	0.81
	Total				100.00

Zn-1.3 at. % Cd Sample

Figure 5. The EDX analysis of Cd–Zn alloys.

the total thermal conductivity, the Wiedemann–Franz relation could hold, which is given by $K = LT\sigma$, where K is the thermal conductivity, L is the Lorenz number, T is the absolute temperature and σ is the electrical conductivity. The thermal and electrical conductivity values were used to obtain the Lorenz number [18].

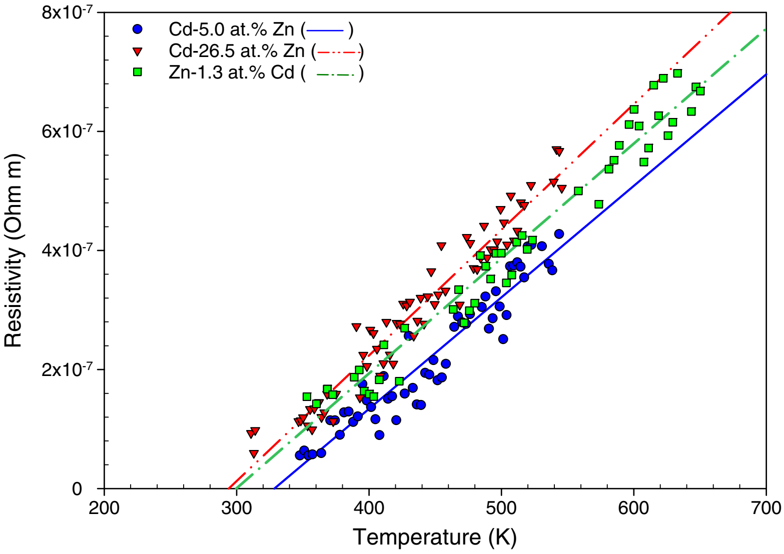


Figure 6. Electrical resistivity of the Cd–Zn alloys versus temperature.

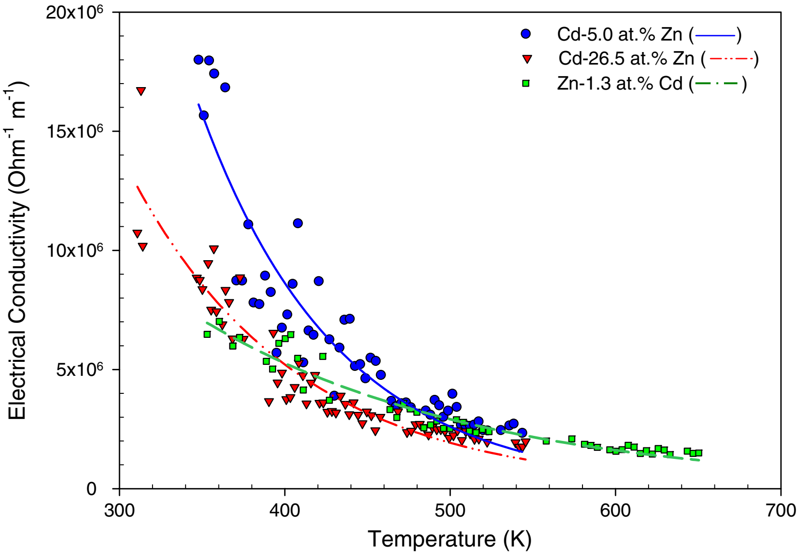


Figure 7. Temperature and composition dependence of electrical conductivity for the Cd–Zn alloys.

The experimental results of the thermal and electrical conductivity of the samples have been used to calculate Lorenz number for 373 and 533 K. The calculated Lorenz numbers are given in table 1. In the low temperature range ($T < 373$ K), the obtained values of Lorenz number approximately approach the typical metallic Lorenz number of 2.44×10^{-8} W Ω K^{-2}. These results indicate that Cd–Zn alloys satisfy the Wiedemann–Franz relation for $T < 373$ K and the dominant carriers of thermal conduction in the Cd–Zn alloys are electrons rather than phonons. Above this temperature ($T > 373$ K), the calculated Lorenz numbers are different from the typical metallic values. This indicates that the phonon component of the thermal conduction starts to dominate the transport processes. In this temperature range, the Weidemann–Franz relation is no longer valid.

In addition, the temperature coefficient of the Cd–Zn alloys, α, was also estimated by using the following equation in the temperature range between 373 and 533 K [19]:

$$\alpha = (1/K_1)(\mathrm{d}K/\mathrm{d}T) = (1/K_1)(\Delta K/\Delta T) \qquad (3)$$

where α is the temperature coefficient in the temperature range $\Delta T = T_2 - T_1$. $\Delta K = K_2 - K_1$; K_1 and K_2 are the thermal conductivities at T_1 and T_2, respectively. The results of the temperature coefficient for different Cd–Zn alloys (Cd solid solution, eutectic phase and Zn solid solution) are given in table 1. The results show that the temperature coefficient is independent of composition of Cd and Zn.

6. Conclusions

In summary, the thermal and electrical conductivities of the Cd–Zn alloys have been investigated. Thermal conductivities have been obtained by the radial heat flow method and compared with the results of the pure Cd and the pure Zn. Our thermal conductivity results are consistent with the pure Cd and Zn results. The temperature coefficient of the Cd–Zn alloys has been determined by using the thermal conductivity values. The results show that the temperature coefficient of the Cd–Zn alloys is independent of composition of Cd and Zn.

In addition, the patterns of electrical conductivity of Cd–Zn alloys with different compositions have been investigated as a function of temperature by using the four-point probe technique. The electrical resistivity increases linearly and the electrical conductivity decreases exponentially with temperature. The electrical conductivity and resistivity values are slightly dependent on Cd and Zn composition.

Also, the experimentally obtained thermal and electrical conductivities are used in order to determine Lorenz number. In the low temperature range ($T < 373$ K), the results indicate that the Cd–Zn alloys satisfy the Wiedemann–Franz relation and the carriers are electrons. Above this temperature, the phonon component of thermal conduction dominates the processes and the Wiedemann–Franz relation is not valid.

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