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Effect of Adding Indium on Wetting Behavior, Microstructure and Physical Properties of Tin- Zinc Eutectic Alloy

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Abstract: Effect of adding indium on microstructure, wetting process, thermal, electrical and mechanical properties of tin- zinc eutectic alloy have been investigated. Microstructure (started base line, lattice parameters, unit cell volume, crystal size and the shape of formed crystalline phases) and measured physical properties of tin- zinc eutectic alloy changed after adding different ratio of indium content. A little variation occurred in thermo-graph (Endo-thermal peaks) of $Sn_{91}Zn_9$ alloy after adding indium. The contact angle, melting temperature and specific heat of $Sn_{91}Zn_9$ alloy decreased after adding indium content. Also elastic modulus and internal friction values of $Sn_{91}Zn_9$ alloy decreased after adding indium content. But electrical resistivity and Vickers hardness values of $Sn_{91}Zn_9$ alloy increased after adding indium content. The $SnZn_9In_5$ alloy has adequate properties for solder applications.

Key words: tin- zinc eutectic alloy, thermal and mechanical properties, electrical resistivity, wetting process

1. INTRODUCTION

Over the past few years the study of lead free solder has become a hot subject. New lead free solder alloy has great attention from researchers around the world. Lead free solders fall into two groups: first has lower melting points and chiefly includes alloys of tin and bismuth. Second is group of alloys, those with higher melting points than tin-lead. The leading choice seems to be a tin-silver-copper alloy. There is a tin-rich ternary eutectic at about 217 °C. These alloys at 30 °C higher melting point are a challenge to use. Sn-Zn solder alloys are quite capable in terms of mechanical integrity but have poor oxidation and corrosion resistance. Many studies have been made on various alloy system solders based on Sn such as $SnZn_9$, $SnAg_{3.5}$, $SnAg_3Cu_{0.5}$, etc. as possible replacements [1, 2]. T. Ohoak et al investigated the dependence of frequency over a range of 0-3 Hz on Young's modulus and internal friction in $SnZn_9$ and $SnAg_{3.5}$ eutectic lead free solder alloys [3]. Several researchers [4- 7] operated to improve the properties of Sn-Zn lead free alloy by adding small amount of alloying elements such as Bi, Cu, In, Ag, Al, Ga, Sb, Cr, Ni, Ge to develop ternary and even quaternary Pb free alloys. The microstructures of the tin- zinc- aluminum lead free solder alloys, which prepared from the Zn-5Al master alloy and Sn, using scanning electron microscopy were investigated [8]. The microstructures and mechanical properties of $SnZn_{8.55}Ag_xAl_{0.45}Ga_{0.5}$ (x= 0.5-3 wt. %) lead free solder alloys were studied [9]. Small additions of Ag decreased the melting point of the $SnZn_{8.55}Ag_xAl_{0.45}Ga_{0.5}$ solder alloys while maintaining the same strength and ductility as the $Sn_{63}b_{37}$ solder alloy. The effects of adding alloying elements such as Ag, Al, and Ga, on melting temperature, microstructures and mechanical properties of the $SnZn_9$ lead free solder alloy were studied [10]. The results show, $SnZn_9Ga_{0.5}$ alloy has very good UTS and elongation, which are better than both those of the $SnZn_9Ag_{0.5}$ and $SnZn_9Al_{0.45}$ alloys. Also effect of adding Al and Cu on microstructural and mechanical properties as well as thermal behavior of $SnZn_9$ lead free solder alloy was investigated [11]. The results indicate the microhardness of the $SnZn_9Al_{0.5}$ alloy was also higher than that of the $SnZn_9Cu_{0.5}$ alloy. Tin- zinc is desired to have a lead free solder with a melting temperature close to the eutectic temperature of the tin- lead alloy. Aluminum has a high melting temperature and good electrical conductivity. It may form solid solutions with tin and zinc. . So that, adding aluminum to tin- zinc solder alloy retain the melting point as low as possible but the soldering temperature higher than that eutectic tin- lead alloy. The aim of this work was to investigate the effect of adding different ratio from indium on microstructure, wetting behavior, thermal, electrical and mechanical properties of tin- zinc eutectic lead free solder alloy.

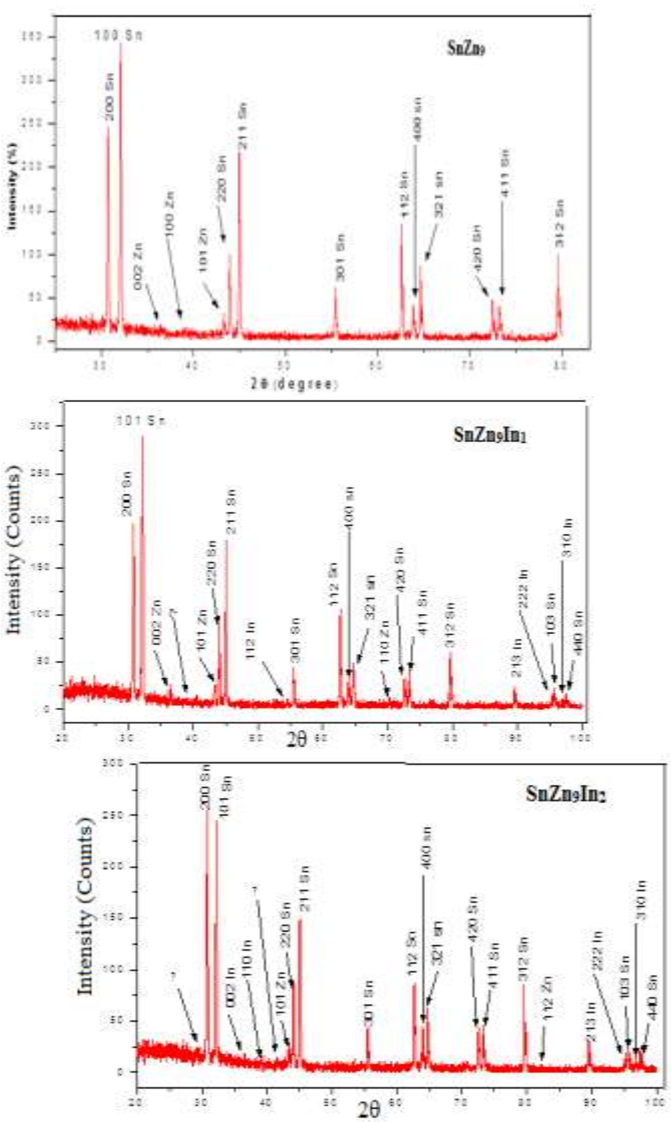
2. EXPERIMENTAL WORK

The alloys $Sn_{91-x}Zn_9In_x$ (X=0, 1, 2, 3, 4 and 5 wt. %) which used tin, zinc and indium elements with a high purity, more than 99.95%, were molten in the muffle furnace. The resulting ingots were turned and re-melted several times to increase the homogeneity of the ingots. From these ingots, long ribbons of about 3-5 mm width and ~ 80 μm thickness were prepared as the test samples by directing a stream of molten alloy onto the outer surface of rapidly revolving copper roller with surface velocity 31 m/s giving a cooling rate of 3.7 × 10^5 k/s. The samples then cut into convenient shape for the measurements using double knife cuter. Structure of used alloys was performed using an Shimadzu x–ray diffractometer (Dx–30, Japan) of Cu–Kα radiation with λ=1.54056 Å at 45 kV and 35 mA and Ni–filter in the angular range 2θ ranging from 20 to 100° in continuous mode with a scan speed 5 deg/min. Scanning electron microscope JEOL JSM-6510LV, Japan was used to study microstructure of used samples. The melting endotherms of used alloys were obtained using a SDT Q600 V20.9 Build 20 instrument. A digital Vickers micro-hardness tester, (Model-FM-7- Japan), was used to measure Vickers hardness values of used alloys. Internal friction Q^{-1} and the elastic constants of used alloys were determined using the dynamic resonance method [12- 14].

3. RESULTS AND DISCUSSIONS

Microstructure

X-ray diffraction patterns of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) rapidly solidified alloys show that sharp lines of body centered tetragonal Sn and hexagonal Zn phases as presented in Figure 1. From x-ray analysis, adding In content to $SnZn_9$ alloy produced a change in its matrix microstructure (lattice parameters, unit cell volume and crystal size) and the shape of formed phases such as peak intensity, peak broadness and peak position. That is because In atoms dissolved in $Sn_{91}Zn_9$ matrix formed a solid solution\or and some In atoms formed a traces of undetected phases (In or In intermetallic phases). Also the calculated lattice parameters, (a and c), unit volume cell and crystal size of tetragonal tin phase in $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are listed in Table 1. The results illustrated that, adding In content to $Sn_{91}Zn_9$ alloy caused a little variation in lattice parameters and unit cell volume with a significant variation in crystal size of tetragonal tin phase of $Sn_{91}Zn_9$ alloy after adding In content.



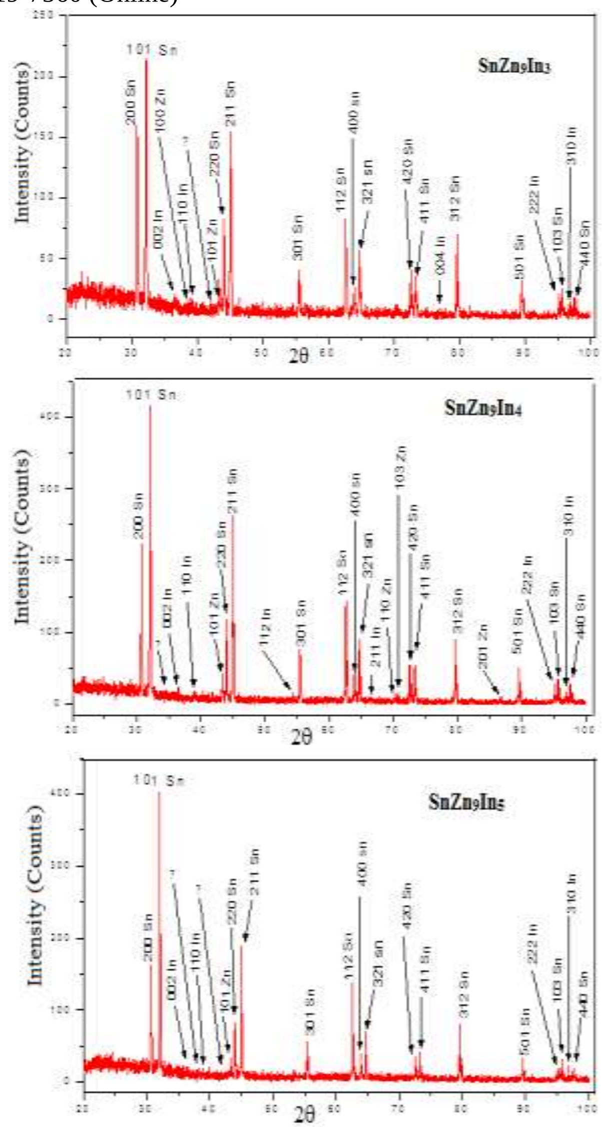


Figure 1:- x-ray diffraction patterns of $Sn_{91-x}Zn_9In_x$ alloys

Table 1:-lattice parameters, unit cell volume and crystal particle size of β-Sn in $Sn_{91-x}Zn_9In_x$ alloys

Samples	a Å	c Å	V Å³	τ Å
$SnZn_9$	5.82	3.183	107.83	681.943
$SnZn_9In_1$	5.801	3.182	107.11	454.054
$SnZn_9In_2$	5.805	3.188	107.45	465.097
$SnZn_9In_3$	5.814	3.189	107.78	707.90
$SnZn_9In_4$	5.812	3.189	107.70	593.691
$SnZn_9In_5$	5.821	3.180	107.76	658.568

Scanning electron micrographs, SEM, of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys show heterogeneous structure (different features) as seen in Figure 2 and it's agreed with x-ray analysis.

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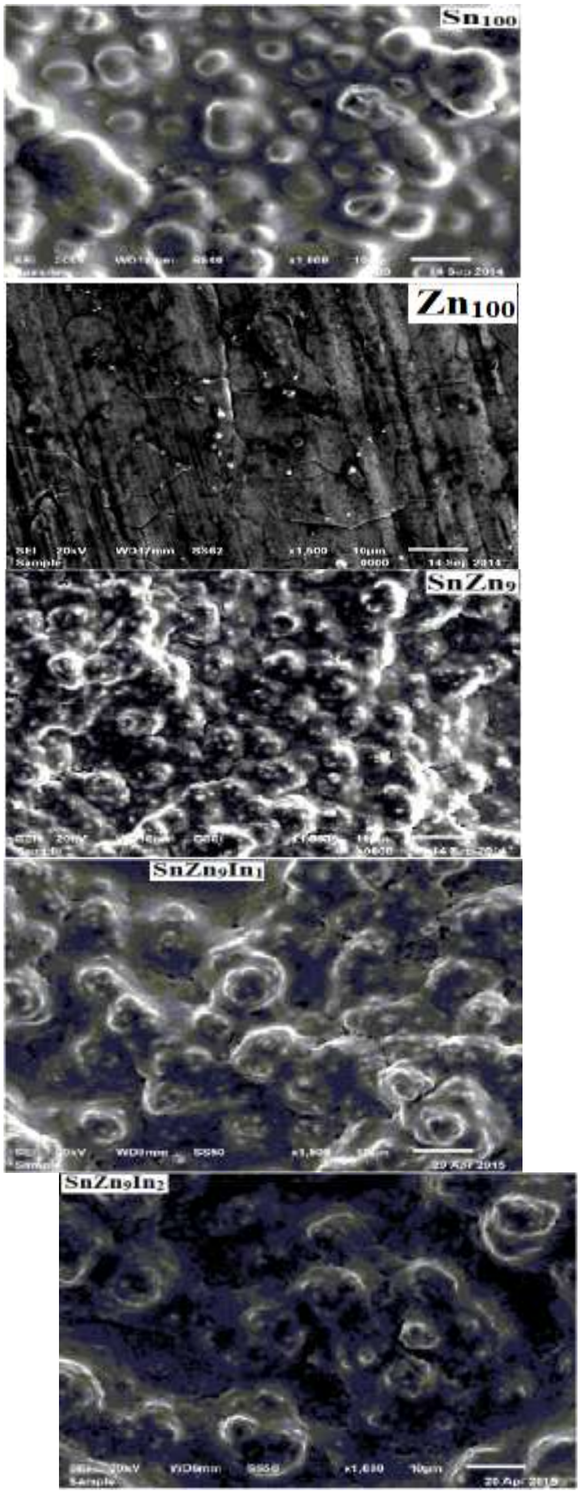


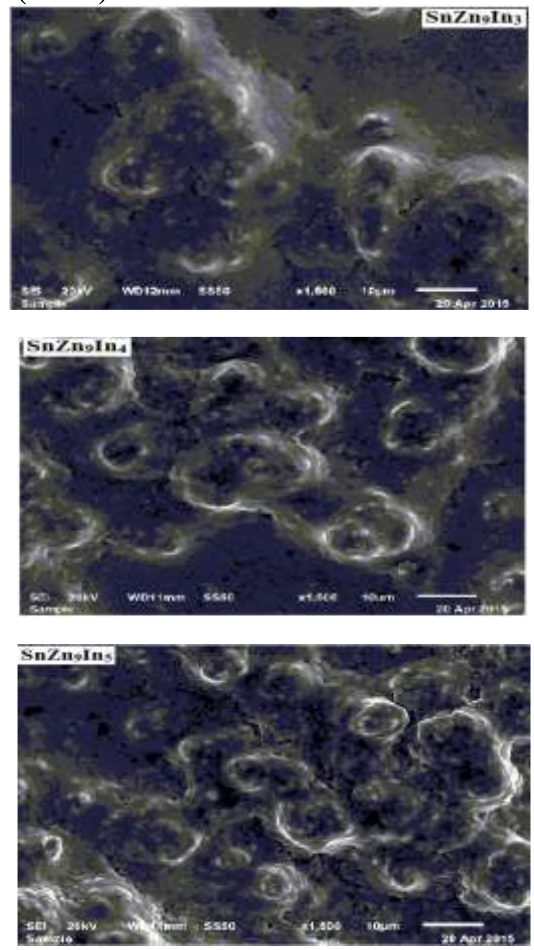


Figure 2:- SEM of $Sn_{91-x}Zn_9In_x$ alloys

Soldering properties

Wettability

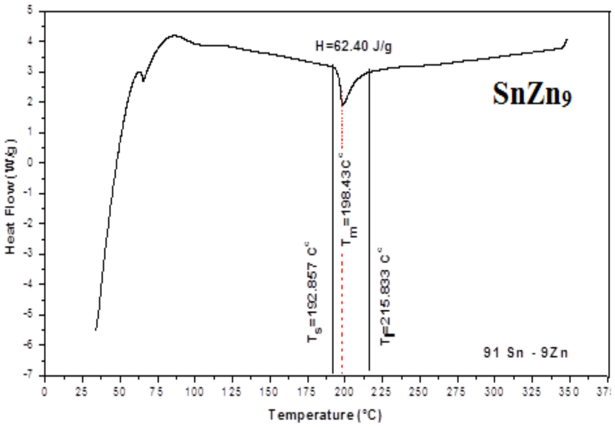
Wettability is quantitatively evaluated by the contact angle formed at the solder substrate's flux triple point. The contact angles of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys on Cu substrate in air are shown in Table 2. The results show that, the contact angle of $Sn_{91}Zn_9$ alloy is variable decreased after adding In content. The $Sn_{86}Zn_9In_5$ alloy has low contact angle value and also adequate for solder applications.

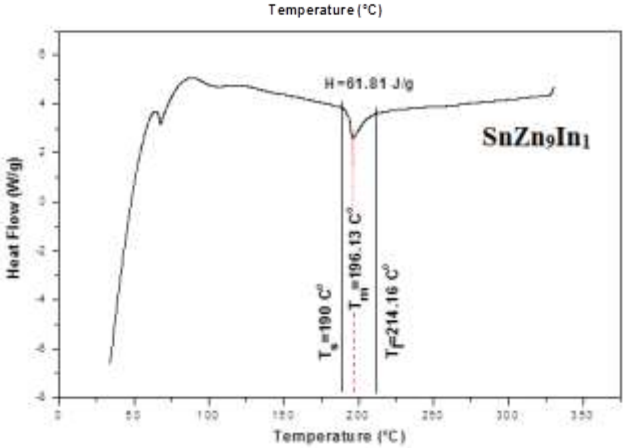
Table 2:-contact angles of $Sn_{91-x}Zn_9In_x$ alloys

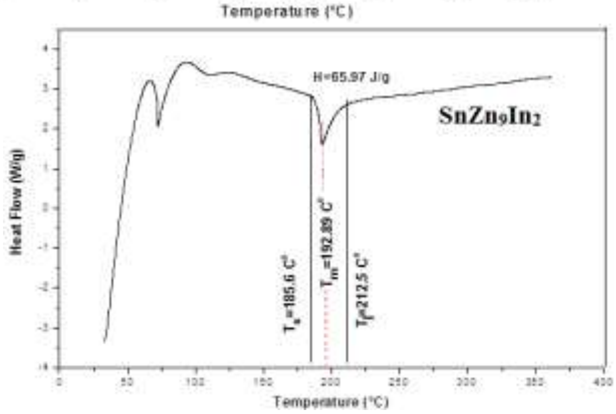
Samples	Contact angle (θ°)
$SnZn_9$	46
$SnZn_9In_1$	42.5
$SnZn_9In_2$	45.75
$SnZn_9In_3$	45
$SnZn_9In_4$	42
$SnZn_9In_5$	38.25

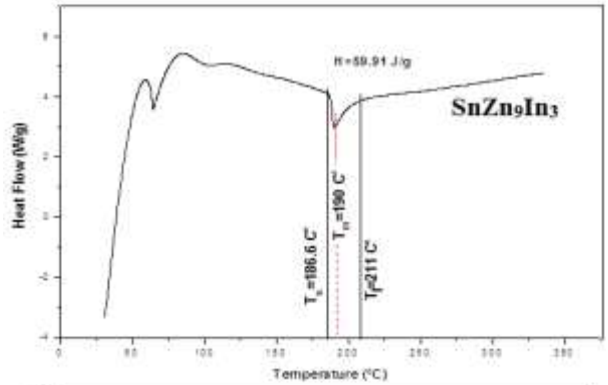
Thermal properties

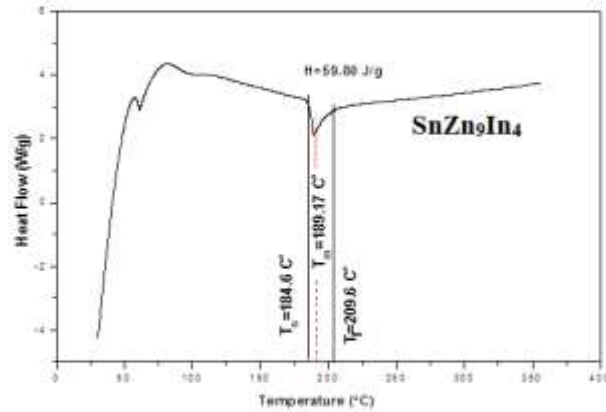
The amounts of thermal properties depend on the nature of solid phase and on its temperature. The DSC thermographs were achieved with heating rate 10 °C /min in the temperature range 0- 400 °C. The DSC thermographs of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are shown in Figure 3. From these graphs the melting point, pasty range and other thermal parameters (specific heat, C_P, enthalpy, ΔH, entropy, ΔS) of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are identified and then listed in Table 3. A little variation occurred in thermo-graph (Endo-thermal peaks) of $Sn_{91}Zn_9$ alloy after adding indium. That is because In atoms dissolved in matrix alloy changed its structure and that is agreed with x-ray diffraction analysis. The melting temperature of $Sn_{91}Zn_9$ alloy decreased after adding indium content. Also the pasty range and other thermal parameters of $Sn_{91}Zn_9$ alloy varied after adding indium content.











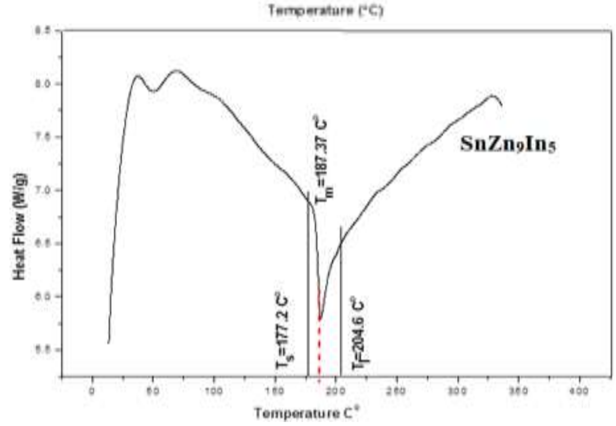


Figure 3:- DSC graphs of $Sn_{91-x}Zn_9Al_x$ alloys

Table 3:- melting point and other thermal parameters of $Sn_{91-x}Zn_9Al_x$ alloys

Samples	Melting point °K	C_P J/g. °K	Δ S J/g. °K	ΔH J/g
$SnZn_9$	471.43	2.716	130.772	62.40
$SnZn_9In_1$	469.13	2.558	130.155	61.81
$SnZn_9In_2$	465.89	2.45	139.889	65.97
$SnZn_9In_3$	463.12	2.46	127.025	59.91
$SnZn_9In_4$	462.17	2.392	127.256	59.80
$SnZn_9In_5$	460.37	2.271	151.858	71.45

Electrical resistivity

Crystalline imperfections and plastic deformation raises the electrical resistivity as a result of the increased number of electron scattering centers. The measured electrical resistivity of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys at room temperature using double bridge method are shown in Table 4. Electrical resistivity of $Sn_{91}Zn_9$ alloy variable increased after adding indium content. That is because indium atoms dissolved in the $Sn_{91}Zn_9$ matrix, formed solid solution/or and some traces, played as scattering center for conduction electrons increased electrical resistivity value.

Table 4:- electrical resistivity of $Sn_{91-x}Zn_9In_x$ alloys

Samples	$\rho x10^{-8}$ Ω.m
$SnZn_9$	33.65
$SnZn_9In_1$	35.6
$SnZn_9In_2$	39.94
$SnZn_9In_3$	40.83
$SnZn_9In_4$	38.27
$SnZn_9In_5$	39.39

Mechanical properties

Elastic moduli

The elastic constants are directly related to atomic bonding and structure. It is also related to the atomic density. The measured elastic modulus and calculated bulk modulus, B, and shear modulus, μ, of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are listed in Table 5. Elastic modulus value of $Sn_{91}Zn_9$ alloy is variable decreased after adding indium content as shown in Table 5. That is because the dissolved indium atoms, formed solid solution\ or stick on grain boundary/ or formed small cluster from phases in $Sn_{91}Zn_9$ matrix, affected on bond matrix strengthens.

Internal friction and thermal diffusivity

Internal friction is a useful tool for the study of structural aspects of alloys. Resonance curves of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are shown in Figure 4 and the calculated internal friction values are presented in Table 5. Also from resonance frequency at which the peak damping occur using the dynamic resonance method the thermal diffusivity value was calculated and then listed in Table 5. The results show that, internal friction value of $Sn_{91}Zn_9$ alloy is variable decreased by adding indium content.

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Table 5:- elastic moduli, internal friction and thermal diffusivity of $Sn_{91-x}Zn_9In_x$ alloys

Samples	E GPa	μ GPa	B GPa	Q^{-1}	D_{th} x10^{-7} m^2\sec
$SnZn_9$	44.34±3.1	16.42	49.30	0.085	4.476
$SnZn_9In_1$	30.75±1.2	11.39	34.05	0.052	3.392
$SnZn_9In_2$	29.53±1.2	10.95	32.57	0.054	2.947
$SnZn_9In_3$	33.04±2.1	12.25	36.3	0.081	3.484
$SnZn_9In_4$	32.9±2.19	12.20	35.99	0.057	2.773
$SnZn_9In_5$	30.09±1.6	11.17	32.80	0.059	2.316

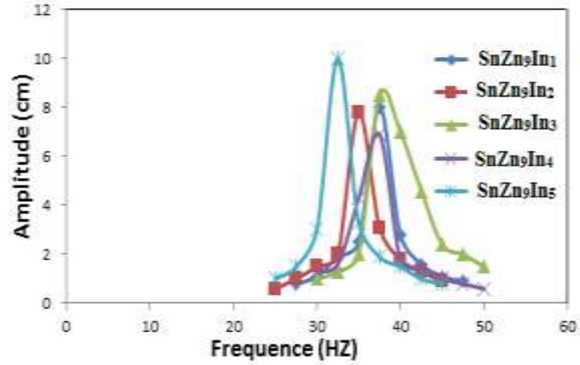

Figure 4:- resonance curves of $Sn_{91-x}Zn_9Al_x$ alloys

Vickers microhardness and minimum shear stress

The hardness is the property of material, which gives it the ability to resist being permanently deformed when a load is applied. The greater of material hardness is the greatest of the resistance to deformation. The Vickers hardness number of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys at 10 gram force and indentation time 5 sec are shown in Table 6. Also calculated minimum shear stress of $Sn_{91-x}Zn_9In_x$ (x= 0, 1, 2, 3, 4, 5 wt. %) alloys are listed in Table 6. Vickers hardness value of $Sn_{91}Zn_9$ alloy is variable increased by adding indium content.

Table 6:- Vickers hardness and minimum shear stress of $Sn_{91-x}Zn_9In_x$ alloys

Alloys	H_v kg/mm^2	μ_n kg/mm^2
$SnZn_9$	22.75±1.1	7.508
$SnZn_9In_1$	23.53±1	7.766
$SnZn_9In_2$	27.8±1.23	9.174
$SnZn_9In_3$	30.7±1.9	10.131
$SnZn_9In_4$	26.37±1.2	8.701
$SnZn_9In_5$	36.82±2.5	12.149

4. CONCLUSIONS

1. Adding indium content to $SnZn_9$ alloy produced a change in its matrix microstructure (lattice parameters, unit cell volume and crystal size) and the shape of formed phases
2. The contact angle, melting temperature, elastic modulus and internal friction values of $SnZn_9$ alloy are variable decreased after adding indium content. The $SnZn_9In_5$ alloy has low contact angle and melting point values.
3. The electrical resistivity and Vickers hardness values of $SnZn_9$ alloy are variable increased after adding indium content.
4. The $SnZn_9In_5$ alloy has adequate properties for solder applications

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