



Mechanical Properties of the (81Sn–xCu–(19-x) In) % Alloys with x=1, 2, 3, 4, and 5

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ABSTRACT: Research on Material Science has a long tradition, and lead has been used widely in many applications. Because of their low melting temperatures, low cost, and good wettability, tin -lead alloys have been preferred as solder alloys in industrial production, particularly in the electronics sector. However, due to the negative effects of lead on the environment and human health, studies on lead-free solder alloys have been increasing, and the subject is still important today. Furthermore, environmental protection agencies (RoHS and WEEE) have prohibited the use of lead-containing solder alloys in the manufacture of electronic and microelectronic products. In this work, the effect of adding Cu to alloys in five compositions of (81Sn-xCu-(19-x)In) % with x=1, 2, 3, 4, and 5 on the mechanical properties has been studied. The five samples were prepared from high purity (99%) elements (Sn –Cu –In) using the melting technique. The creep of the ternary alloys was examined at different temperatures (25, 50, and 80°C) under three different loads (2.5, 6.3, and 11.4 MPa). It was found that the creep rate of the ternary alloys was increasing with the increases in Cu –content in all samples at different temperatures and loads. The mean values of the stress exponent were found to be in the range of (0.1-2.5) as well as the activation energies varied in the range of (0.48-5.33) eV for all alloys under different loads (2.5, 6.3, and 11.4 MPa).

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1. INTRODUCTION

The alloy is a macroscopically uniform material with metallic properties; it contains two or more metals and nonmetals. Only the composition of the alloy and the equilibrium temperature specify whether it is a single phase or a mixture

of phases. Alloys can be categorized in an array of ways based on their compositions and distinguishing characteristics to; best metals, joining alloys and soldering alloys. Solder alloys might be lead-tin solders or lead-free solders [1]. Lead-free solders are likely to be tin-rich alloys with alloying elements such as Bi, Ag, Cu, In,

Zn, Sb, etc. A large number of topics of Pb-free solder alloys have been proposed. All these solder alloys were developed from Sn-based binary alloys [2]. The soldering process has been a critical component in the creation of all electronic devices since the dawn of the electronic age, and it is expected to remain the primary assembly and linking technology for some time to come [3]. In the manufacturing of electronics, solder joints are utilized to connect electrical components and printed circuit boards. Traditionally, Sn-Pb solder was extensively used in the electronic packaging industry due to its superior performance. However, due to the growing concern for environmental protection and human health, the toxicity of lead has received a lot of attention [4,5]. Environmental and health concerns prompted the elimination of SnPb solder from all electronic applications in so many countries. In this environment, the interest in developing lead-free solder alloys has grown [6, 7]. Lead-free solders such as Sn-58Bi, Sn-52In and Sn-3.5Ag might be used to replace Sn-37Pb solder in low-cost electronic assembly [8]. Recent studies show that room temperature has a significant impact on the mechanical properties of solders [9]. Sn-0.7Cu solders are the least expensive and most promising new Pb-free alloy candidates [4].

Sn-In and Sn-Ag solder alloys are already in use, but only under certain conditions. Considering this, it is probable that the Ag-In-Sn ternary system would be one of the appropriate alternatives to traditional solders [10].

Sn-In alloying solders are clearly stronger than Pb-In (50 Pb/50 In) solders at low temperatures [7, 11]. Also, it was noticed that, the addition of In element had a significant effect on the onset point of the Sn-58Bi solder, which implied that the addition of 2 wt.% In could decrease the solder's onset point from 139.060°C to 129.680°C and when 2 wt.% In and 2 wt.% Ag were added at the same time, the melting temperature of the

composite solder fell from 143.750°C to 133.590°C [7].

2. Materials and Methods

The compositions of $(81\text{Sn} - x \text{Cu} - (19-x)\text{In}) \%$ where, $(x=1, 2, 3, 4, \text{ and } 5)$ are prepared from pure metals of (99%)(Sn, Cu, In) supplied by the (Basic Material Company in the UK).

At first, the pure elements of each composition were weighted, then mixed together homogeneously in a Pyrex tube. The, mixtures were then carried out for 4hrs at 700°C, then left to cool slowly to room temperature in order to obtain samples containing the fully precipitated phases. After that, all Pyrex tubes were broken to obtain the samples.

Secondly, all samples were polished using grades of silicon paper and then washed in a solution of $(\text{CH}_3\text{COOH}_3)$, after that, the samples were drawn using the rolling mechanism technique; the first group was as wires of a diameter $(D=1\text{mm})$ and a length $(L=70\text{Cm})$ for Creep properties, Finally, the samples were washed again in a solution of $(\text{CH}_3\text{COOH}_3)$ in order to remove residual stress and defects induced during specimen formation [12,13,14].

3. Results and Discussion

3.1 Mechanical Properties:

3.2.1 Creep Strain – Time

The creep behavior of solder alloys is an important input for accurate materials [16]. This section summarises and discusses the main findings of our work. The mechanical properties of the five alloys were determined by the temperatures $(25, 50, \text{ and } 80^\circ\text{C})$ [17]. However, the increase in Cu content, will increase the melting temperature significantly, which directly affects the soldering temperature [4]. Creep tests were carried out with the materials studied.

Figure. 1 (a, b, c, d, e, f, j, h, and k) shows the creep strain-time, the curves of $(81\text{Sn}-x\text{Cu}-(19-x)$

In) %, (x = 1, 2, 3, 4, and 5) alloys, represent average results of the carried out tensile tests. It can be observed that Cu content can significantly change the tensile properties of the (81Sn-xCu-(19-x) In) % alloy with (x=1, 2, 3, 4, and 5) [5]. The samples were studied after annealing (T=700 °C) and testing at temperatures of (T= 25, 50, and 80°C) under three different constant loads (2.5, 6.3, and 11.4 MPa).

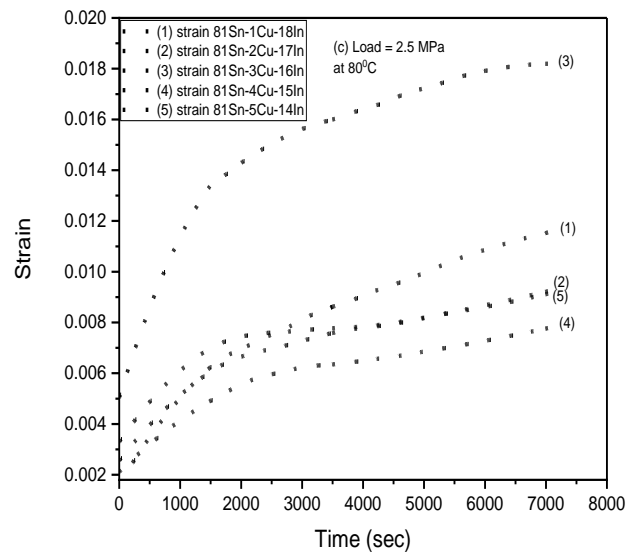
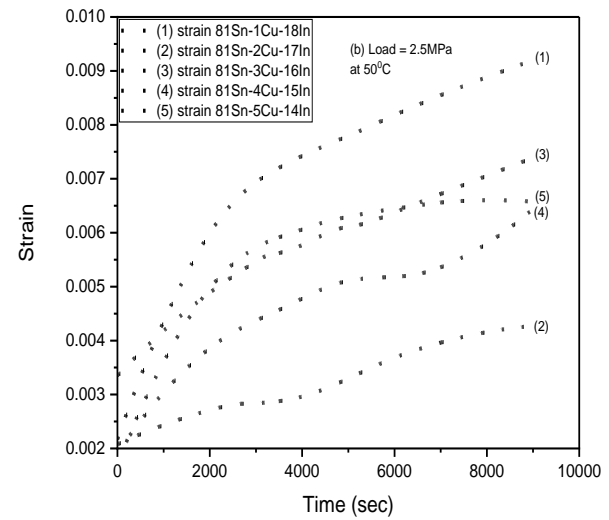
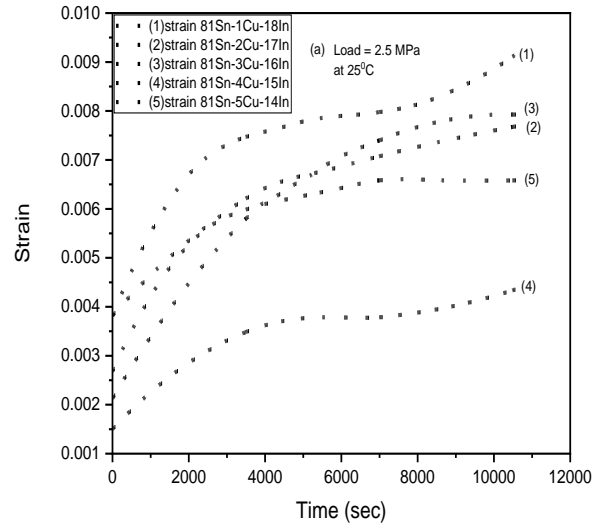
From the experimental results, it can be observed that the curves in Figure.1 (a, d, and i), represent the creep time and resistance of the 81Sn-xCu-In alloy with a 2.5 MPa load at room temperature, are longer compared to the other loads (6.3, and 11.4 MPa).

Figure.1 (b, e, and h) shows that the creep time and resistance of (81Sn-xCu-(19-x) In) %, with (x= 1,2,3,4 and 5) alloys with 11.4 MPa load are lower compared to the other loads (2.5 and 6.3MPa).

In general, the creep resistance of (81Sn-xCu-(19-x) In) %, with (x= 1, 2, 3, 4, and 5) alloys was high, and that may be due to the refinement of the structure.

The effect of ternary alloying additives on the mechanical properties of an alloy can be seen from the strain–time curves shown in Figure. 1 [18]. The creep behavior of (81Sn-xCu-(19-x)In)% alloys increases with deformation temperature and applied stress, which is in agreement with the study of Saad et. al. [19].

The creep strain rate (S^{-1}) of the (81Sn-xCu-(19-x) In) % alloys was calculated using the slope between strain-time curves in the secondary stage, as shown in Figure. 1 (a to k). And from Figure. 1 (a, b, i, f, and k) for the five Alloys, only the third stage of creep is shown, while Figure. 1 (c, d, e, and h) only shows the first two stages of creep.



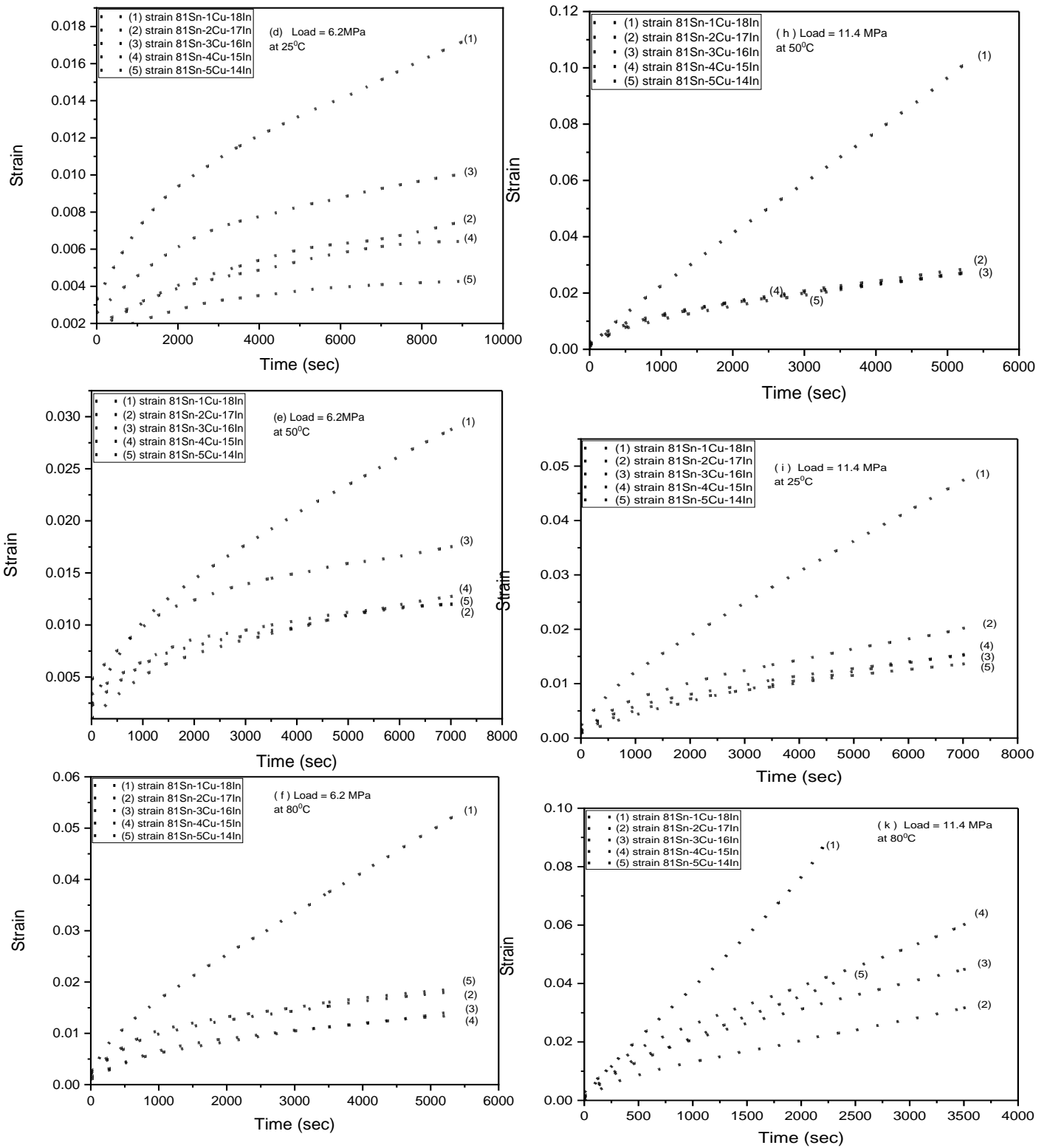


Figure. 1 (a, b, c, d, e, f, h, I, and k) : Strain–time for the five alloys at different loads (2.5, 6.3, and 11.4MPa) with different temperatures (25, 50, and 80°C).

From Table (1) and Figure. 2 below, with a small amount of Cu to (81Sn-xCu-(19-x)In)% with (x= 1, 2, 3, 4, and 5), it was observed that the secondary creep rate (strain rate ($\dot{\epsilon}$)) decreases while the creep life increases at this stage at different

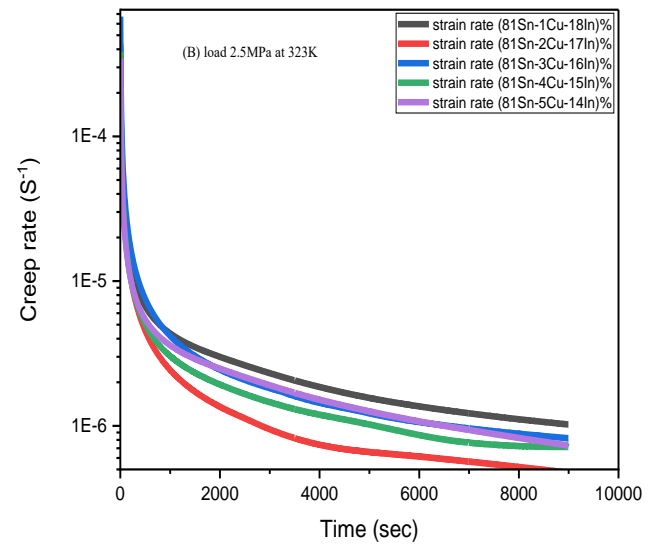
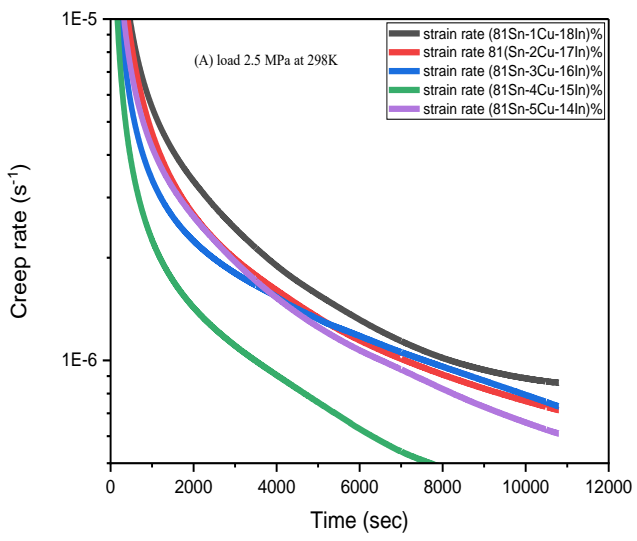
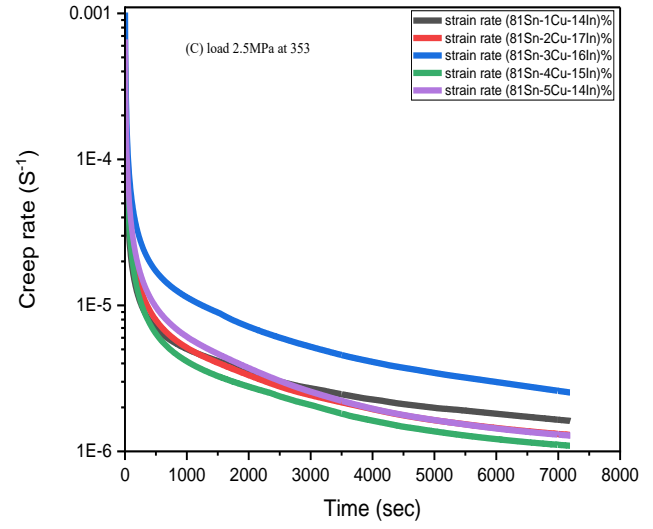
temperatures and loads. It may be due to its large lattice distortion. The increasing of creep rate (S^{-1}) of the (81Sn-xCu-(19-x) In) % alloys with Cu additions is attributed to the appearance of (111)

Cu (Cubic), which is basically returned to their different crystal structures.

Sample in wt.%	Creep rate(S ⁻¹)								
	2.5MPa			6.3MPa			11.4MPa		
Tem.	298K	323K	353K	298K	323K	353K	298K	323K	353K
81Sn - 1Cu - 18In	1.52E-07	3.80E-07	9.18E-07	1.02E-06	2.83E-06	8.05E-06	5.73E-06	1.83E-05	3.90E-05
81Sn - 2Cu - 17In	2.23E-07	3.06E-07	4.64E-07	4.09E-07	8.85E-07	1.75E-06	1.99E-06	3.55E-06	7.33E-06
81Sn - 3Cu - 16In	3.94E-07	3.11E-07	7.77E-07	5.02E-07	9.07E-07	1.47E-06	1.72E-06	3.19E-06	9.50E-06
81Sn - 4Cu - 15In	7.87E-08	2.29E-07	3.63E-07	4.06E-07	1.12E-06	1.60E-06	1.50E-06	3.19E-06	1.34E-05
81Sn - 5Cu - 14In	1.27E-07	1.68E-07	3.12E-07	1.02E-09	7.25E-07	1.72E-06	1.37E-06	3.66E-06	1.42E-05

Table (1): Creep rate(S-1) for the five alloys at different temperatures (25, 50, and 80°C) at different loads

Typical creep rate curves of (81Sn-xCu-(19-x) In) % with (x=1, 2, 3, 4, and 5) alloys are presented in Figure. 2 from (A-I). The materials showed secondary creep immediately after loading with very little primary creep. It appears that, the hardening of the matrix with the creep strain was recovered immediately. These curves illustrate the change in creep rate that occurs during the creep test. This variation is the result of changes in the internal structure, especially in the variation of the dislocation movement. From Figure. 2 (G, -I) and Table (1) above, it can be observed that, secondary creep increases with high temperature and load 11.4MPa. Also, it was noticed that all alloys showed only the two creep stages.



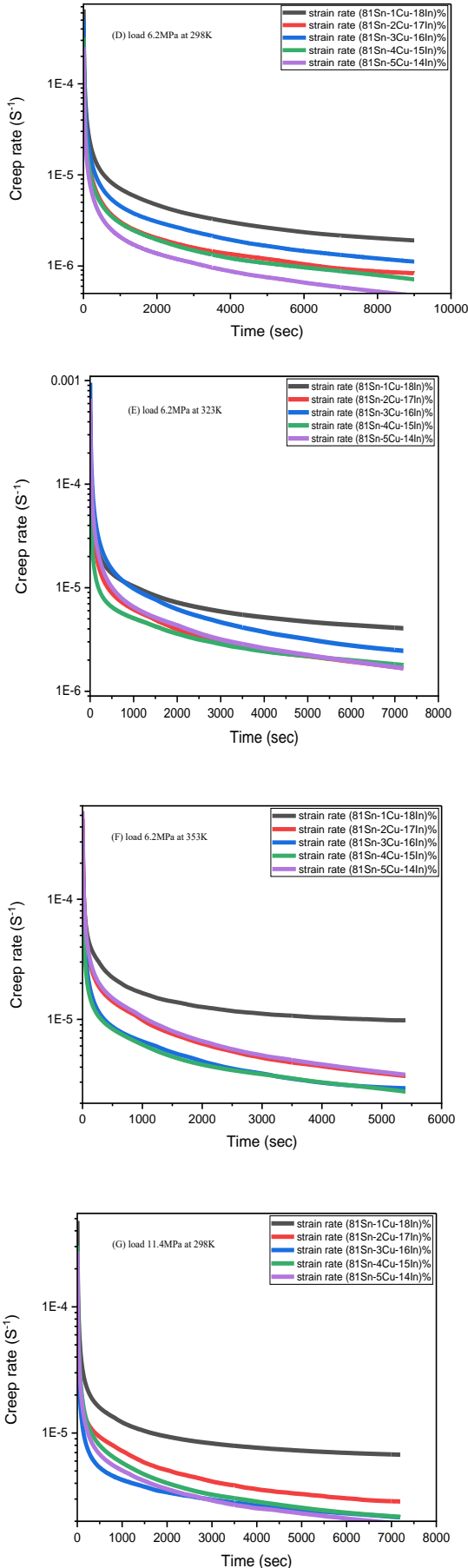


Figure. 2 (A, B, C, D, E, F, G, H, and I): Creep rate – time for (81Sn – xCu – (19-x)In)% at
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different loads (2.5, 6.3, and 11.4 MPa) with different temperatures (25, 50, and 80°C). We observed from Figure. 3 the relation between the creep rate (s^{-1}) and Cu – content (wt.%), that Cu – additions have improved the creep rate for all alloy systems with variant conditions. The minimum creep rate values were calculated using the slope ($\dot{\epsilon} = \frac{\partial \epsilon}{\partial t}$) of the secondary region in the strain–time curves Figure. 1.

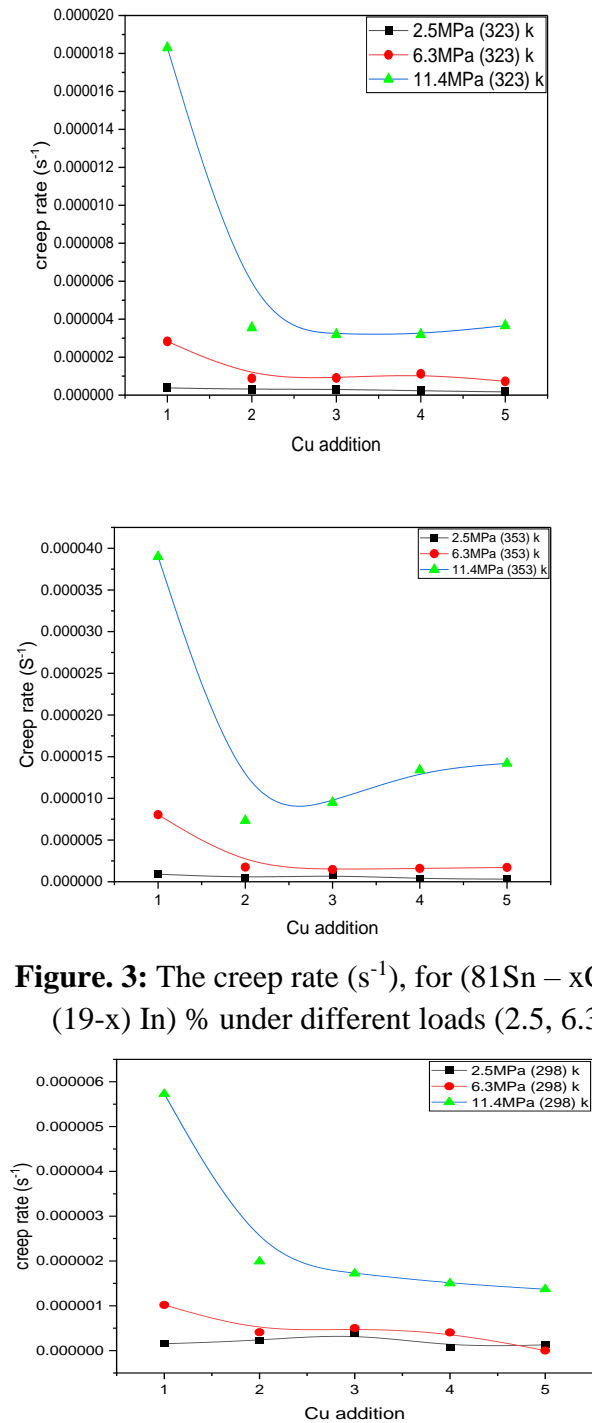


Figure. 3: The creep rate (s^{-1}), for (81Sn – xCu – (19-x) In) % under different loads (2.5, 6.3,

and 11.4 MPa) with Cu (wt.%) additions.

3.2.2 The Elongations: -

Figure. 4 (a, b, c) showed the results of Elongation (EL.%), which were calculated using the following relation:

$$EL\% = Strain \times 100 \dots (1)$$

The EL.% plots are drawn in Figure. 4 and their values are listed in Table (2) for the five alloys.

The elongation of the (81Sn-xCu-(x-19) In) % alloy at (25, 50, and 80°C) with 2.5, 6.3, and 11.4 MPa are shown in Figure. 4. The EL.% of (81Sn-xCu-(x-19) In) % alloys at T (25, 50, and 80°C) under 6.3, and 11.4 MPa are increasing compared to the EL.% of (81Sn-xCu-(x-19) In) % under 2.5 MPa, as shown below. Also, it was observed that the elongation decreased with the addition of Cu content to the alloy composition.

Table (2): The variation of elongation of three samples with temperatures and Cu addition

Sample in wt.%	EL.%								
	(2.5MPa)			(6.3MPa)			(11.4MPa)		
Tem.	298K	323K	353K	298K	323K	353K	298K	323K	353K
81Sn - 1Cu - 18In	4.465	4.7	5.875	8.46	14.805	26.32	24.44	52.405	43.475
81Sn - 2Cu - 17In	3.76	2.115	4.7	3.76	6.345	9.165	10.34	14.335	16.215
81Sn - 3Cu - 16In	3.29	3.055	7.285	3.995	7.05	5.64	6.345	4.7	18.565
81Sn - 4Cu - 15In	2.115	3.055	3.995	3.29	6.345	7.05	7.755	13.63	31.02
81Sn - 5Cu - 14In	3.29	3.29	4.465	2.115	6.345	9.635	7.05	14.1	21.15

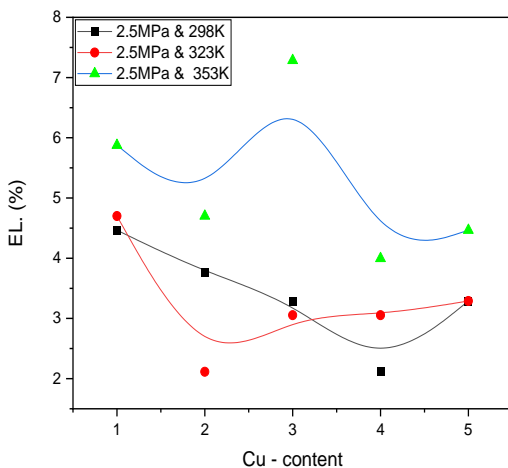
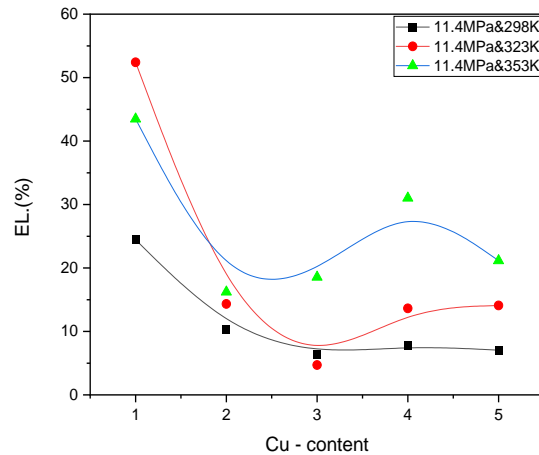
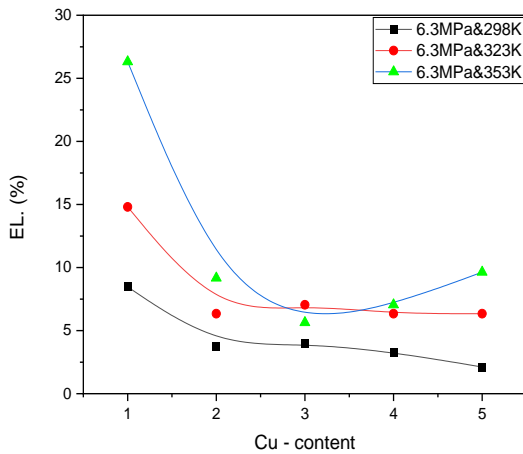


Figure. 4: The EL.% for (81Sn – xCu – (19-x) In) % alloys under different loads (2.5, 6.3, and 11.4 MPa) at (25, 50, and 80°C).

3.2.3 Creep stress exponents and activation energy for (81Sn – xCu – (19-x) In) %alloys: -

The stress exponent (n) parameter and activation energy (Q) of the steady state creep were calculated using the relation [14, 20]:

$$n = \frac{\partial \ln(\dot{\epsilon}^0)}{\partial \ln(\sigma)} \dots \dots \dots (2) \quad \text{where,}$$

σ is the stress

$$Q = R \frac{(\partial \ln(\epsilon^0))}{\partial \ln(\frac{1}{T})} \dots \dots \dots (3)$$

where, R is the gas constant and T is the temperature. Table (3) shows the result of the stress exponent and strain rate sensitivity (n and m) at different loads and temperatures for the tested

alloys. The values of the strain rate sensitivity, (m) were represented by the slope of each segment line ($\partial \ln \sigma / \partial \ln \epsilon^0$), and varied in the range from (0.4 at low temperatures to 6.33E-01 at high temperature) [21]. According to the literature review, the values of n depend on temperature [22].

Sample in wt. %	Stress exponent and strain rate sensitivity					
	n			m		
Tem.	298K	323K	353K	298K	323K	353K
81Sn - 1Cu - 18In	2.38616	2.54	2.48	0.419083381	3.94E-01	4.03E-01
81Sn - 2Cu - 17In	1.38777	1.58909	1.80143	0.7205805	6.29E-01	5.55E-01
81Sn - 3Cu - 16In	0.91696	1.52	1.5799	1.090560112	6.60E-01	6.33E-01
81Sn - 4Cu - 15In	1.94496	1.75026	2.32908	0.514149391	5.71E-01	4.29E-01
81Sn - 5Cu - 14In	0.98217	2.01048	2.4802	1.01815368	4.97E-01	4.03E-01

Table (3) stress exponent (n) and (m) values for five alloys at temperatures (25, 50, and 80°C).

The results listed in the Table (3) reveal that n decreases at high temperatures, which is due to the instability of microstructure at elevated temperatures.

The difference between n values for all alloys at different temperatures is due to the dislocation movement (climb – creep) mechanism, while at temperature T=50°C the n value of the (81Sn-xCu-(x-19) In) % alloy with (1, 2, 3, 4, and 5) is due to the dislocation movement (slip–creep) mechanism.

Also, the activation energies in the steady state creep for five alloys were calculated from the slope of the linear between Ln (ϵ^0) and 1000/T, and they were found to be vary in the range (0.4 -5) eV as shown in Table (4). Figure. 6 (a, b, and c) shows the activation energy under three different applied loads. It is noticeable that Q of the (81Sn-4Cu-15In) % alloy has the highest activation energy value compared to the other alloys, and the activation energy decreases with the increase of applied loads, which means that activation energy depends strongly on the applied loads [20].

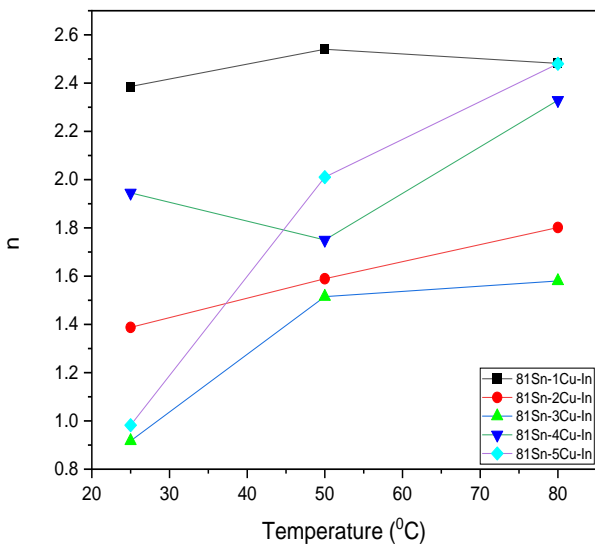
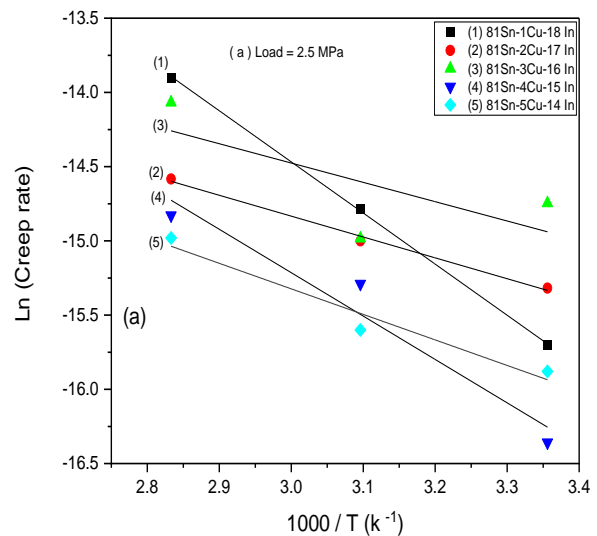


Figure. 5: The stress exponent (n) values at different temperatures.



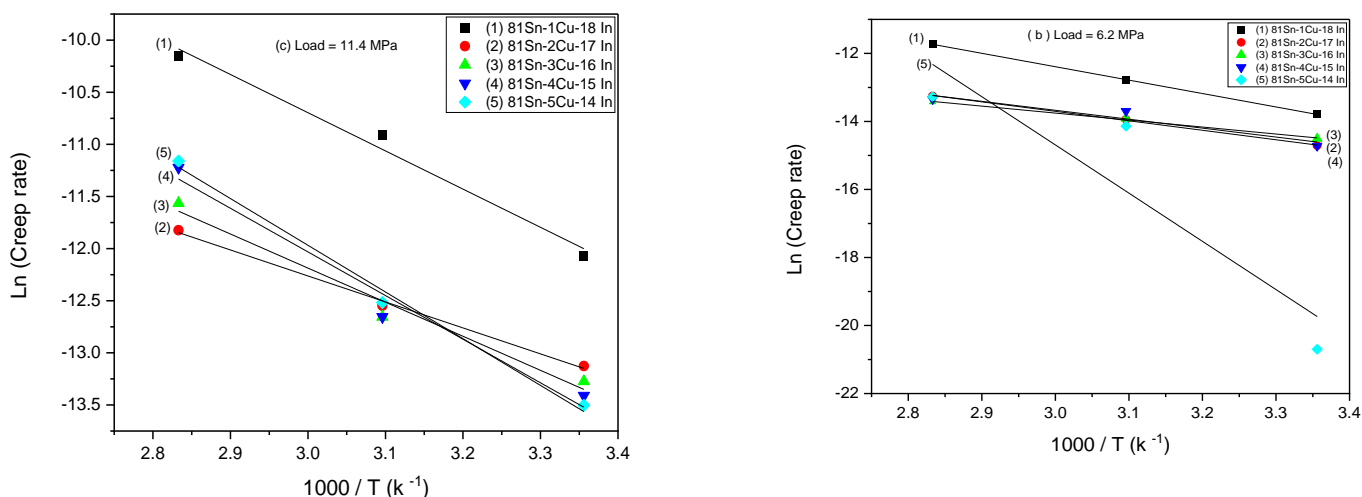


Figure. 6: The relation between Ln ($\dot{\epsilon}$) and 1000/T for five alloys at (a) 2.5 MPa, (b) 6.3Mpa, and (c) 11.4 MPa.

Sample in wt. %	Activation Energy (Q) (eV)		
	2.5 MPa	6.3 MPa	11.4 MPa
81Sn - 1Cu - 18In	1.296671472	1.484	1.380
81Sn - 2Cu - 17In	0.5291	1.049	0.938
81Sn - 3Cu - 16In	0.490	0.773	1.231
81Sn - 4Cu - 15In	1.101	0.987	1.576
81Sn - 5Cu - 14In	0.648	5.337	1.687

Table (4): Activation energy values for five alloys at different loads (2.5, 6.2, and 11.4 MPa)

4. Conclusion

We conclude that:

- The creep rate of (81Sn-xCu-(19-x) In) % alloys with x = (1, 2, 3, 4, and 5) at high temperatures and high loads is higher than that at low temperatures and low loads.
- The stress exponent for (81Sn-xCu-(19-x) In) % alloys with x = (1, 2, 3, 4, and 5) decreases at high temperatures; this is due to the instability of the microstructure at elevated temperatures.
- The activation energy decreases with the increase of applied loads, which means that activation energy depends strongly on the applied loads, (high rupture time, low strain rate, and high creep activation energy).
- The mechanical properties of creep rate, stress exponent, and activation energy are the best in the composition (81Sn-3Cu-16In) % alloy and show the best performance and stability compared to the other compositions.

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