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Effect of quaternary addition on structure, electrical, mechanical and thermal properties of bismuth-tin-zinc rapidly solidified fusible alloy

Abu Bakr El- Bediwi, Feryal Dawood*, Mustafa Kamal
Metal Physics Lab., Physics Department, Faculty of Science, Mansoura University, Mansoura, Egypt
baker_elbediwi@yahoo.com
*Basic education college, University of Diayala, Iraq

Abstract

Effect of quaternary addition, silver or indium, on structure, electrical, mechanical and thermal properties of bismuth-tin-zinc rapidly solidified fusible alloy have been investigated. Adding silver or indium caused change in alloy matrix microstructure such as matrix parameters and crystal structure of formed phase. A significant increase in bismuth-tin-zinc alloy strengthens with a little decreased in melting point after adding silver content. But a significant decrease in bismuth-tin-zinc alloy melting point with a very little increase in alloy strengthens after adding indium content.

Keywords

Structure; electrical resistivity; elastic modulus; melting point hardness; fusible alloy

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Introduction

An alloy may be defined as a substance that has the metallic properties and is composed of two or more chemical elements of which at least one is a metal. Most alloys are of great importance in industry and in the arts, in fact far more so, when quantitatively considered, than are the pure metals. Fusible alloys are one such category of materials, which have attracted the attention of scientists and technologists all over the world. Structure, wettability, melting point, electrical and mechanical properties of Sn-Zn-Bi-Cu-In, Sn-In, Sn-In-Ag, Sn-Zn-Ag-In, Sn-Zn-In, Bi- Sn, Sn-Bi- In and Sn-Ag lead free solder alloys have been investigated [1-6]. The results show that, these alloys have required properties for solder applications. The Sn-3.5%Ag-1%Zn with superior mechanical properties [7] and Sn-Zn-In based alloys with lower melting that are sufficiently similar as to serve as a drop-in replacement for the eutectic Pb-Sn solder [8]. The aim of present work is to investigate the effect of quaternary addition on structure and physical properties

Experimental work

In this work two groups of quaternary fusible alloys, bismuth-tin-zinc-sliver and bismuth-tin-zinc-indium, were used. These groups' alloys were molten in the muffle furnace using high purity, more than 99.95%, bismuth, tin, zinc, silver and indium. 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 ~ 70 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 0 to 100° in continuous mode with a scan speed 5 deg/min. Electrical resistivity of used alloys was measured by double bridge method. 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 [9-11].

Results and discussions

Structure

X-ray diffraction patterns of $Bi_{71-x}Sn_{25}Zn_4Ag_x$(x= 0, 0.5, 1.5, 2.5 and3.5 wt. %) and $Bi_{71-x}Sn_{25}Zn_4In_x$(x= 0, 2, 4, 6, 8 and 10 wt. %) rapidly solidified fusible alloys are shown in Figure 1. X-ray diffraction analysis show that, $Bi_{71}Sn_{25}Zn_4$ alloy consisted of β- Sn and hexagonal Bi phases. That is meant that, Zn dissolved in alloy matrix. $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys consisted of β- Sn and hexagonal Bi phases. That is meant that, Ag or In dissolved in alloy matrix.From these analysis it obvious that, adding Ag or In content to $Bi_{71}Sn_{25}Zn_4$alloy caused a change in its matrix microstructure such as lattice parameters and crystalshape of formed phases,(crystallinity which is related to intensity of the peak, crystal size which is related to full width half maximum and the orientation which is related to the position of the peak, 2θ).

Lattice parameters, (a and c), and unit volume cell of β- Sn phase in $Bi_{71-x}Sn_{25}Zn_4Ag_x$ (x= 0, 0.5, 1.5, 2.5 and 3.5 wt. %) and $Bi_{71-x}Sn_{25}Zn_4In_x$ (x= 0, 2, 4, 6, 8 and 10 wt. %) rapidly solidified fusible alloys were determined from equations (1) and (2) and then listed in Table 1.

$$\frac{1}{d^2} = \frac{(h^2+k^2)}{a^2} + \frac{l^2}{c^2} \qquad (1)$$

$$V = a^2c \qquad (2)$$

Lattice parameters and unit volume of β- Sn changed after adding Ag or In content to $Bi_{71}Sn_{25}Zn_4$alloy. That is because Ag or In atoms dissolved in matrix alloy.

Table 1:- lattice parameters and unit volume cell of β- Sn phase

Samples	aÅ	c Å	Unit cell volume $Å^3$
$Bi_{71}Sn_{25}Zn_4$	4.754	11.888	71.035
$Bi_{69}Sn_{25}Zn_4In_2$	4.756	11.8878	71.213
$Bi_{67}Sn_{25}Zn_4In_4$	4.756	11.891	71.987
$Bi_{65}Sn_{25}Zn_4In_6$	4.756	11.891	71.114
$Bi_{63}Sn_{25}Zn_4In_8$	4.753	11.887	70.988
$Bi_{61}Sn_{25}Zn_4In_{10}$	4.754	11.885	71.108

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Samples	a Å	c Å	Unit cell volume Å^3
$Bi_{71}Sn_{25}Zn_4$	4.754	11.888	71.035
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	4.751	11.874	71.003
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	4.755	11.891	71.022
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	4.752	11.885	70.929
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	4.754	11.888	70.979

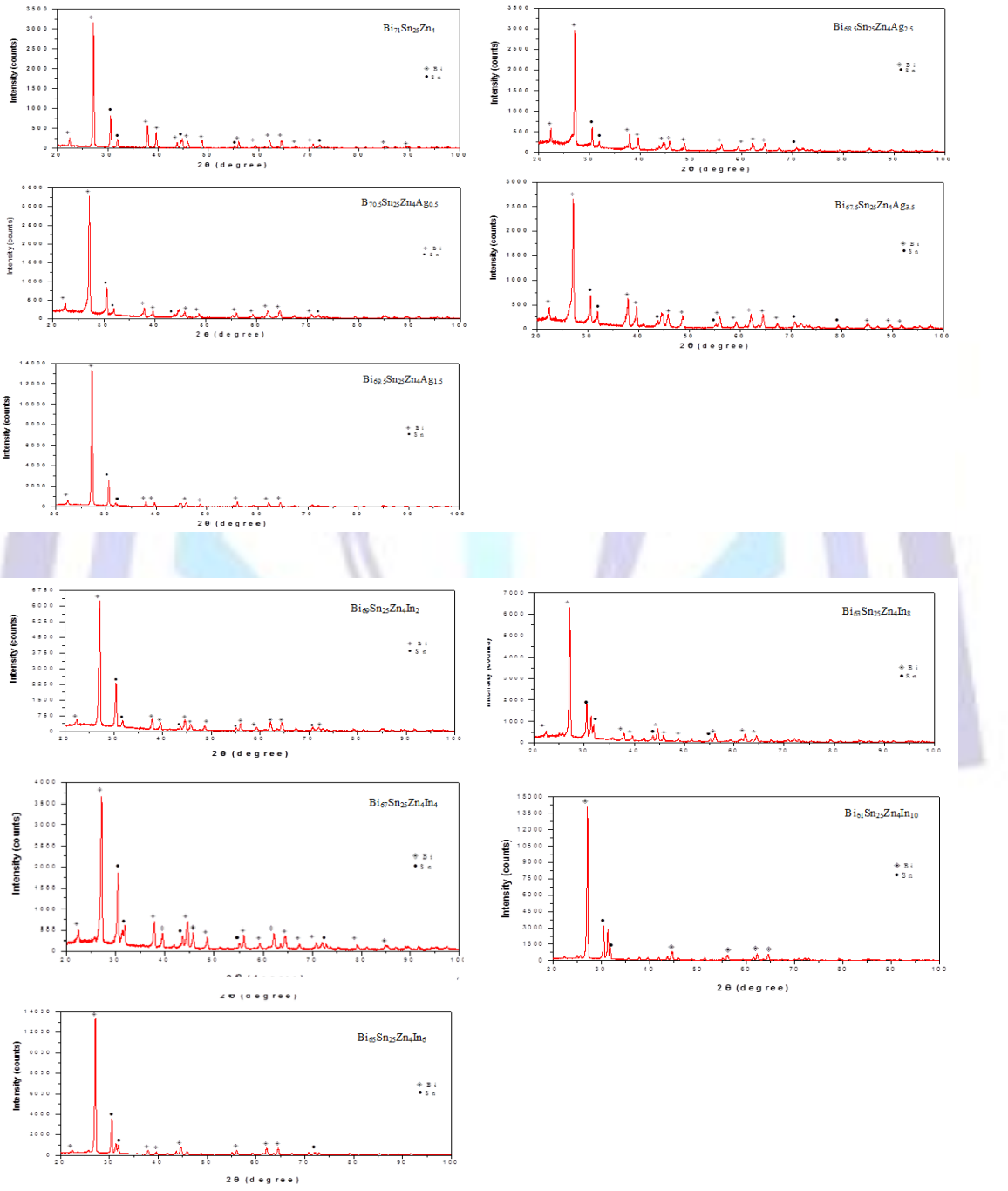

Figure 1:- x-ray diffraction patterns of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

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Thermal properties

Thermal analysis is often used to study solid state transformations as well as solid-liquid reactions. DSC thermographs were obtained by SDT Q600 (V20.9 Build 20) with heating rate 10 °C /min in the temperature range 0-400 °C. Figure 2 shows the DSC thermographs for $Bi_{71-x}Sn_{25}Zn_4Ag_x$ (x= 0, 0.5, 1.5, 2.5 and 3.5 wt. %) and $Bi_{71-x}Sn_{25}Zn_4In_x$ (x= 0, 2, 4, 6, 8 and 10 wt. %) alloys. From these thermographs, a little variation in the exo-thermal peaks shape. That means there is a change in alloy matrix after adding Ag or In content.The melting point and other thermal properties of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$alloys are shown in Table 2. A little decreased in $Bi_{71}Sn_{25}Zn_4$ alloy melting point after adding Ag content but a significant decreased after adding In content. The pasty range is the difference between solidus and liquidus points. The pasty range of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$alloys are listed in Table 2. The $Bi_{61}Sn_{25}Zn_4In_{10}$ alloy has lower melting point.

Table 2:- melting point and other thermal properties of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

Samples	Melting point ºC	Pasty rang ºC	C_P J/g. C	S Δ J/g. C
$Bi_{71}Sn_{25}Zn_4$	142.25	12.44	1.5	0.128
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	136.25	7.49	2.09	0.112
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	140.25	7.41	2.18	0.112
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	139.78	7.1	2.68	0.133
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	139.23	7.96	2.003	0.111

Samples	Melting point ºC	Pasty rang ºC	C_P J/g. C	S Δ J/g. C
$Bi_{71}Sn_{25}Zn_4$	142.25	12.44	1.5	0.128
$Bi_{69}Sn_{25}Zn_4In_2$	127.69	9.03	1.25	0.085
$Bi_{67}Sn_{25}Zn_4In_4$	121.65	11.15	0.75	066
$Bi_{65}Sn_{25}Zn_4In_6$	116.91	8.09	0.78	0.052
$Bi_{63}Sn_{25}Zn_4In_8$	110.93	8.59	0.475	0.035
$Bi_{61}Sn_{25}Zn_4In_{10}$	106.76	7.3	0.208	0.014

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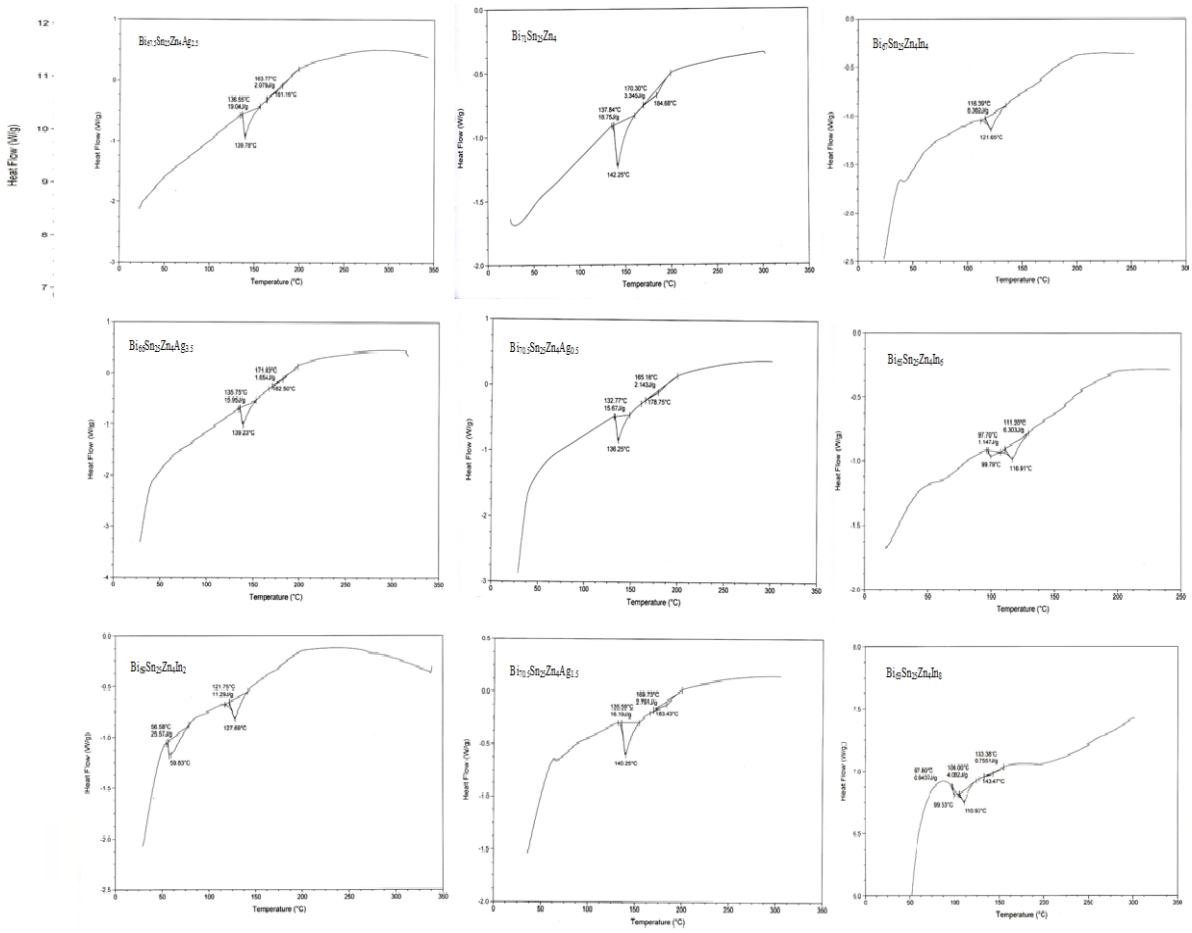

Figure 2:- DSC thermographs for $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

Wettability

Now wettability is a center of attention in nanotechnology and nanoscience studies due to the advent of many nanomaterials in the past 2 decades. Wetting is the ability of a liquid to maintain contact with a solid surface, resulting from intermolecular interactions when the two are brought together.Low contact angle, less than 90°, usually indicates that wetting of the surface is very favorable, and the fluid will spread over a large area of the surface but high contact angle, greater than 90°, generally means that wetting of the surface is unfavorable so the fluid will minimize contact with the surface and form a compact liquid droplet. The contact angles of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys on Cu substrate are shown in Table 3. The contact angle value of $Bi_{71}Sn_{25}Zn_4$ alloy varied after adding Ag or In content.

Table 3:- contact angle of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and$Bi_{71-}Sn_{25}Zn_4In_x$alloys

Samples	θº
$Bi_{71}Sn_{25}Zn_4$	50.5
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	64
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	46.5
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	50
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	69

Samples	θº
$Bi_{71}Sn_{25}Zn_4$	50.5
$Bi_{69}Sn_{25}Zn_4In_2$	73.5
$Bi_{67}Sn_{25}Zn_4In_4$	68
$Bi_{65}Sn_{25}Zn_4In_6$	72
$Bi_{63}Sn_{25}Zn_4In_8$	71

Electrical resistivity and thermal conductivity

In general, the plastic deformation raises the electrical resistivity as a result of the increased number of electron scattering centers. Crystalline defects serve as scattering center for conduction electrons in metals, so the increase in their number raises the imperfection. Electrical resistivity and calculated thermal conductivity values of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys are shown in Table 4. Electrical resistivity value of $Bi_{71}Sn_{25}Zn_4$ alloy increased after adding Ag

or In content. That is because Ag or In atoms dissolved in the alloy matrix playing as scattering center for conduction electrons. Thermal conductivity value of $Bi_{71}Sn_{25}Zn_4$ alloy decreased after adding Ag or In content.

Table 4:-electrical resistivity and thermal conductivity of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

Samples	ρx10^{-8}Ω.m	K (W.m^{-1}.K^{-1})
$Bi_{71}Sn_{25}Zn_4$	184.02±4.7	4.01
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	189.49	3.895
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	247.3	2.984
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	258.27	2.857
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	282.01	2.617

Samples	ρx10^{-8}Ω.m	K (W.m^{-1}.K^{-1})
$Bi_{71}Sn_{25}Zn_4$	184.02±4.7	4.01
$Bi_{69}Sn_{25}Zn_4In_2$	196.45±15.7	3.756
$Bi_{67}Sn_{25}Zn_4In_4$	198.39±19.5	3.720
$Bi_{65}Sn_{25}Zn_4In_6$	213.94±4.81	3.449
$Bi_{63}Sn_{25}Zn_4In_8$	221.08±20.23	3.338
$Bi_{61}Sn_{25}Zn_4In_{10}$	246.74±5.07	2.991

Elastic properties

The elastic constants are directly related to atomic bonding and structure. Also it is related to the atomic density. Elastic modului valuesof $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys are listed in Table 5. A little decreased in elastic modulus of $Bi_{71}Sn_{25}Zn_4$ alloy after adding In content but a significant increased after adding Ag content.

Calculated internal friction and thermal diffusivity values of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys are seen in Table 5. Internal friction value of $Bi_{71}Sn_{25}Zn_4$ alloy decreased after adding Ag or In content.

Table 5:- elastic modului, internal friction and thermal diffusivity values of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

Samples	E GPa	B GPa	μ GPa	Q^{-1}x10^{-3}	D_{th}x10^{-8} (m^2/sec)
$Bi_{71}Sn_{25}Zn_4$	34.94±5.7	35.167	13.1	84.41±5.46	11.587
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	39.87	40.15	14.94	53.71±3.26	4.81
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	41.45	41.84	15.53	38.07±1.46	26.64
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	44.51	45.04	16.67	75.09±4.41	1.74
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	54.05	54.83	20.23	29.15±1.24	31.44

Samples	E GPa	B GPa	μ GPa	Q^{-1}x10^{-3}	D_{th}x10^{-8} (m^2/sec)
$Bi_{71}Sn_{25}Zn_4$	34.94±5.7	35.167	13.1	84.41±5.46	11.587
$Bi_{69}Sn_{25}Zn_4In_2$	33.74±3.25	33.82	12.65	55±3.24	6.8
$Bi_{67}Sn_{25}Zn_4In_4$	32.95±3.15	32.99	12.37	47.06±3	19.95
$Bi_{65}Sn_{25}Zn_4In_6$	32.19±1.24	32.35	12.14	42.3±2.4	33.71
$Bi_{63}Sn_{25}Zn_4In_8$	30.79±1.8	31.05	11.66	79.34±4	23.79
$Bi_{61}Sn_{25}Zn_4In_{10}$	30.19±3.2	30.56	11.48	72.15±3.85	30.70

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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. Vickers hardness of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys at 10 gram force and indentation time 5 sec are shown in Table 6. The minimum shear stress (τ_m) value of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys was calculated using the equation (3) [10], where ν is Poisson's ratio of the elements in the alloy and then listed in Table 6.

$$\tau_m = \frac{1}{2}H_v\left\{\frac{1}{2}(1-2\nu)+\frac{2}{9}(1+\nu)[2(1+\nu)]^{\frac{1}{2}}\right\} \qquad (3)$$

Vickers hardness value of $Bi_{71}Sn_{25}Zn_4$ alloy increased after adding Ag or In content.

Conclusion

1. Adding Ag or In content to $Bi_{71}Sn_{25}Zn_4$ alloy caused a change in its matrix microstructure such as lattice parameters and crystal shape of formed phases.
2. A little decreased in elastic modulus of $Bi_{71}Sn_{25}Zn_4$ alloy after adding In content but a significant increased after adding Ag content.
3. Electrical resistivity value of $Bi_{71}Sn_{25}Zn_4$ alloy increased after adding Ag or In content.
4. A little decreased in melting point of $Bi_{71}Sn_{25}Zn_4$ alloy after adding Ag content but a significant decreased after adding In content.
5. Vickers hardness value of $Bi_{71}Sn_{25}Zn_4$ alloyincreased after adding Ag or In content.

Table 6:- Vickers hardness and minimum shear stress of $Bi_{71-x}Sn_{25}Zn_4Ag_x$ and $Bi_{71-x}Sn_{25}Zn_4In_x$ alloys

Samples	H_V (Kg/mm^2)	μ_n (Kg/mm^2)
$Bi_{71}Sn_{25}Zn_4$	14.43±0.327	4.763
$Bi_{70.5}Sn_{25}Zn_4Ag_{0.5}$	14.4±0.22	4.75
$Bi_{69.5}Sn_{25}Zn_4Ag_{1.5}$	18.78±0.4	6.2
$Bi_{68.5}Sn_{25}Zn_4Ag_{2.5}$	23.78±1.4	7.85
$Bi_{67.5}Sn_{25}Zn_4Ag_{3.5}$	24.83±1.57	8.19

Samples	H_V (Kg/mm^2)	μ_n (Kg/mm^2)
$Bi_{71}Sn_{25}Zn_4$	14.43±0.33	4.763
$Bi_{69}Sn_{25}Zn_4In_2$	14.38±0.48	4.75
$Bi_{67}Sn_{25}Zn_4In_4$	18.8±0.68	6.20
$Bi_{65}Sn_{25}Zn_4In_6$	15.6±0.69	5.15
$Bi_{63}Sn_{25}Zn_4In_8$	14.48±0.29	4.78
$Bi_{61}Sn_{25}Zn_4In_{10}$	17.5±1.42	5.78

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