

# The Influence of Thermoelectric Power and Grain Size on Three Groups $\text{Cu}_x\text{Zn}_{1-x}\text{O}$ , $\text{Fe}_x\text{Zn}_{1-x}\text{O}$ and $\text{Mg}_x\text{Zn}_{1-x}\text{O}$

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**Abstract:** The aim of this research is to study the influence by adding ZnO to the oxides CuO, FeO and MgO preparing by metallurgical powders according to chemical formula's  $\text{Cu}_x\text{Zn}_{1-x}\text{O}$ ,  $\text{Fe}_x\text{Zn}_{1-x}\text{O}$  and  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  respectively, at values of x equal to 0, 0.2, 0.4, 0.6, 0.8, 1 from thermoelectric Power (seeback effect). The influence addition of ZnO from the values of thermoelectric Power (seeback effect) at about temperatures (300-450) K, the values indicate that these values were decreased by increasing the temperatures of these groups, specially at high temperatures in general for all samples accordingly about the percentages of ZnO inside them in which it's effect on the mobility of the charge carriers and it's effective mass distributed with defects. Also studying the influence the grain size from thermoelectric Power through Gamma Ray and scanning electron microscope (SEM), then proceeded this way about the samples having same concentrations and samples with different concentrations, therefore the results indicated that thermoelectric Power factors increased by increased the grain size of samples that having different concentrations

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## Introduction

ZnO and it's groups such as  $\text{Cu}_x\text{Zn}_{1-x}\text{O}$ ,  $\text{Fe}_x\text{Zn}_{1-x}\text{O}$  and  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  (ceramic materials) have been widely used in various components for application, due to their highly electrical resistivity, mechanical hardness, and chemical stability. The aim of this work is to investigate the effects of the compositional change of a stoichiometric ZnO by partial oxides CuO, FeO, and MgO. The effect of ZnO on thermoelectric power of some powders alloys of some metal oxides (MgO, CuO, and FeO) for different percentages (0, 0.2, 0.4, 0.6, 0.8, 1)% in temperature ranges (300-450K). Thermoelectric power (seeback effect) is one of best methods to obtain cleaner energy through the conversion of subtractive heat to electrical energy in which can be used in many fields such as cooling solid device found in circuits by removing the heat which have negative effect about it's function, although introduced in manufacturing biotermol batteries and the important clinic fields representing of the manufacturing regulator pacemakers heart, sensitive resistors heat <sup>(1)</sup>, and thermoelectric equipment's such as Beltier refrigerator, in spite of experimental employment which gives to us data about electrical conduction mechanisms, the concentration and nature of charge carriers <sup>(2)</sup>.

The thermoelectric phenomenon discovered firstly by Johan Thomas Seeback in 1821 while generating electrical current in closed circuit made of two different materials, if it's junction points at two different temperatures which related to his name seeback effect <sup>(3)</sup>, since that discovery has been cared by researchers <sup>(4)(5)</sup> to study the thermoelectric properties in semi-conductors due to it's importance in many applied fields, in addition to each researchers <sup>(6)</sup> and <sup>(7)</sup> were interested in studying the dependence seeback factor on temperature in conduct semiconductors at different ranges of temperatures which interacted with it, other researchers studied the influence of charge carriers concentration on seeback effect <sup>(8)</sup>, some researchers were interested in studying the effect of magnetic field and it's direction on seeback factor <sup>(9)(10)</sup>, now a days thermoelectric power is used as a measure of semi-insulators such as gallium nitrite  $\text{GaN}^{(11)}$ . The researcher <sup>(12)</sup> improved the capacity of thermoelectric power materials at high temperatures and introduce in manufacture devices, besides some research restudied generating high thermoelectric power by using nano technique from  $\text{Y}_2\text{O}_3^{(13)}$ , also thermoelectric power were used in manufacturing many sensors <sup>(14)</sup>.

## Materials and Methods

Present study was done in experimental laboratory to study the oxide powders that were prepared by metallurgical powders. The samples were divided in three groups  $\text{Cu}_x\text{Zn}_{1-x}\text{O}$ ,  $\text{Fe}_x\text{Zn}_{1-x}\text{O}$  and  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  according to the test. The required weight of the oxides ZnO, CuO, FeO, and MgO powders was weighted by using electronic balance for each group. Mixing of powder oxides was done by mortar and pestle until a homogenous powder is produced by manual method named as metallurgic powder.

## Experimental

we prepared the required samples in this work from pure zinc oxide ZnO, copper oxide CuO, iron oxide FeO and magnesium oxide MgO according to atomic weight with percentages ranged by (0,0.2,0.4,0.6,0.8)wt%, so we have three groups, each group having four samples with different concentrations, the table(1) indicates the chemical structures of samples.

At the beginning of preparing samples we heated the oxides powder by using furnace at  $70^\circ\text{C}$  in 3 hours in order to guarantee removing any humidity that influenced about the operation of definition the percentage weight, after there we compressed the samples at pressure 17 ton/cm<sup>2</sup> by form of a disks it's diameter 13mm and thickness of 2-3mm by using England mechanical piston <sup>(15)</sup>, final stage we proceeded heat treatment of all samples in term of 4 hours at temperature  $1000^\circ\text{C}$  then cooled them inside the furnace gradually until room temperature.

**Table (1) the chemical structures of prepared samples**

First group	Second group	Third group
CuO	FeO	MgO
$\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$	$\text{Fe}_{0.8}\text{Zn}_{0.2}\text{O}$	$\text{Mg}_{0.8}\text{Zn}_{0.2}\text{O}$
$\text{Cu}_{0.6}\text{Zn}_{0.4}\text{O}$	$\text{Fe}_{0.6}\text{Zn}_{0.4}\text{O}$	$\text{Mg}_{0.6}\text{Zn}_{0.4}\text{O}$
$\text{Cu}_{0.4}\text{Zn}_{0.6}\text{O}$	$\text{Fe}_{0.4}\text{Zn}_{0.6}\text{O}$	$\text{Mg}_{0.4}\text{Zn}_{0.6}\text{O}$
$\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$	$\text{Fe}_{0.2}\text{Zn}_{0.8}\text{O}$	$\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$
ZnO	ZnO	ZnO

## Result and Discussion

### 1. Thermoelectric Power (Seeback Effect)

The above way as followed to measure the seeback effect parameter  $S$  to make difference in temperature  $\Delta T$  from the two sides of the sample after connecting it to heater on one side, since the fall in temperature  $\Delta T$  reproducing thermoelectric power which appears potential difference  $V$  about the side of the sample in order to measure temperature, we fixed the two couples of thermocouples from copper-constantan on both upper and lower disk of the two sample faces. Fig(1) shows the thermoelectric circuit using for measuring seeback factor, where  $T_1$  and  $T_2$  represented the temperature about the two faces of the sample, so the average temperature  $T$  is equal as  $T = (T_1 + T_2)/2$ , but the difference between the two temperatures  $\Delta T$  is equal to  $\Delta T = T_1 - T_2$ , when  $\Delta V$  represented the thermoelectric power produced by the temperature difference on two sides of the sample, therefore seeback factor is equal to

$$S = \Delta V / \Delta T$$

Many important points were taken into consideration in order to lower the percentage error in the measurements specially the heating losses through heating radiate from the side's surfaces in order to obtain the best contact between the sample and sample holder. Measurement apparatus from type of Marconi were used in measuring the required potential difference. Temperature controller used to fix the lower disk temperature (Heat sink).

Fig (2) shows the relation between seeback factor  $s$  and temperature  $T$  ranged between 280K to 460K of the group  $\text{Mg}_x\text{Zn}_{1-x}\text{O}$  which noted that seeback factor decreased linearity for all samples when temperature increased. This would eventually lead to increase the number of charge carriers and finally increased the mobility is which lead to increase the electrical conduction which inversely proportional to seeback factor.

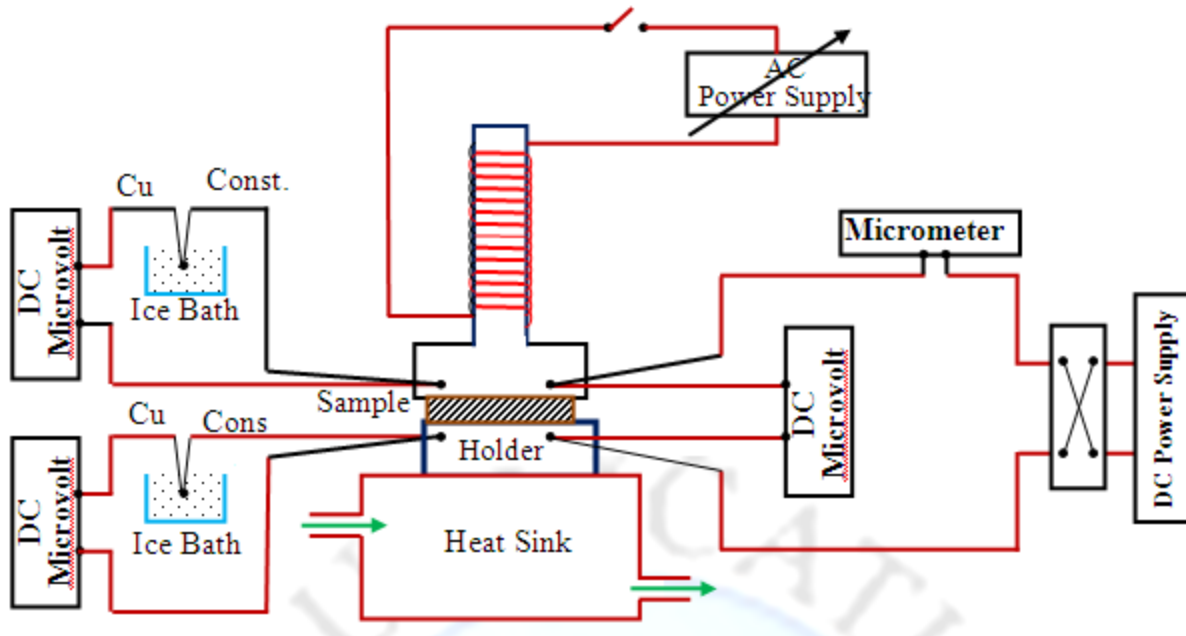


Fig. 1 : circuit of thermoelectric power

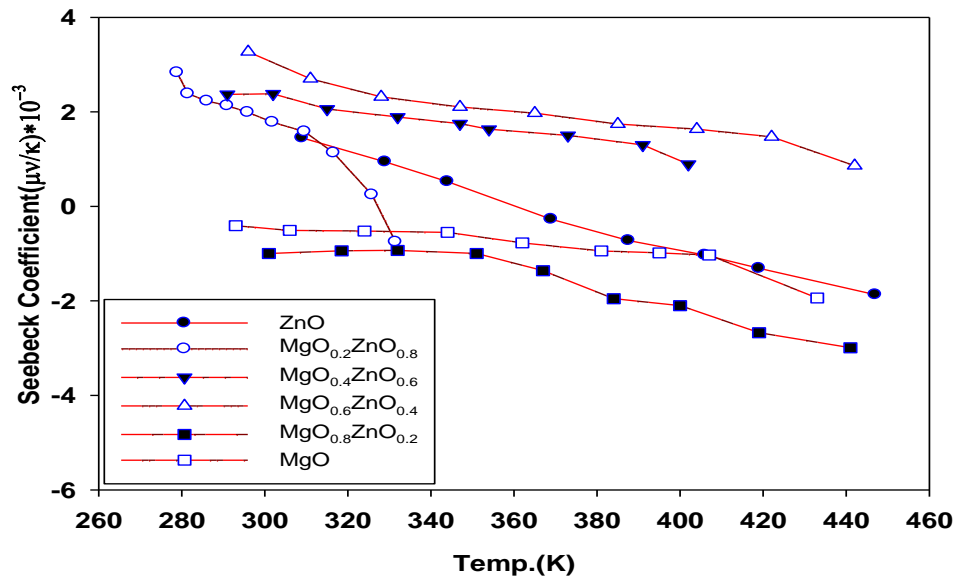


Fig. 2: The relation between seeback factor and temperature of  $Mg_xZn_{1-x}O$

Fig (3) shows the relation between seeback factor  $s$  and temperature  $T$  ranged between 280K to 460K of the group  $Fe_xZn_{1-x}O$  which noted that seeback factor increased for the samples  $Fe_{0.2}Zn_{0.8}O$  and  $Fe_{0.4}Zn_{0.6}O$  by increasing temperature. This would eventually lead to decrease the mobility of charge carriers or change it and finally increased the mobility or change to it effective mass in the same range, in addition the constant increased in return to exist more than one energy defect plane in covalent band with different ionic energies which lead to obtain this increasing, these results are coincide with the results of seeback effect from germanium but the remaining samples noted that there seeback factor increased slowly in the range between temperatures 300K and 350K, this is because the numbers of charge carriers which produced from the ionic about most ions and defects keep constant [2], never the less decreasing in values of seeback factor in the next range for temperature nearly 360K, this is because that the numbers of charge carriers in these samples resultant the defects possessions more than one energy plane in which having different ionic energies.

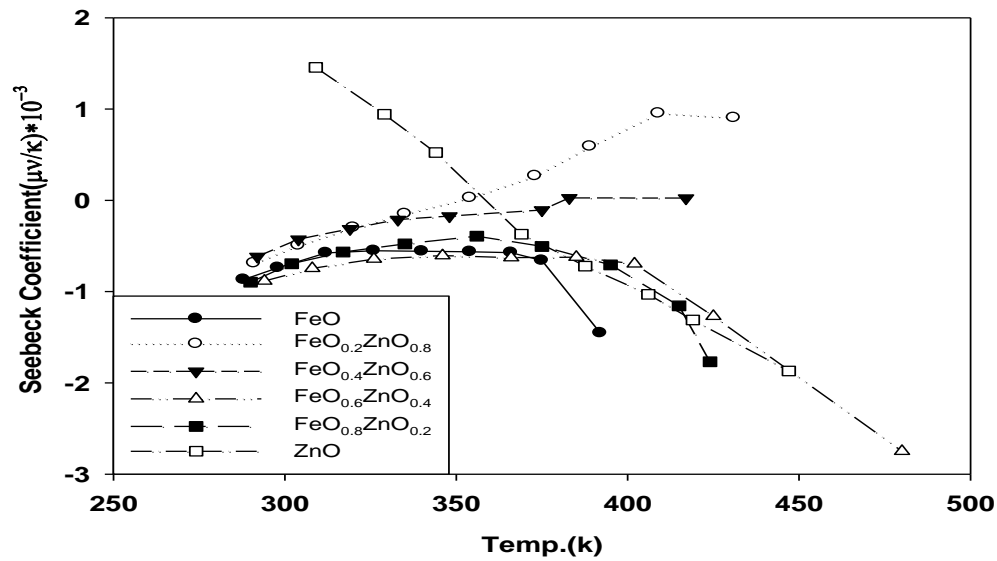


Fig. 3: The relation between seeback factor and temperature of  $\text{Fe}_x\text{Zn}_{1-x}\text{O}$

Fig (4) shows the relation between seeback factor  $s$  and temperature  $T$  of the group  $\text{Cu}_x\text{Zn}_{1-x}\text{O}$  which noted that' seeback factor decreased from increasing ZnO percent obviously but the little swung to higher and lower because the defects possessions more than one energy plane in which having different ionic energies. Although the net sample CuO noted that slowly increasing in the value of seeback factor as temperature increased, this is because of the increasing the number of charge carriers which produced from most ions and defect.

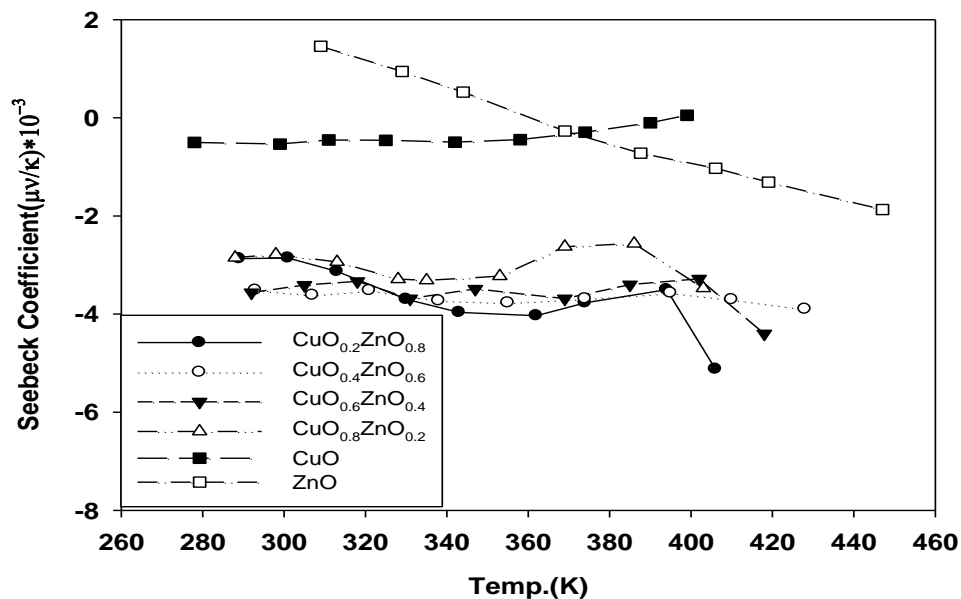


Fig. 4: The relation between seeback factor and temperature of  $\text{Cu}_x\text{Zn}_{1-x}\text{O}$

we discussed the influence each percent addition from all three groups in this work through Fig(5), which shows the variation of seeback factor and temperature from the samples having 0.2% Zinc Oxide, since it may be noted that this percent is affected in separate form, but from Iron Oxide FeO making increasing of the value seeback factor approximately in the range 290K and 350K after that we noted decreasing the value of seeback factor in behind this rang.

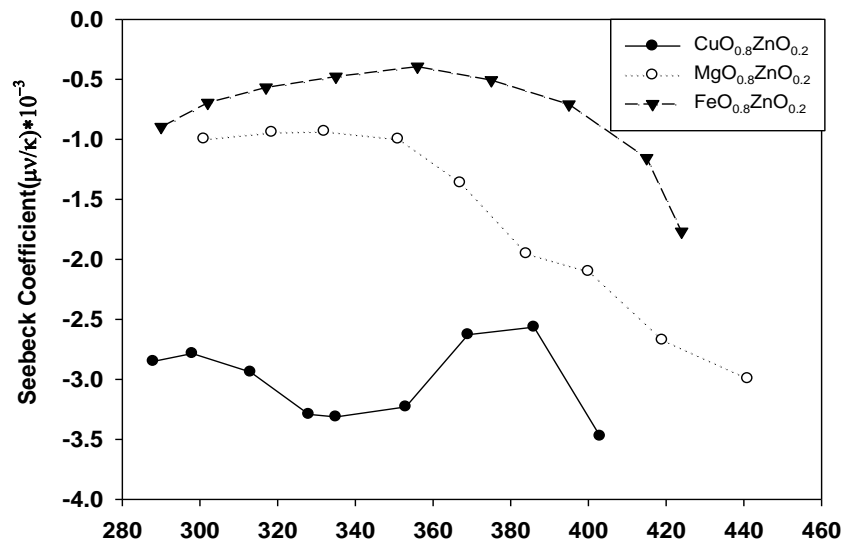


Fig. 5: The relation between seeback factor and temperature of 0.2% ZnO

Fig(6) shows the relation between seeback factor and temperature ranged between 300K to 350K, in which the samples having 0.4% Zinc Oxide, since it may be noted that there are continuously decreasing about magnesium oxide MgO, this is because of the increasing number of charge carriers and it's mobility by increasing temperature which execute to decrease the value of seeback factor upon this effect from FeO and CuO are semi linearity till to temperature 400K which referred to fixing the number of charge carriers in that range.

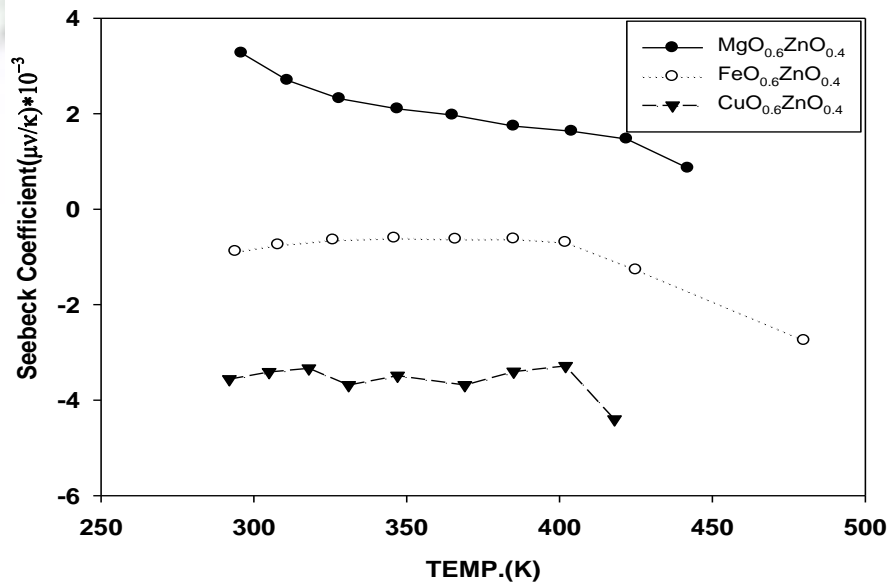


Fig. 6 : The relation between seeback factor and temperature of 0.4% ZnO

The same thing we noted about the samples having 0.6% Zinc Oxide, from Fig(7) which showed that there were decreasing the values of seeback factor about magnesium oxide MgO, upon this effect from CuO is semi linearity which means that this effect making fixed of their number of charge carriers produced from thermionic of most ions and defects, but this effect for FeO increases slowly the values of seeback factor which produced from decreasing the mobility of charge carriers and it's effective mass.

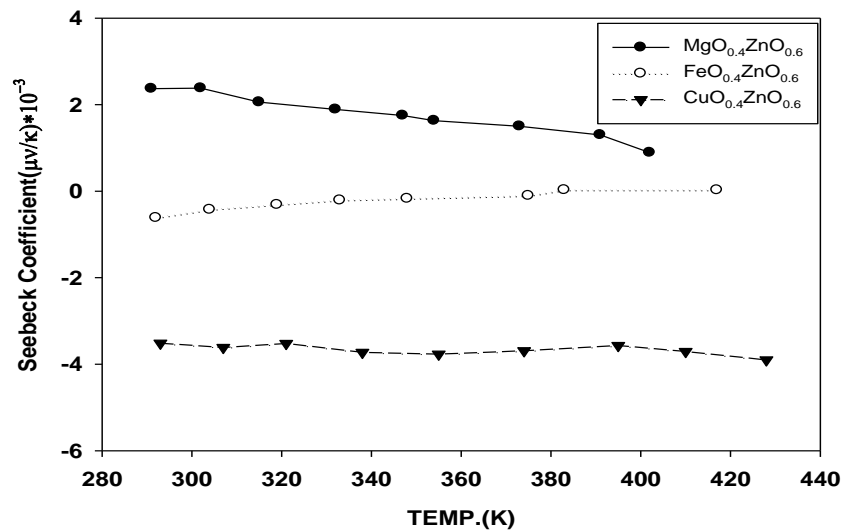


Fig. 7: The relation between seeback factor and temperature of 0.6% ZnO

Fig(8) shows the samples having 0.8% Zinc Oxide which affects the values of seeback factor of the three oxides MgO, CuO and FeO in temperatures ranged between 280K to 450K, this affect is strong according to MgO in the temperature range 280K to 330K from seeback factor because the majority of charge carriers which produced from the ionic of most the acceptor atoms still constant and highly more in number than the charge carriers produced from self ionic or electronic transmission via the energy gap.

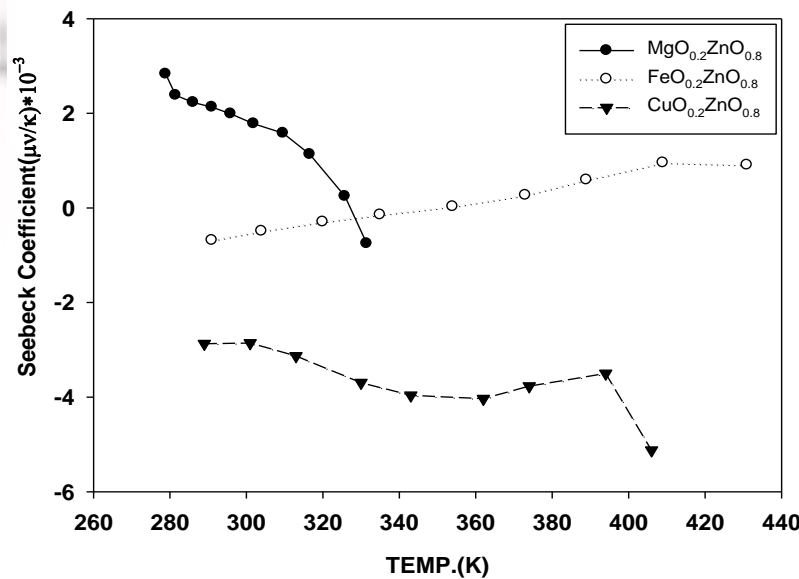


Fig. 8: The relation between seeback factor and temperature of 0.6% ZnO

## 2. The Grain Size and it's Influence on Seeback Factor

In this section we study the grain size about three samples of  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$ , which prepared under the same conditions of hydrostatic press and heat treatment by using scanning electroscopic (SEM), which dealt at temperatures (280-300, 350-360, 390-410)K, we found the values of seeback factors a results obtained from table(2), which indicate that values are very proximate, these results are coincided with microstructure pictures in Fig(9) from three samples of  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$ .



Table (2): The values of seeback factor at various temperatures from three samples of  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$

$\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$	Seeback Coefficient( $\mu\text{V/K}$ ) $\times 10^{-3}$		
	(290-300)K	(350-360)K	(390-400)K
1	-2.85	-3.22	-3.47
2	-2.91	-3.14	-3.51
3	-2.76	-3.10	-3.44

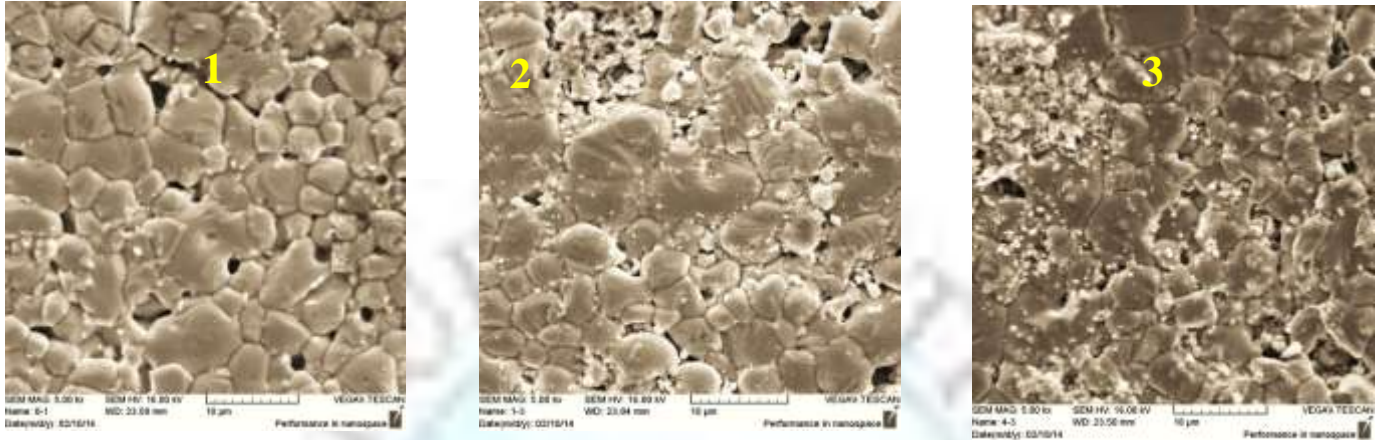


Fig. 9: The microstructure pictures from three samples of  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$

Another study in this section the effect of grain size with seeback factor by using two samples with different percent are  $\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$  and  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$  respectively, which dealt at temperatures 300K, 350K and 400K, we found values of the seeback factor as results obtained from table(3), these results are coincided with microstructure pictures in Fig (10) from the two samples of  $\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$  and  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$ .

Table (2): The values of see back factor at various temperatures from three samples of  $\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$  and  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$

Samples	Seeback Coefficient( $\mu\text{V/K}$ ) $\times 10^{-3}$		
	(300-310)K	(340-360)K	(400-410)K
$\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$	-2.9	-3.22	-3.47
$\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$	-2.85	-3.96	-5.12



Fig. 10: The microstructure pictures from three samples of  $\text{Cu}_{0.2}\text{Zn}_{0.8}\text{O}$  and  $\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}$  and

### References

- [1]. Terry M.Tritt and M.A. Subramanian, (2006), "Thermoelectric Materials, phenomenon, and Applications", Mrs Buletin.Vol. 31.
- [2]. Bowers R., YreR. W., Bauerle J.E., and Cornish A.J.,(1959)," InAs and InSbas Thermoelectric Materials" J, Appl, Phys.Vol.30, No.6: P.930-934.
- [3]. Safa Kasap, (2001), "Thermoelectric Effects In Metals: Thermocouples", Canada.
- [4]. HaroM,Corrales C, VillaresP, MarquezE, and R. Jimenez-Garay , (1993)," Thermoelectric and Hall Effects In Amorphous AlloyAs<sub>0.20</sub>Se<sub>0.40</sub>Te<sub>0.40</sub>"Meteriais Letters,Vol.17:p.263-267.
- [5]. Yamaguchi Y., (1990)," Thermopower of Several Ternary Compounds of Cerium and Uranium: CeTX and UTX(T=Ni,Pd or pt:X=In or Sn", J.Phys:condens Matter, Vol2:p.5715-5721.
- [6]. Rathanyaka K.D.D., Trodahi H.J., KaiserA.B.,and Dillis,(1986),"Transport properties and Loclized Spin Fuctuations in P<sub>i</sub>Ni Alloys, " Phys.Rev.B33, No.5:P.3399-3402.
- [7]. KoyanoM , Negishi H., Ueda Y, Sasaki M., and Inoue M.,(1986),"Electrical Resistivity and Thermoelectric power of Intercalation Compounds M<sub>x</sub>TiS<sub>2</sub>(M=Mn,Fe,Co and Ni)" Phys.State. Sol.(b) Vol.138:p.357-362.
- [8]. Have S.and Hansberger R.,(1985)," Thermoelectric properties ofGal<sub>x</sub>Al<sub>x</sub>As"J.Appl,phys.Vol.57,No.112:p.5330-5335.
- [9]. 9.Burns M.J.(1989)"Thermoelectric power During Magnetic Field Induced Localization in Degenerately Doped n-Type Ge"Phyz.Rev.B20,No.8:p.5473-5478.
- [10]. TomoyochiAono,(1977),"Magneto-Seebeck of pb-Doped Bi-Sb", Hap.J.of Appl.phys.,Vol.17.No.5:p.843-849.
- [11]. M.Pavlovic,U.V.Desnica, Z.Q.Fang D.C.Look, (2003),"Thermoelectric effect spectroscopy measurements on semi-insulating Gan Science direct Vacuum"Vol.71, P. 153-158.
- [12]. R.D.Purohit, M. Syambadu and P.K. Sinha, (2011), "Development of High Temperature Thermoelectric materials and Fabrication of Device" Technology Development Article,Issue No.320.
- [13]. K. Koumoto.W.Ssed and S. Ozawa Huge,(1997),"Thermopower of porous Y<sub>2</sub>O<sub>3</sub>",Appl. Phys.Lett.,Vol.71.No.11,15.
- [14]. A.W.Van Herwaarden and P.M. Sarro,(1986), "Thermal Sensors Based on the Seeback Effect", Sensors and Actuators, 10321-346.
- [15]. K.A. Mohammed,H.A. Mohammed,(2010)."Electrical Conductivity and Thermoelectric Power of Iron- Magnesium(MgO)<sub>x</sub> (Fe<sub>2</sub>O<sub>3</sub>)<sub>1-x</sub>Compounds"Abhath Al- Yarmovk.