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Thermoelectric properties of zintl phase antimonite compounds, $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ prepared by microwave-assisted solid-state technique

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Abstract: **In a relatively short duration (10 minutes) under a microwave irradiation method prepared the chemical Zintl compounds $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ by using an active carbon as a susceptor material. These samples were then analyzed by the XRD, SEM. The XRD revealed that the compounds crystallized in the hexagonal structure where ZnSb precipitated as a secondary phase in almost all the compounds. Thermoelectric characterizations have been performed for three obtained samples a maximum power factor of 2.62 $\mu W/cmK^2$ at 523K was noticed for $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$ sample with carrier concentration of 5.11×10^{21} cm^{-3} at 300K. And the compound $YbCd_{1.6}Zn_{0.4}Sb_2$ is found the power factor of 8.20 $\mu Wcm^{-1}k^{-2}$ at 523K with carrier concentration of 3.04×10^{21} cm^{-3} at 300K. The compound $YbZn_{1.6}Mn_{0.4}Sb_2$ the power factor of 1.78 $\mu W/cm\ k^2$ at 523 K with carrier concentration of 4.8×10^{21} cm^{-3} at 300 K.**

Keywords: **microwave irradiation, zintl phase, active carbon, power factor.**

1. INTRODUCTION

Thermoelectricity refers to a phenomenon of thermal and electrical energies can be converted to each other under varied temperatures and different voltages [1-4]. According to the current development in the industrial material fields, ingots have emerged associated with much attention due to their use in many thermal and electrical applications[5]. High thermoelectric figure of merit $ZT=(S^2\sigma/k)T$, where S, σ , K, and T are Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature, respectively [6-9]. The power factor ($S^2\sigma$) is considered as the most important part in the study of thermoelectric materials (TEMs). To reach the high value in the figure of merit (ZT), higher value of power factor and lower thermal conductivity are required [10]. Zintl phase compounds have been prepared by direct solid state technique and found taking a long time under heating for example: $YbZn_{2-x}Mn_xSb_2$ was synthesized at 1323k for 30h[11] ,$Ca_{16}Sb_{11}$ was synthesized at 1423k for several hours [12], $YbCd_{2-x}Mg_xSb_2$ at 1273k for 72h [13], $Ca_{1-x}Yb_xZn_2Sb_2$ at 1273k for 48h[14], $YbZn_2Sb_2$ at 1323k for 30h[15], $SrZn_2Sb_2$ at 1073k for 5days[16]. $Ca_{1-x}Eu_xZn_2Sb_2$ at 1273k for 3 days[17], $Yb_xEu_{1-x}Cd_2Sb_2$ at 1200 °C for 24 h[18]. While in our work the Zintl compounds $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ are synthesized in short time 10 mins under a microwave-assisted solid state. Microwave irradiation is an efficient method employed to prepare the thermoelectric materials in short time. In comparison with bulk samples, metal powders can feasibly couple under microwave fields at 2.45 GHz and heating reach to 1273 K without causing visible electric discharges[19]. The Zintl phase families like 1-2-2-family which includes $EuZn_2Sb_2$,$YbZn_2Sb_2$, $YbCd_2Sb_2$, $YbMn_2Sb_2$ are considered as important TEMs[11,13,17]. The Zintl compounds crystal structure consist of layered structures with a triangle lattice formed by cation vacancies A^{+2} between two dimensional $(B_2Sb_2)^{-2}$ network is covalently bounded slabs separated by cationic layers [20]. Importantly, a microwave-assisted solid state provides high energy, easy work up and eco-friendly methodology than other traditional techniques[21].

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2. EXPERIMENTAL

The rapid microwave synthesis was used to prepare the Zintl phase antimonite compounds from pure elements (Eu, Yb, Zn, Cd, Mn and Sb > 99.99%) semiconductor compounds were weighted 2 g according to the stoichiometric $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$. The powder is mixing into an agate mortar and pestle to create a homogenous mixture for 20 mins, and put it inside a clean quartz ampoule and sealed under high vacuum of 10^{-5} mbar. The ampoule was irradiated with a maximum power of 1000W microwave oven (LG) (MS2147C 1000 W) at 2.45 GHz. An ctive carbon (susceptor) was surrounded the quartz ampoule to absorb microwave irradiation and initiate heating under a microwave-assisted solid state synthesis. The active carbon helps to raise reaction temperature to reach 1123K for 10 mins (2on:2off) as shown in Fig.1. The temperature of ampoule was measured using an infrared thermometer (S-CA-1168), temperature range 223-1123 K. The fusing materials were further cooled to room temperature to obtain ingot. The morphological, and stoichiometric ratio, structural and were measured . The polycrystalline selected portions of the ingots were imaged using field emission scanning electron microscopy (FESEM), after grinding, the powders were then measured to determine their crystal structure using X-ray diffraction (XRD, PANalytical X'Pert PRO MRD PW3040-Netherlands). The Seebeck coefficient (S) of polished disks was measured by the slope of the linear relationship between the thermoelectromotive force and the temperature difference between the two ends of each sample, more detail in previous report [22].The four-point probe method was using to measure the electrical conductivity (σ) in a vacuum at 10-3 mbar at a temperature range of 298K -523K. The carrier concentration (n) was determined at room temperature from the Hall voltage measurement with an applied magnetic field of 1 T using a PHYWE electromagnetic (model: 6480, Germany).

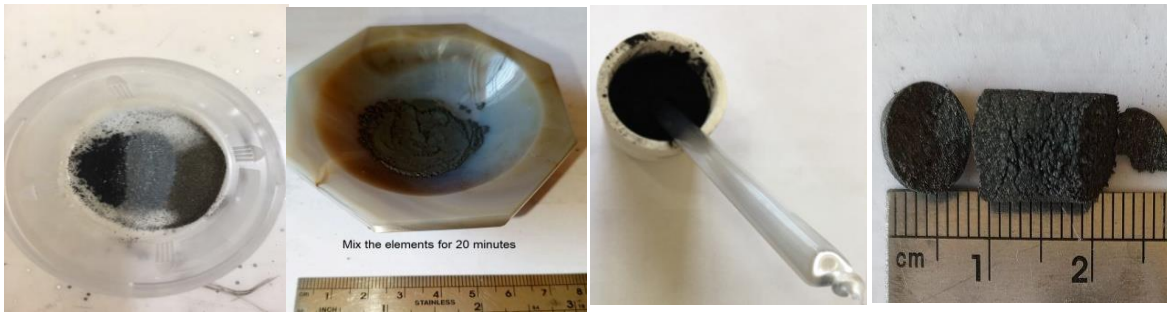

Fig.1: Synthesize the ingots of the compounds

3. RESULTS AND DISCUSSION

X-ray diffraction (XRD) of $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ are shown in fig.2. The XRD revealed that the compounds crystallized in the hexagonal structure where ZnSb precipitated as a secondary phase in compounds. The appearance of the secondary phase in the X-ray diffraction results of zintl compounds is consistentwith published works in this field[23].

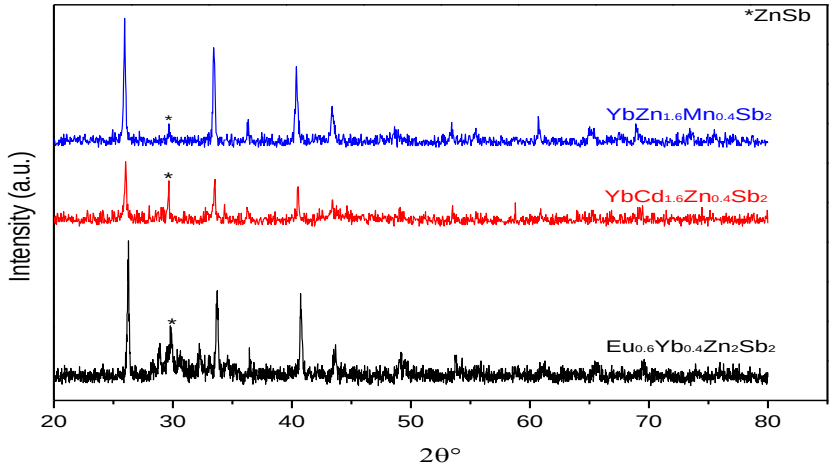

Fig.2: XRD patterns of $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$

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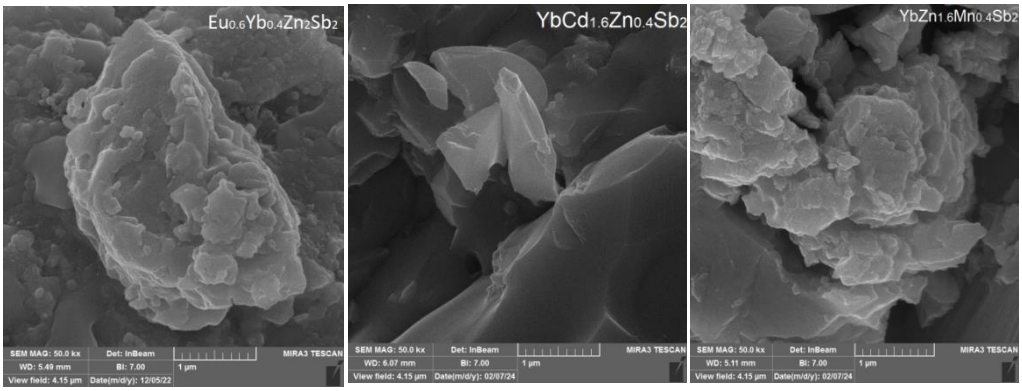

Fig.3: SEM image of $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$

The electrical conductivity (σ) of the $YbCd_{1.6}Zn_{0.4}Sb_2$ increased when the temperature was increased and this behavior was in agreement with degenerate semiconductor behavior of while the compounds $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ have relatively little change as shown in Fig.4 .

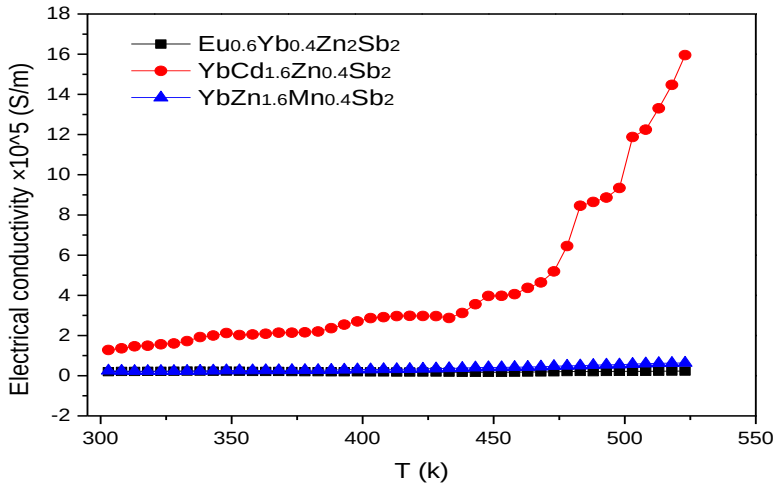

Fig.4: Electrical conductivity (σ) of the $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$

The Fig.5 shows the dependent temperature of the Seebeck coefficient for $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$. The measured values of Seebeck coefficient for are a negative value n-type conductivities which means that the majority of carriers are electrons, while the compound $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$ is p-type as the holes which are represented the majority of carriers of electrical transports.

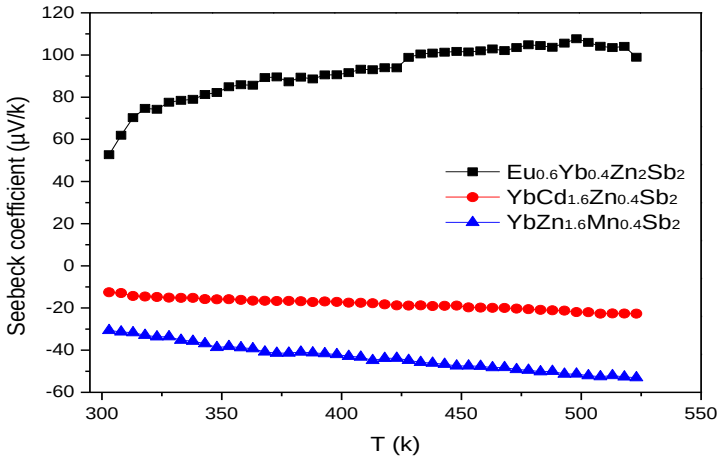

Fig.5: Seebeck coefficient of the $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$

The dependent temperature of the power factor ($S^2\sigma$) of Zintl compounds the $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ that were prepared under microwave assisted solid state as shown in Fig.6. The high calculated value of power factor ($S^2\sigma$) for the prepared sample $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$ was 2.62 µW/cmk^2 at 518 K .

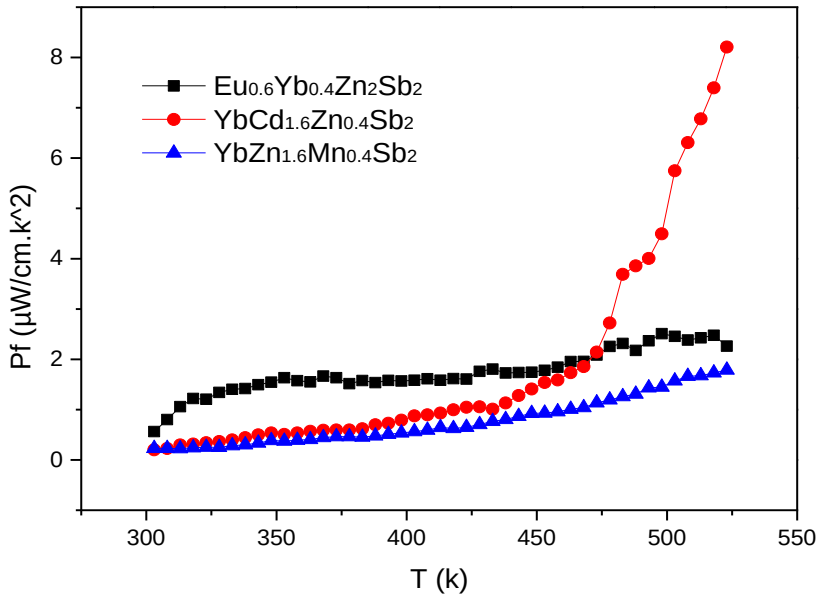

Fig.5: Power factor ($S^2\sigma$) of the $Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$

4. CONCLUSIONS

$Eu_{0.6}Yb_{0.4}Zn_2Sb_2$, $YbCd_{1.6}Zn_{0.4}Sb_2$, $YbZn_{1.6}Mn_{0.4}Sb_2$ are Zintl chemical compounds have been successfully produced using microwave assisted solid state at short time . The XRD analysis detects that prepared compounds has a hexagonal structure where ZnSb precipitated as a secondary phase in almost all the compounds. The $YbCd_{1.6}Zn_{0.4}Sb_2$ sample reveals moderate electrical conductivity and a higher Seebeck coefficient with maximum power factor of 8.20 µW/cmK^2 at 523 K.

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Ethical Approval: The study protocol was approved by local ethics committee.

Disclaimer: None

Conflict of interest: There are no conflicts of interest to declare.

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