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INVESTIGATION OF SUPERPLASTIC BEHAVIOR OF CERTAIN (Zn – Al) ALLOYS

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ABSTRACT

Super plasticity behavior finds applications in so many fields, for example the aerospace manufacturing that is the main bazaar for super plasticity, but automotive, medical, sports, cookware and architectural applications have their share too. "In this work a study of the superplastic behavior of a new Zn-Al alloy was conducted. In addition to the investigation of the possible superplastic behavior of Zn-0.5Al alloy. These alloys were prepared by using gravity and chill casting techniques. Zn-0.5Al alloy was subjected to hot rolling at 250 ºC and cold rolling at room temperature, while Zn-48Al alloy was also hot rolled at 250 ºC to 20% reduction in the thickness of sample followed by partial remelting at 500 ºC. Several tests were carried out such as physical, mechanical and chemical which include (XRF, XRD, OP, SEM, Microhardness (HV) and Tensile (cold, hot) test). Results showed that the Zn-0.5Al alloy has poor mechanical properties and may not be regarded as a superplastic alloy compared with Zn-48Al alloy. The Zn-48Al alloy generally enhanced all properties. The maximum elongation of (450%) was obtained in Zn-48Al alloy after thermomechanical controlling process and partial remelting.

KEYWORDS: Zn-Al alloy, Superplastic behavior, hot rolling, partial remelting, hot tensile

التحقيق في سلوك اللدونة الفائقة لسبائك (خارصين – ألمنيوم) معينة

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الخلاصة

يجد سلوك اللدونة الفائقة تطبيقات في العديد من المجالات، مثلا الصناعات الفضائية التي هي أكبر سوق للدونة الفائقة، الا أن التطبيقات مثل السيارات والتطبيقات الطبية والرياضية وتجهيزات المطابخ والهندسة المعمارية لها حصتها أيضا. في هذا العمل تم إجراء دراسة لسلوك اللدونة الفائقة لسبيكة (زنك – ألمنيوم) جديدة. بالإضافة إلى التحقيق في سلوك اللدونة الفائقة المحتمل لسبيكة (زنك - 0.5 المنيوم). تم تحضير هذه السبائك باستخدام تقنيات السباكة بالجاذبية و السباكة بالصقع. تعرضت سبيكة (زنك - 0.5 المنيوم) للدرفلة على الساخن عند 250 درجة سيليزيه وللدرفلة على البارد بدرجة حرارة الغرفة, في حين أن سبيكة (زنك - 48المنيوم) و سبيكة (زنك - 48 المنيوم مع إضافة B_4C) أيضا تم درفلتهم على الساخن عند 250 درجة سيليزيه لتخفيض بنسبة (20%) من سمك العينة يليه اعادة صهر جزئي عند 500 درجة سيليزيه. أجريت العديد من الاختبارات مثل الفيزيائية والميكانيكية والكيميائية والتي تشمل اختبار الاشعة المتفلورة، حيود الاشعة السينية، المجهر الضوئي، المجهر الالكتروني الماسح، الصلادة الميكروية (HV) والشد (البارد، الساخن)). وأظهرت النتائج أن أن سبيكة (زنك - 0.5 المنيوم) أظهرت خصائص ميكانيكية ضعيفة و ربما لا تكون سبيكة فائقة اللدونة مقارنة مع سبيكة (زنك - 48 المنيوم). عموما سبيكة (زنك - 48 المنيوم) ذات خواص جيدة من ناحية اللدونة الفائقة. ان أقصى استطالة تم الحصول عليها في هذه السبيكة هي (450%) بعد عملية التحكم الميكانو حرارية و إعادة الصهر الجزئي.

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INTRODUCTION

Superplasticity is the ability of a polycrystalline material to exhibit, in a generally isotropic manner though there are some works reported on anisotropic behavior also, very large elongations without necking prior to failure, usually observed with the elongation greater than 200% in tensile test [G. Wang, & M. W. Fu, 2007]. There are two main advantages in utilizing superplastic materials for metal forming operations. First, large strains can be achieved without necking. Second, the stresses required for superplastic deformation are generally low [F. A Mohamed, 2011].

The basic requirements for superplasticity are: A fine and stable grain size, typically 10 μm. Relatively high temperature, i.e. ($T \geq 0.5Tm$) where (Tm) is the absolute melting temperature of the material in Kelvin degrees. Very small strain-rate of the order of 10^{-3} to 10^{-5} s^{-1}. The strain rate sensitivity index, m value ≥ 0.3 and tensile elongations in extra of (200%) are generally revealing of superplastic behavior [G. G. Costanzo Bellini & L. Sorrentino, 2015].

A wide range of metals and alloys were identified to exhibit structural superplasticity. Several Zn-Al alloys exhibit superplastic behavior such as Zn-0.2Al, Zn-4.9Al, Zn-22Al and Zn-40Al [G. Kumaresan, 2014]. One of the best known superplastic materials is the Zn-Al eutectoid containing 22Al, and the deformation characteristics of this alloy have been reported by a number of workers. In this study, an investigation has been carried out of the effect of increasing the aluminum content on the occurrence of superplasticity in Zn-Al alloys. The compositions Zn-0.5Al and Zn-48Al have been studied.

EXPERIMENTAL PROCEDURE:

The samples were prepared by melting quantities of pure Zn and Al in electrical resistance furnace type (Via P.da Cannobia, 10, 20122 MILANO, Italy). Graphite crucible with 0.5 Kg capacity was used as container of the alloy. The temperature was setup at approximately 500 ^oC for (Zn-0.5Al) alloy and approximately 700 ^oC For (Zn-48Al) alloy to insure the melting of aluminum (660 ^oC) completely. As for zinc (420 ^oC) was added gradually with decreasing molten temperature to reduce or prevent zinc evaporation. After the zinc melted completely, the alloy was stirred by the graphite rod to achieve homogenization.

Before the pouring of the molten alloy in the mold the slag removed by the same rod. The (Zn-0.5Al) alloy produced by gravity casting, while (Zn-48Al) alloy produced by gravity and chill casting techniques. The initial dimensions of the cast are (150 × 30 × 8) mm, 1 mm of the top and bottom surface of the sample were cut away in order to eliminate the high porosity regions. The cast was homogenized at 400 ^oC for 5 hours for both alloys. The chemical compositions of these alloys were analyzed after casting process as shown in Tables (2) & (3).

These alloys subjected to rolling process, the (Zn-0.5Al) alloy hot rolled at 250 °C to 40-60% reduction, then room temperature rolled to 90% reduction. While (Zn-48Al) alloy subjected to hot rolled only at 250 ^oC to 20% reduction in the thickness of samples, followed by partial remelting at 500 ^oC for 30 min, to produce suitable microstructure for superplastic properties examination.

Microstructural analyses were carried by optical and scanning electron microscopy (1280XEQ-MM300TUSB) and (FEI Quanta 450 FMG SEM (USA)), respectively. The X- ray Diffraction test (XRD) was performed by using (XRD) device type (D8 II machine, Bruker axs). Vickers microhardness measurements test was performed using (a Digital Micro hardness tested HV-1000) test device, with load 500g with socking time 10 sec. Tensile test was performed on a universal testing machine at temperature 260-300 ^oC and strain rate of 0.2 mm/min. The dimensions of tensile test sample according to the (ASTM B 557M-02) a sub size sample 6 mm width.

RESULTS AND DISCUSSION

Mechanical Tests

Tensile Test

The results of the tensile test and mechanical properties (ultimate tensile strength, yield strength and percentage of elongation) of the prepared samples are listed in Table (4). The tensile properties of A_3 - sample was poor because of low percent of aluminum and high percent of zinc which has HCP structure and the number of slip planes is low, therefore total elongation to failure 18% as shown in figure (2a), while it is possible to see that the total elongation of B_2-sample was 88% as shown in figure (2b).

Comparisons of tensile elongation data of B_2 - sample to those of C_2 - sample reveals that the chilled alloy (i.e. C_2 - sample) significant increased the superplasticity in the Zn-48Al alloy. It is believed that the microstructure evolution including the changes in grain size, grain shape and the properties of grain boundary are directly responsible for the changes in the superplasticity of the Zn-48Al alloy.

Because the superplasticity refers to the ability of a material to pull out uniformly to a very high elongation by grain boundary sliding. Thus, a fine grain size provides large quantity of grain boundary for the grain boundary sliding (GBS) mode of deformation during superplastic deformation. In addition, the flow stress decreases as the grain size decreased.

Closer observations on the microstructures of the Zn-48Al alloy reveal the fact of grain coarsening, because the grain coarsening is bad for the GBS mode superplasticity and the formation of high angle grain boundaries. Directly after thermomechanical controlling process, the C_4 - sample demonstrated significant improvements in tensile properties due to the elimination of porosities, the refinement of the microstructure to 0.45 μm and the ultrafine of grain structure.

To compare between the tensile properties for the prepared samples, it's clear that the maximum elongation obtained for C_4-sample and the minimum for A_3- sample. While the other properties such as (yield strength, ultimate tensile strength…) vice versa.

Microhardness Test Results:

Table (5) demonstrates the results of the hardness tests. Each result was considered as an average of three readings. From the results, it can be noticed that, by adding 0.5 and 48 wt. % Al the hardness values markedly improved compared with pure zinc (A_0). However, due to the higher solubility of Al in Zn and thereby low second phase volume fraction as shown in A_2 and B_2 – sample respectively in the phase diagram for Zn-Al system.

The dendritic structure of Zn-48Al alloy is not superplastic; it is harder and less ductile than the equiaxed fine grained structure as shown in B_2 - sample. This indicates the importance of a very fine equiaxed microstructure because the large grain boundary area associated with such a microstructure makes grain boundary diffusion the rate-controlling process.

By apply homogenizing heat treatment some of the material properties changed, such as strength and hardness. It reduces the internal stresses and improve the internal structure of the alloy, but become less hardness and the ductility increases.

Interestingly, after rolling no apparent improvement of hardness was observed in any sample; because the work hardening is equal to softening (recrystallization), hence no apparent increase in hardness. Also, indicating that even strong grain refinement does not influence the hardness of the Zn - based alloys significantly (A_3, and C_3).

MICROSTRUCTURES CHARACTERIZATIONS:

Optical Microscope Analysis:

Microstructure of any metal or alloy affected by several factors, including the chemical composition, thermal and mechanical processes, which in turn determines the quality of the resulting phases. The internal structure can be controlled by controlling the ratio G/R between the thermal gradient (G) to the rate of cooling (R). Since whenever this ratio was little they could get on with equiaxal structure. The control of this ratio through the appropriate choice of method of casting that considered as amid for cooling from the viewpoint of specialists mineral-class basis [T. S. Habeb**,**2006].

Figure (6) shows the microstructure of the investigated materials. As seen, the pure Zn as shown in figure (6a) possesses quite large grains with size 148 μm. The measuring is carried by putting listed ruler (100 degree per mm) on sample to be measuring the microscopic structure. Then, it has been compared the image scale with ruler scale to determine the image scale. The computer program (its name was scope Image plus, 3-0.4 copyright 2003-2007, accompanier with software of microscopy) and use the following techniques.

However, after alloying with 0.5 wt. % Al the structure generally created of β-Zn solid solution because of the high solubility of aluminum in zinc (1.1 wt. % at 382 °C). Examination by greater magnification showed a little quantity of eutectoid at grain boundaries. The grain size was reduced to about 129.5 μm as shown in figure (6b). Moreover, due to the rapid solidification, the alloy had a supersaturated single phase microstructure [E. Mostaed, M. Hashempour, M. Bestetti, A. Tuissi, & M. Vedani, 2014].

Figure (6c) shows the homogenization treatment. The cast alloys are usually homogenized by keeping the material at a certain temperature for a pre-defined time to allow diffusion of the alloying elements from the grain boundaries and other segregated areas. Homogenization treatment assists in reduction of the micro-segregation, removal of low melting point eutectics which may cause incipient melting during thermo-mechanical processing, and controlling precipitation, that Zn and Al gradually diffuse from higher to lower concentrations. But the grain size increases to 156 μm due to temperature [H. M. M. Rashed**,** 2010].

The microstructure of the rolled sample figure (6d) would propose that a smallest grain-size was got, resulted through the induction of dislocations because of more distortion. This grain-size seems to have been made possible through the occurrence of the dispersion of the fine Al-rich particles, these particles causing recrystallization by inhibiting boundary migration and dislocation recovery processes. Solute additions of aluminum in zinc have been revealed to increase the recrystallization temperature and prevent boundary mobility as suggest by other workers [D. A. Saheb, 2011]. The grain size after rolling process is 135 μm, because of the occurrence of grain growth.

The microstructure of B_1 - sample, shown in figure (7a), consists of developed dendritic and interdendritic eutectic structures (bright and dark structures, respectively). The dark interdendritic eutectic (α + β) regions, which represent the last metal to solidify, are enriched with zinc (β-phase), while the aluminum (α-phase), solidifies as primary dendrites. The measuring of DAS of the alloy cast gravitationally into steel molds are over 450 mm long and the grain size is of 35.5 μm [M. A. M. Arif, M. Z. Omar, N. Muhamad, J. Syarif, & P. Kapranos,2013 , M. Agapie, & B. Varga, 2015].

As previously mentioned the effect of homogenization treatment assists in reduction of the micro-segregation, removal of low melting point eutectics which may cause incipient melting during thermo-mechanical processing, and controlling precipitation, that Zn and Al gradually diffuse from higher to lower concentrations, while the grain size increased to about 46.72 μm as shown in figure (7b).

To compare the microstructure results of gravity and chill casting techniques the alloy casted in preheat steel mold and copper mold, respectively. In general, Zn-Al alloys have a dendritic structure resulting from gravity casting with dendritic arm spacing depending on casting parameters. The consequences of the dendritic structure are established, primarily, in the lower ductility of the cast alloy, as well as in relatively high inhomogeneity of mechanical properties.

Attempts in this work were made to produce fine grain by method called chill casting without grain refiner addition. Rapid solidification shows the well distributed fine grains and slow solidification rate shows coarse grain size. Figure (8a) shows the microstructure examination of the sample obtained from this technique. It was expected to produce non-uniform fine grains for α and β phases where have been observed the presence of very small amounts of eutectic at the limit of some crystals rather than dendritic structure because molten metal undergo higher cooling rate. The average grain size is roughly the same (19 µm) as shown in figure (8a) .It is almost within the range measured by reference [M. Agapie, & B. Varga, 2015].

This means that the required time to complete the solidification process (the metal reaches the eutectic temperature) is shorter in chill casting mold and this condition affects the microstructures strongly. Whereas, the pre-heating of 150°C steel mold has effect on the slow absorption of the heat generated from molten metal. This leads to longer solidification time than that of the copper mold. Figure (8b) shows the effect of homogenization treatment on the structure of C_2 - sample.

After being hot rolling deformed by a ratio of 20%, the primary grains in the longitudinal direction of rolling elongated and coarsened as shown in figure (8c). This is possible because of the fragmenting and coarsening of the grains, as a result of the strain force, during this thermomechanical treating process. One can conclude this microstructure is really induced by the prior rolled deformation and the rolling level of 20% is adequate for this change.

During partial remelting at semi-solid temperature of 500 ºC of the pre-deformed Zn-48Al alloy, the primary grains coarsened due to the dissolution of the interdendritic eutectics, its structure changed due to recrystallization. The liquid phase penetrates the grain boundaries. Finally, these grains’ projecting positions remelted and became ultrafine grains. After the refining was completed, coarsening and agglomerating occurred, and the ultrafine grains changed into coarsened grains and irregularly inter connected grains as shown in figure (8d). This structure was resolved by other workers using electron microscope [M. R. Azpeitia, E. E. M. Flores, & G. T. Villasenor,2012].

Scanning electron microscopy (SEM)

The SEM had been carried out for the Zn-48Al (C_4 - sample) as shown in figure (9), respectively. SEM microscope has revealed that the structure is composed of a very fine and homogeneous mixture of α and β, which are Al- and Zn-rich phases (dark and bright phases, respectively).

To compare between the grain sizes for the different samples, it's clear that the largest grain size obtained for A_3-sample and the minimum for C_4- sample as shown in Table (6).

X-Ray Diffraction Analysis

Figures (10 a, b, and c) represent the charts of the x-ray diffraction for the prepared samples (A_3, B_2, and C_4) respectively. By using Bragg's law can calculate d-spacing, the ($I/I_∘$) ratio represented the intensity for all peak (I) divided on the maximum intensity ($I_∘$).

X-ray diffraction patterns obtained from Zn-0.5Al, and Zn-48Al alloys. The results indicate that the figure (10a) sample show that the only element appeared is zinc, aluminum does not appear in this chart because of the low percent (0.5wt. %). While the other figures from (12b) and (12c) show the same result. We found that peak number (2, 5, 7, and 11) for α-phase and others peaks (1, 3, and 4, 6, 8, 9, 10) for β-phase [M. A. M. Arif, M. Z. Omar, N. Muhamad, J.

Syarif, & P. Kapranos,2013, M. R. Azpeitia, E. E. M. Flores, & G. T. Villasenor,2012, and T. J. Chen, Y. Hao, & J. Sun,2002].

CONCLUSIONS

The following Conclusions may be drawn from this work:

- A fracture point was obtained from superplastic (Zn-48Al) after an elongation of 260% in tensile test.
- Further improvement was introduced by hot rolling and partial remelting. A fracture point was also obtained after an elongation of 450% in tensile test.
- No indication of superplasticity behavior was observed in (Zn-0.5Al).
- Both hardness and tensile elongation are expected to increase with decrease of grain size. This behavior is unfortunately, not observed. This is because the deformation mechanisms are different.

Table 1. Refer to codes of the samples prepared in this study

No.	Sample Code	Item
1	A0	(Pure Zinc) as cast
2	A_1	(Zn-0.5Al) as cast produced by gravity casting
3	A_2	(Zn-0.5Al) after homogenizing produced by gravity casting
4	A_3	(Zn-0.5Al) after rolling produced by gravity casting
5	B_1	(Zn-48Al) as cast produced by gravity casting
6	B_2	(Zn-48Al) after homogenizing produced by gravity casting
7	C_1	(Zn-48Al) as cast produced by chill casting
8	C_2	(Zn-48Al) after homogenizing produced by chill casting
9	C_3	(Zn-48Al) after rolling produced by chill casting
10	C_4	(Zn-48Al) after partial remelting produced by chill casting

Tables 2. & 3. The chemical compositions of (Zn-0.5Al) and (Zn-48Al) alloys, respectively.

Table 2. Chemical composition (weight %) of (Zn-0.5Al) alloy

Al%	Si%	Mo%	Fe%	Cd%	Pb%	Zn %
0.46	0.31	0.008	0.06	0.002	0.002	**Rem**

Table 3. Chemical composition (weight %) of (Zn-48Al) alloy

Al%	Mo%	Fe%	Cd%	Pb%	Si%	Zn %
47.835	0.003	0.002	0.01	0.01	0.04	**Rem**

Table 4. Mechanical properties of the prepared samples

Sample	Yield Strength (Mpa)	Ultimate Tensile Strength (Mpa)	Deformation (mm)	Elongation %
A_3	210.029	300.857	5.76	18
B_2	97.83	144	28.16	88
C_2	33.37	42.012	83.2	260
C_4	25.143	37.942	144	450

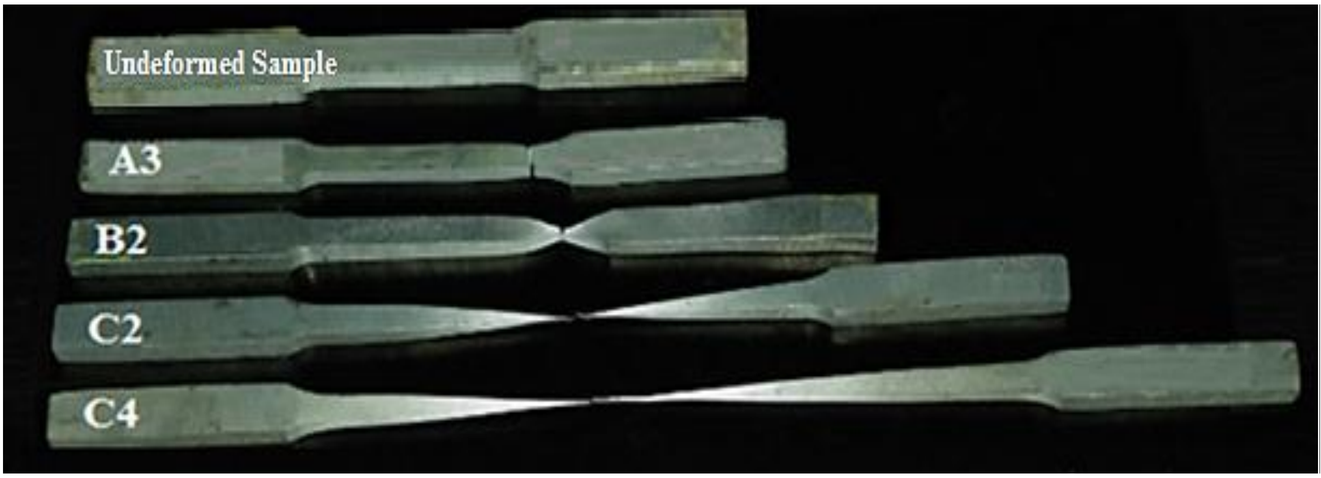

Fig. 1 Fracture of the tensile test samples

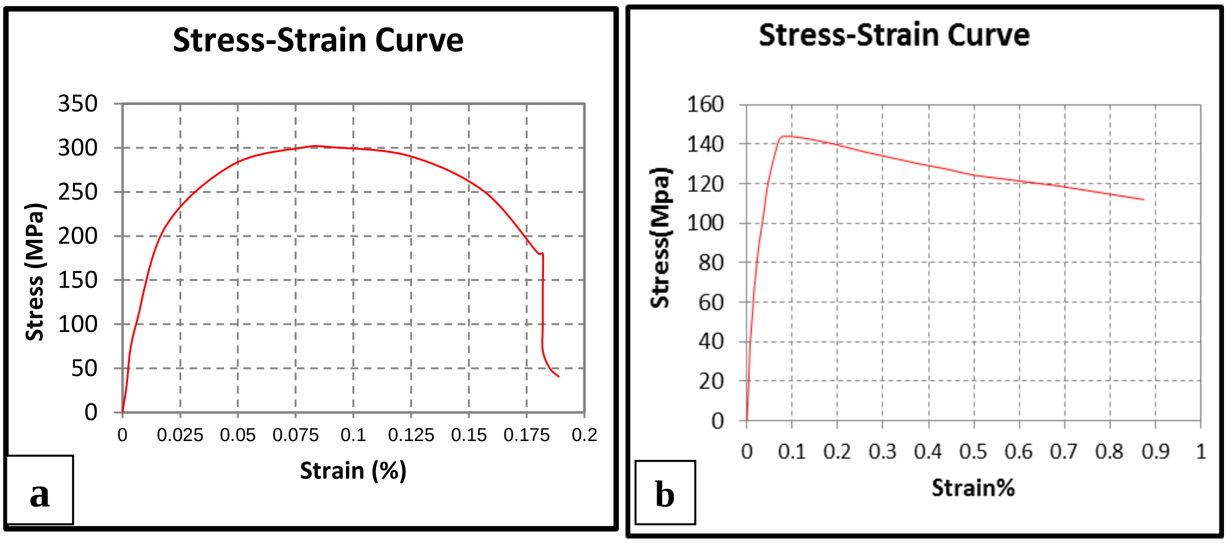

Fig. 2 Tensile test of (a) A_3 – sample (b) B_2 – sample

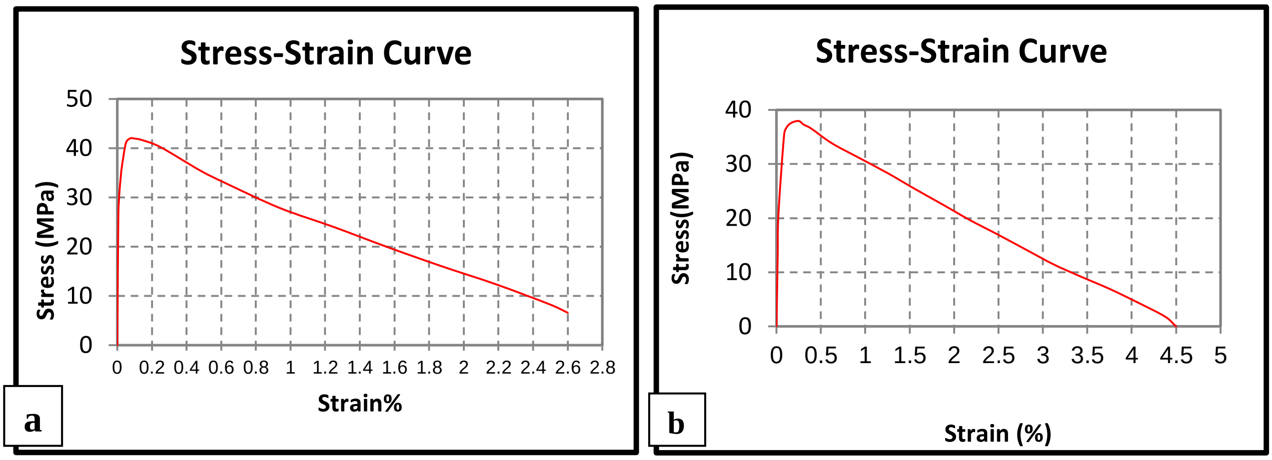

Fig. 3 Tensile test of (a) C_2– sample (b) C_4 – sample

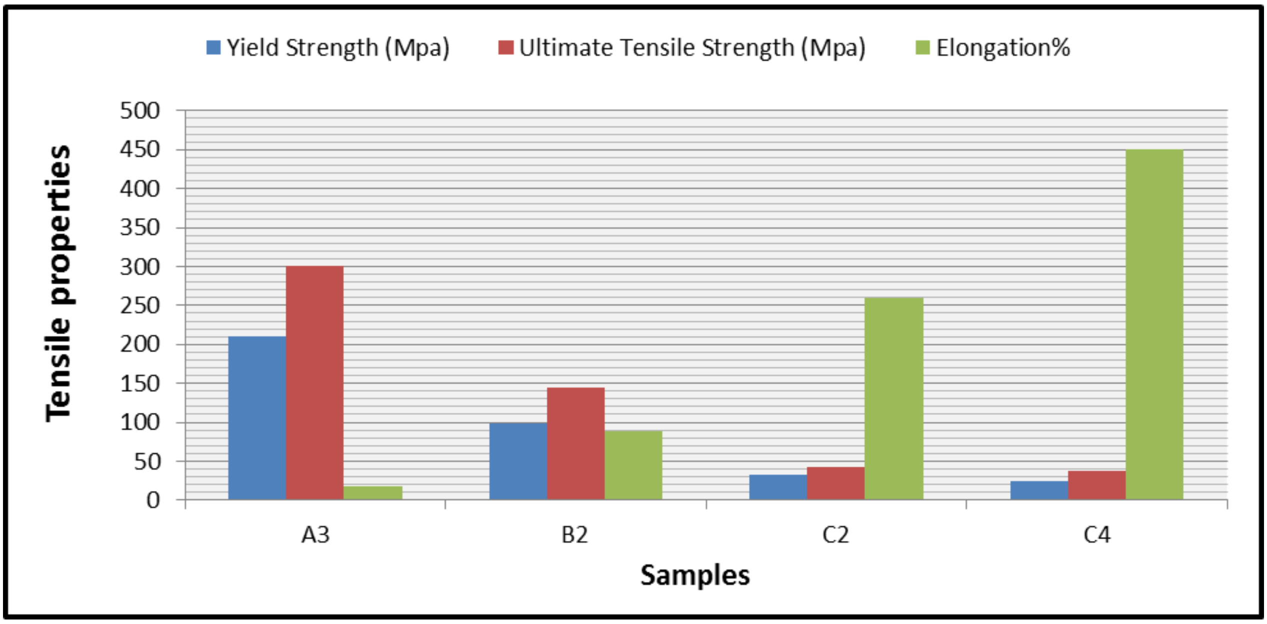

Fig. (4); tensile properties of the prepared samples

Table 5. Microhardness of the prepared samples

Sample	Microhardness (HV)	Sample	Microhardness (HV)
A_0	35.66	B_2	100.125
A_1	66.35	C_1	94.21
A_2	59	C_2	90.025
A_3	43.17	C_3	80.138
B_1	108	C_4	54.63

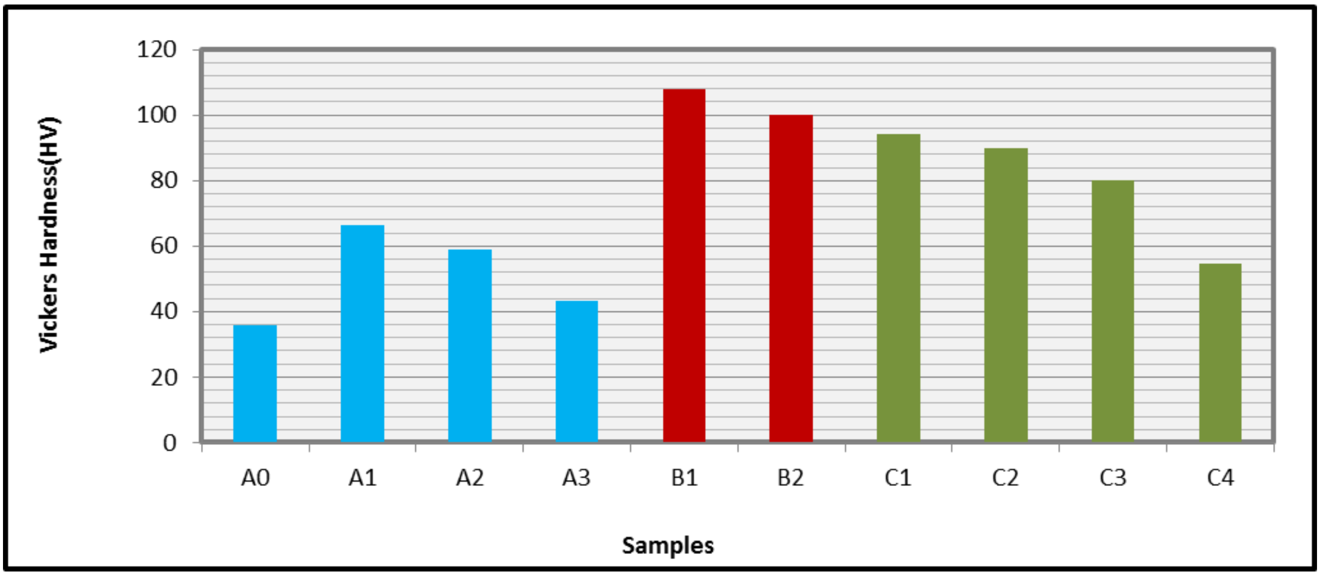

Figure 5. Hardness of the prepared samples

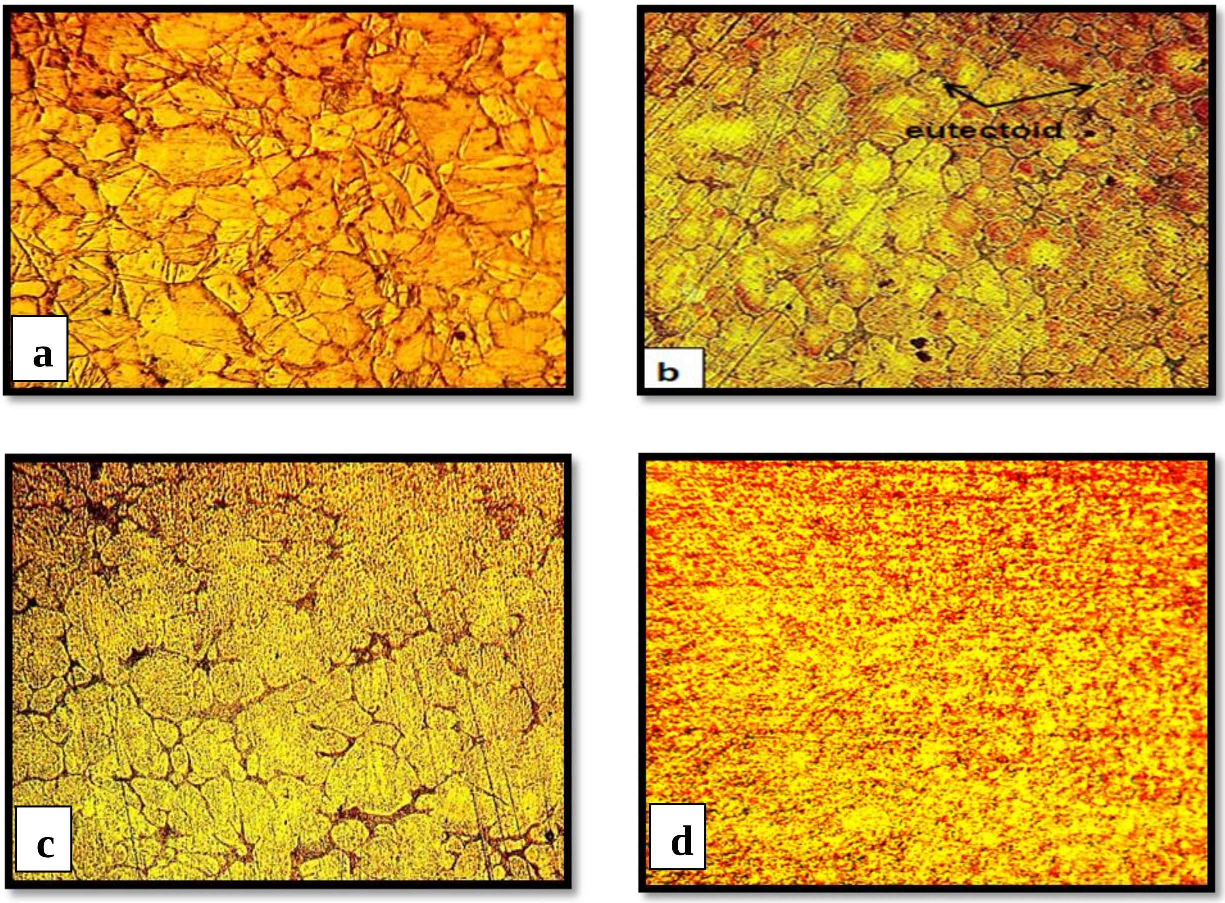

Fig. 6. Shows the microstructure of (a) A_0 (b) A_1 (c) A_2 (d) A_3 with Magnification 200X.

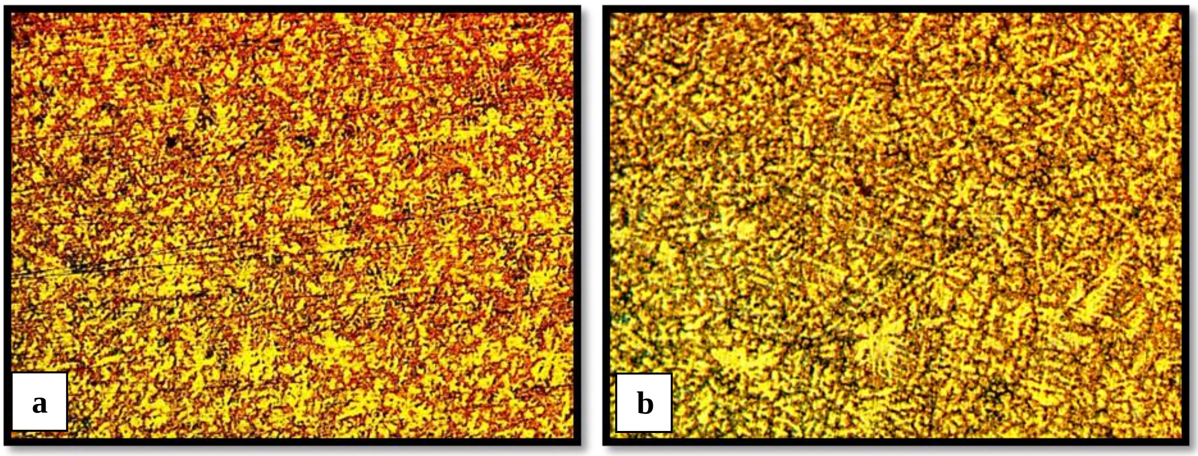

Fig. 7. Microstructures of (a) B_1 (b) B_1 with Magnification 100X.

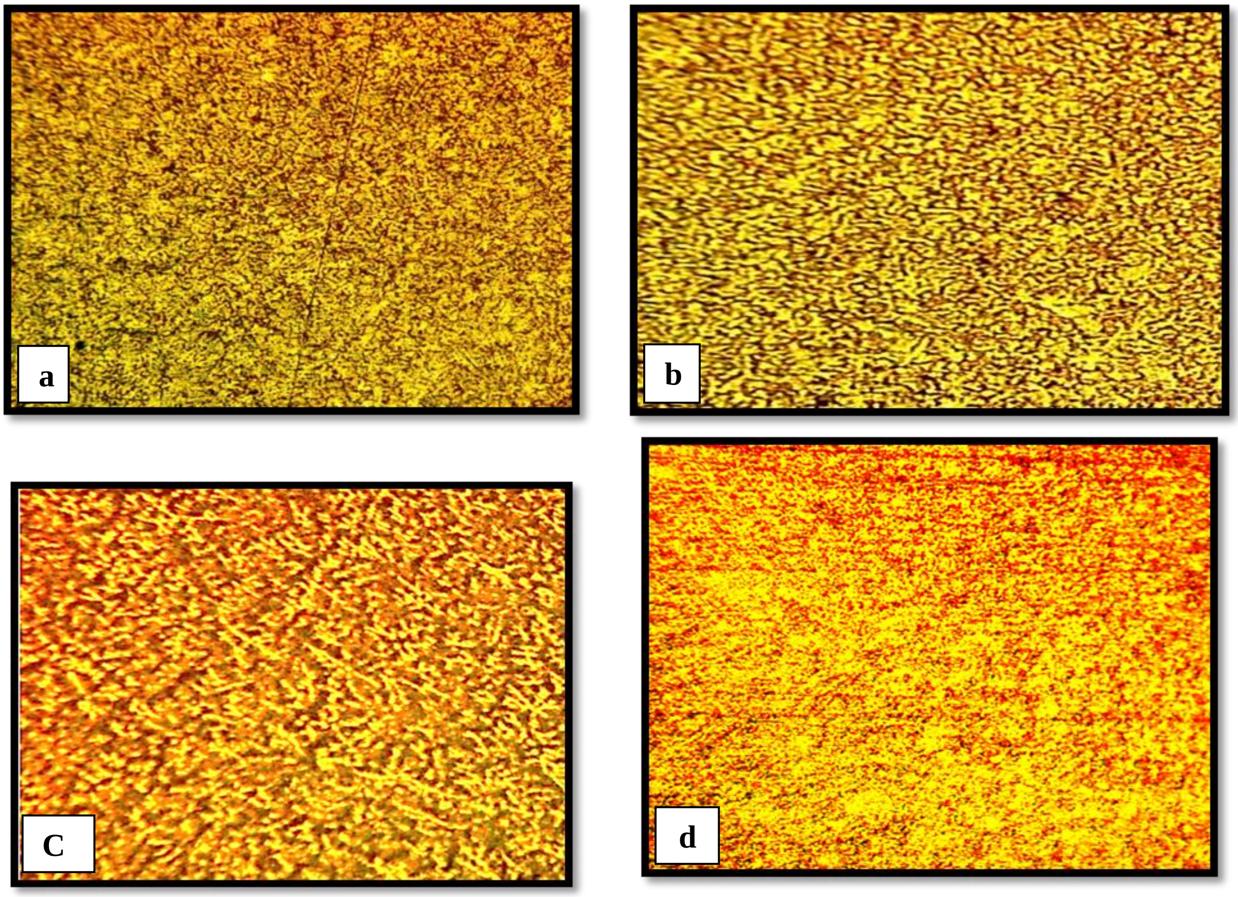

Fig. 8. Microstructure of (a) C_1 (b) C_2 (c) C_3 (d) C_4 with Magnification 100X.

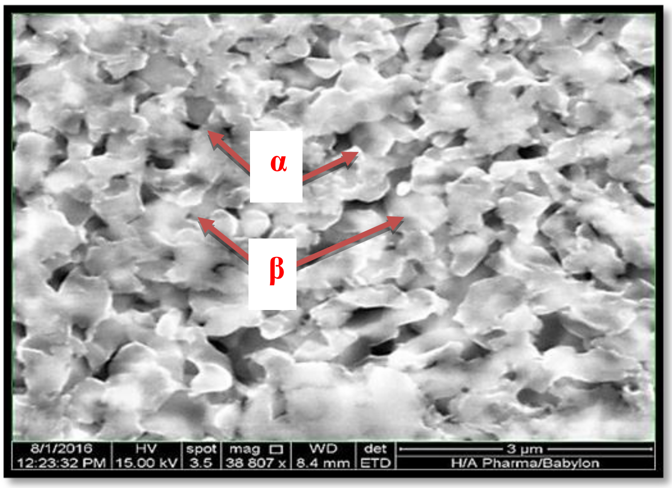

Fig.9 SEM micrograph of C_4-sample

Table 6. The average grain size of the different samples

Samples	A_3	B_2	C_2	C_4
Average Grain size (μm)	135	39.54	19.6	0.45

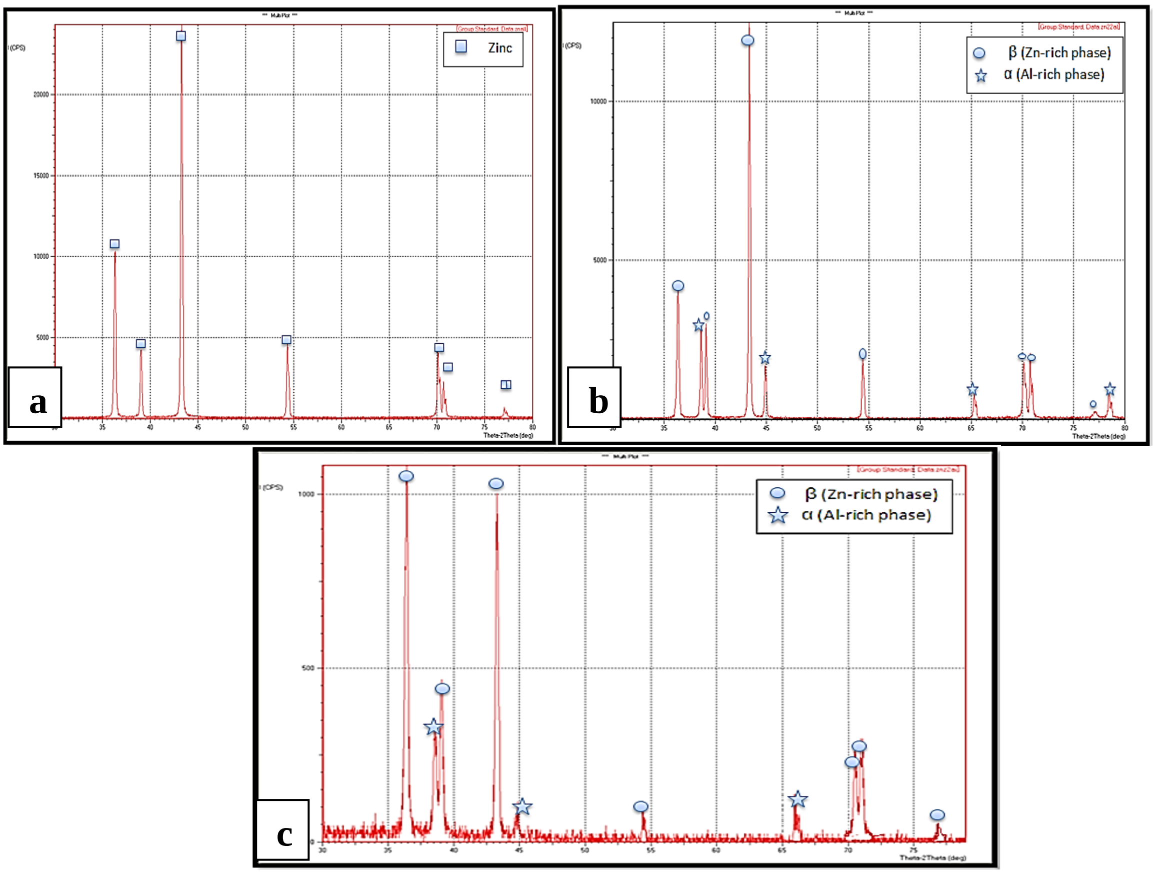

Figure 10. XRD patterns of (a) A_3, (b) B_2, and (c) C_4 – Samples

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