Study the Effect of some Metallic Additives on the Physical Properties of the Commercial Pure Aluminum Metal

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Abstract: The aim of the present work is to develop the 6201 alloy, which is the most used for conductor cables by adding different amount of Ce into Al-Mg-Si alloy namely (0.0, 0.024, 0.043, 0.054, 0.133, 0.166 and 0.194 wt% Ce) concentration. Sample alloys were homogenized by annealing at 540° C for various duration in range (½ to 5 hours), followed by water quenching. Tensile tests, hardness, electrical conductivity tests, microstructure characterization in Scanning Electron Microscope (SEM) have all been investigated as-cast and annealing. The results indicate that the alloys with Ce content make a more refined structure of grains and have higher tensile properties especially in range (0.043 to 0.054 wt% Ce) content and also hardly increase resistivity rather than the alloy which is free of cerium. [Journal of American Science. 2010;6(12):239-252]. (ISSN: 1545-1003).

Keywords: Tensile test, hardness, electrical conductivity, microstructure characterization

1. Introduction:

Aluminum and its alloys are characterized by a relatively low density, high electrical and thermal conductivities and a resistance to corrosion [1-3]. Many of these alloys are formed by virtue of high ductility [3], its ductility is maintained even at very low temperatures. The primary limitation of aluminum is its melting temperature (660°C). Mechanical strength of aluminum can be enhanced by cold work and by alloying [1,3]. Principle alloying elements are copper, magnesium, silicon, manganese and zinc. Aluminum is being widely used as a conductor material. The suitable alloy for aluminum conductors belong to Al-Mg-Si system with varied composition. Alloy 6201 is (Al- 0.8 Mg- 0.25 Si) designed for overhead conductor cables because it has excellent strength and good conductivity when suitably treated [4,5,6].

The use of rare – earth metal as a beneficial addition to nonferrous alloys has affected considerable interest in recent years. The rare – earth additions such as cerium have been claimed to led refinement of structure and improvement in mechanical properties. Rare – earth additions were also explored as a modifier to Al-Si alloys [7].

The main aim of the present work is the modification of the commercial aluminum alloy, AA6201 [Al- Mg- Si] to be used as conductor cable in our industries. For that a series of ternary master alloys [Al- Mg- Si] with different amount of cerium were prepared. A study of the effect of Ce content and heat treatment on the microstructure, the mechanical and electrical properties was made.

2. Experimental Technique:

Commercial aluminum of 99.8% purity, magnesium and silicon of high purity were used to produce a ternary alloy (Al- Mg- Si) in our investigations. The alloys were melted in a graphite crucibles open to atmosphere in a resistance furnace held at (700 – 800 ºC). The tested alloys were stirred well with a graphite rod, from time to time and long melting times were used to ensure dissolution, homogenization and uniform distribution of the alloying elements (Si & Mg) in aluminum. Cerium was added in the form of Al- Ce master alloy, the estimated amount of Al-Ce master alloy wrapped in aluminum foil, was plunged into the molten to produce a series of ternary alloys (Al- Mg- Si) with different amounts of Ce. The melt was stirred with a graphite rod then was poured in a steel mould to solidify in atmospheric air. The alloys were provided in the form bar of circular cross section of dimension 3 cm and length of 28 cm. Specimens test cut from the as cast rod with different dimensions for the required measurements before heat treatment.

For the tensile testing and resistivity samples were drawing into wires of 1 mm diameter. Solution treatment is the supersaturated solid solution of alloy structure is produced to take advantages of its precipitation hardening characteristics. Series of samples of each alloy were homogenized at a fixed temperature of 540ºC with annealing time of 30 minutes to 5 hours and thereafter immediately (within 30 seconds) quenched in cold water (at room temperature). The samples are then rinsed and left to dry the surface completely. Tensile test measurements were used as an indicator of mechanical response to heat treatment. The tensile machine used in this investigation is of type, (2wick-1425). All tensile measurements were performed on wire of 40mm length and 1mm diameter at room temperature. All the specimens had been heat treatment before using.

Hardness measurements were used as an indicator of mechanical response to heat treatment. The sample hardness was measured using Vicker's micro Hardness Tester Shemadzu. All hardness measurements were performed on a block of 5mm thickness at load of 200g and the pressing time is 5 seconds. The sample hardness was measured immediately after heat-treatment.

Microstructures of the as cast samples from each alloy were examined by conventional optical microscopy after mechanical polishing of the specimen followed by etching solution is about 20% hydrofluoric acid in distilled water. The etching was done by immersing the samples into the solution and waiting until a suitable contrast of the grains was obtained. The etched samples were washed, dried and scanned with SEM type. Joel JSM, 5410 to identify the existing phases, their shapes, and size distribution. To obtain further micro structural information, SEM is well suited to identify the existing phases, their shapes, and size distribution. Such investigation was carried on a series of Al- Mg-Si alloys without and with different cerium amounts of 0.024, 0.043, 0.054, 0.133, 0.166 and 0.194 wt% respectively. The tested specimens were solution heat-treated at 540º C for different times ranging from 0.5 hour and the water quenched.

Microstructure of both as-cast and solution heat-treatment alloys were characterized by Scanning Electron Microscopy (SEM) after polishing and etching.

3. Results and Discussion:

The microstructures of the tested alloys were examined by SEM as shown in Fig1, a-g. It can be observed that, the addition of cerium affected the microstructure development in the Al- Mg- Si alloy in various ways. Firstly, for Al- Mg- Si alloy without cerium, the alloy is composed of the primary α matrix and a secondary phase that exists in two kinds of morphologies, i.e. a discontinuous network of coarse particles along grain boundaries Mg_2 Si, and many spherical Si particles that distribute both inside grains and at grain boundaries, as shown in Fig.1, a. Secondly, after adding cerium Fig.1, b, β particles were refined. However, a new kind of distribution characteristics from β precipitate in the alloys with Ce, these particles generally have rod-like shaped and do not have an obvious tendency to distribute in grain [8,9]. The β- phase is found at the grain boundaries as a network of precipitates as observed in Fig.1 b-g. These precipitates depend upon the amount of cerium in the alloys with 0.043, 0.054 and 0.194 wt% {Fig.1, c,d and g}, there are less of spherical particles surrounded by much and continuous rod- shaped precipitates at grain boundaries. While, in alloys with (0.024, 0.133 and 0.166 wt%), there are much discrete spherical particles can be observed either in grains or along grain boundaries, furthermore there are still some rod- shaped precipitates at grain boundaries.

The morphology of all tested alloys (without and with Ce) are changed after the homogenization process as shown in the figures from Fig. 2, a-b to Fig. 8, a-d. The rods like β particles are gradually replaced by a uniform dispersion of new particles. With increasing homogenization time these particles are nucleated and grown. Therefore, the microstructure was found to have a new grains formation. The islands of new formed particles are thickened and spherodized at the expense of the remaining β- phase. At certain annealing time for each alloy, the formation of a homogeneous finely dispersed microstructure occurred. We can conclude that, the suitable time for the concentrations (0.024, 0.043, 0.054 and 0.194 wt% Ce) is 3 hours and it is 0.5 hour for the alloys with the concentrations (0.0, 0.133 and 0.16 wt% Ce). While, further annealing time, (i.e. the number of particles become fewer and bigger caused by coarsening [10].

Tensile testing measurements were carried out for as- received wires of each composition at room temperature as shown in Fig.9. It can be seen that the value of tensile testing for the samples containing Ce addition up to 0.194 wt% is higher compared to the Ce free alloy. The tensile strength and hardness increased from (333 and 70.18 N/mm^2) for the Ce-free alloy to (368.65 and 102.28 N/mm^2) for that containing 0.054 wt% Ce. Further additions up to 0.054 wt% Ce, have little effect on the tensile properties and hardness while, the maximum tensile strength was reduced to (337.436 and 71.27 N/mm^2), when the Ce content was further increased to 0.194 wt %.

Meanwhile, with increasing Ce content in the present alloys, the elongation was decreased compared with the Ce- free alloy. The elongation of alloys with contents 0.043 wt% and 0.194 wt% Ce respectively, were generally lower than that of other alloys. It can be concluded that increasing the Ce content increased the tensile strength and hardness but a slightly decreased in the elongation, compared to the alloy without cerium. This can be accounted due to in Al-Mg-Si alloy, to form the stoichiometric constituent the Mg and Si, which is the primary hardening phase. Any excess of silicon above the required Mg_2Si will contribute significantly to hardening [11].

Distribution of the Mg_2Si inter-metallic around the grain boundaries as was indicated in Fig.1, a in SEM. This kind of distribution of the related inter-metallic phases decreases the mechanical properties of the metal [12].

The addition of cerium improves the mechanical properties, this is ascribed to the grain refining effect during casting as previously shown from SEM micrographs. Also showed the best grain refining which can be obtained when the content of Ce is in the range (0.043-0.054 wt%). The structure refinement is one of the most important methods for improving the strength of alloys [13,14] besides the presence of spheroidal silicon particles and inter-metallic compounds of cerium (Al_2Ce, Al_4Ce,SiCe and $SiCe_4$) [15].

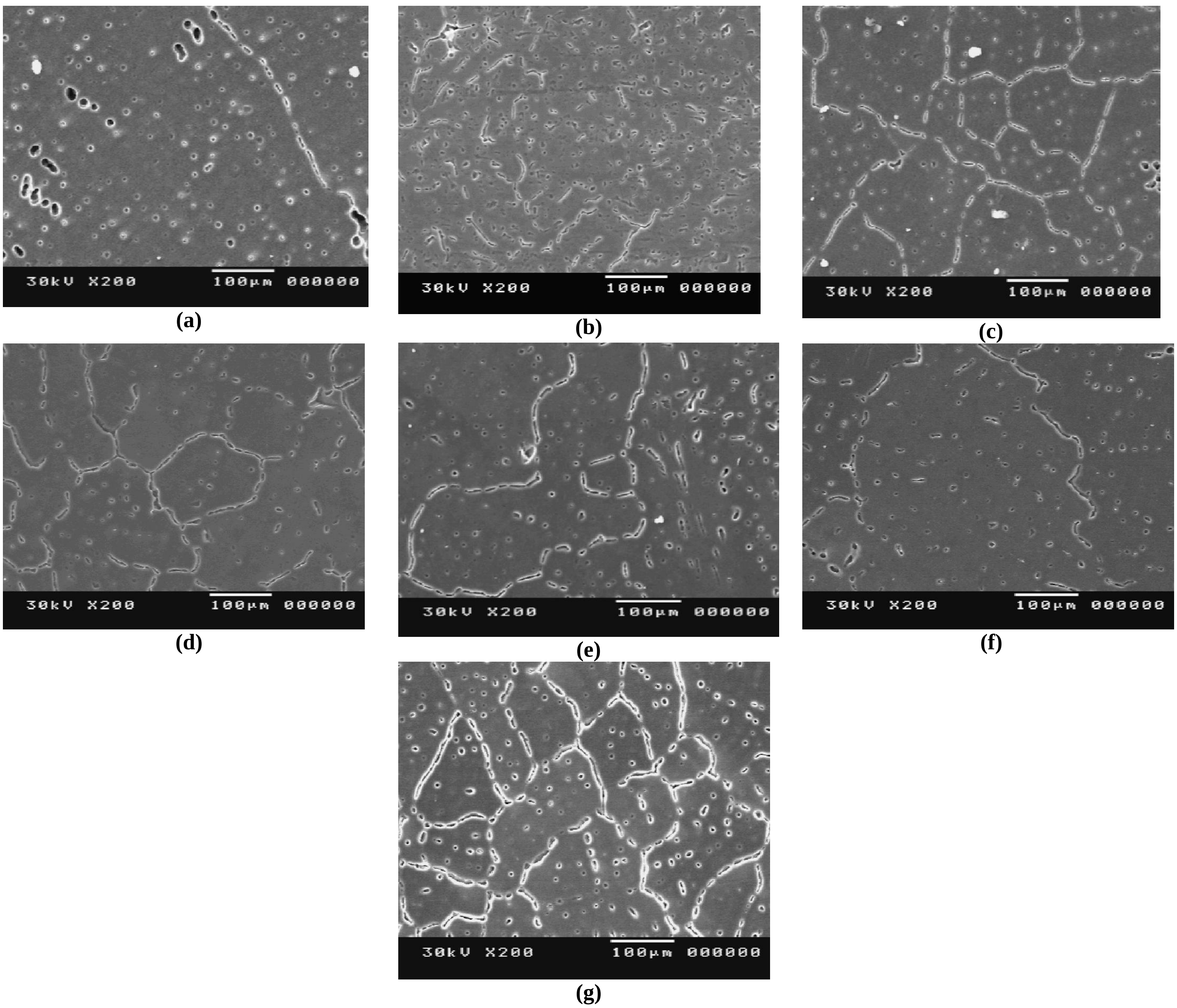
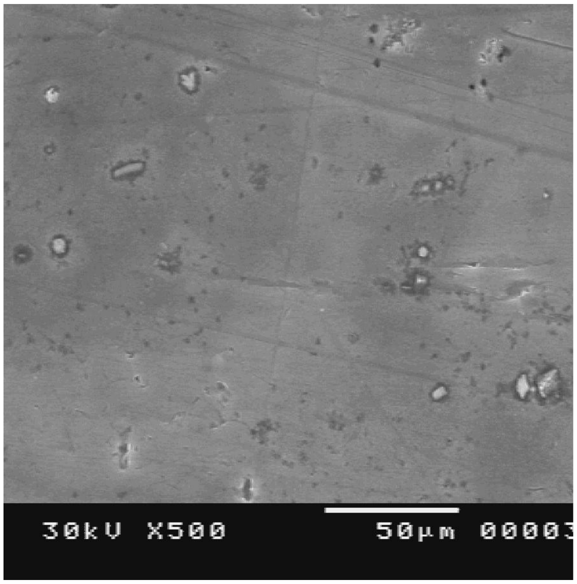
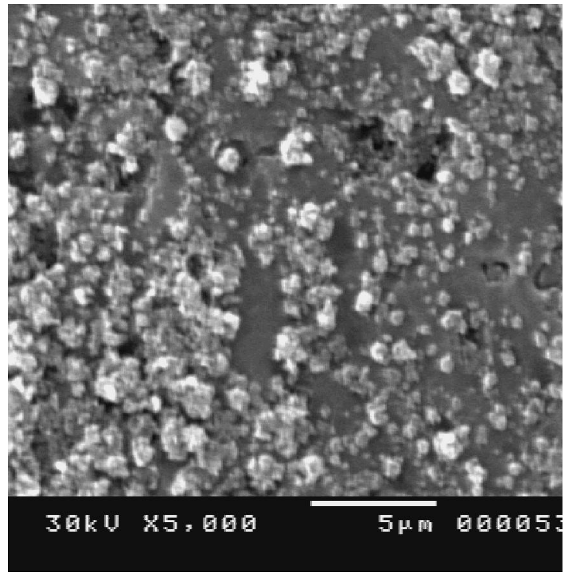


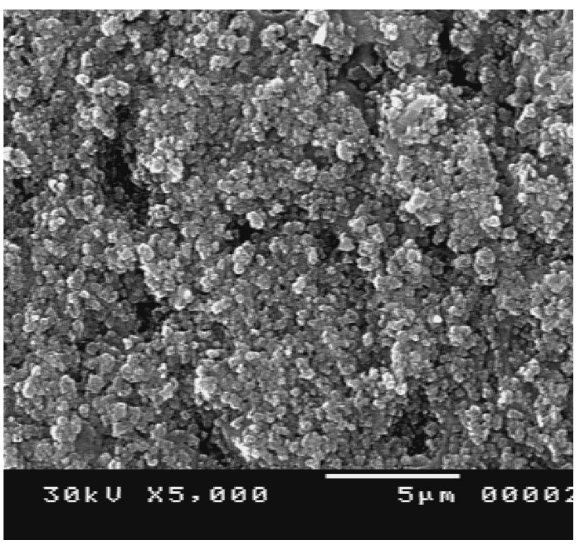
Fig.1a-g: SEM micrographs of the Al-Mg-Si alloy with different Ce concentrations; (a) 0.0 wt%, (b) 0.024 wt%, (c) 0.043 wt%, (d) 0.054 wt%, (e) 0.133 wt%, (f) 0.166 wt% and (g) 0.194 wt %



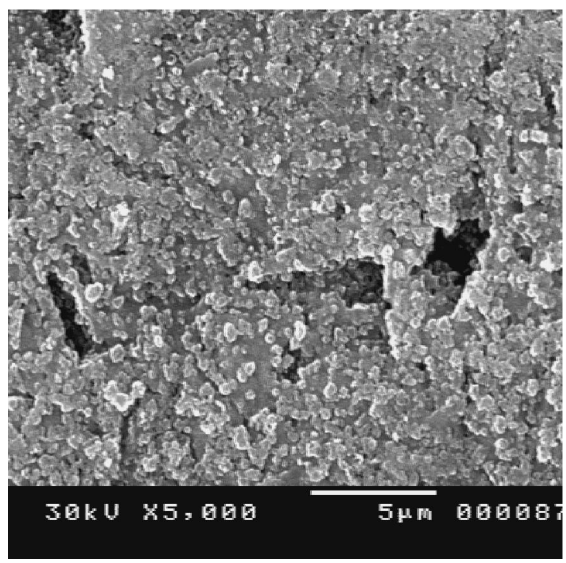
(a) 0.5h



(b) 1h

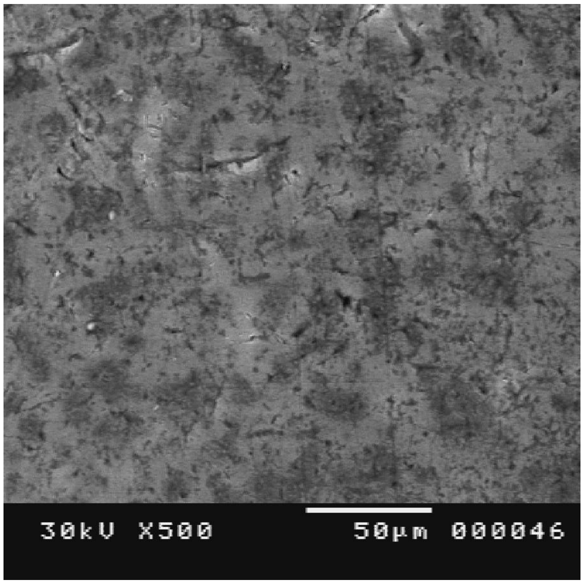


(C) 3h

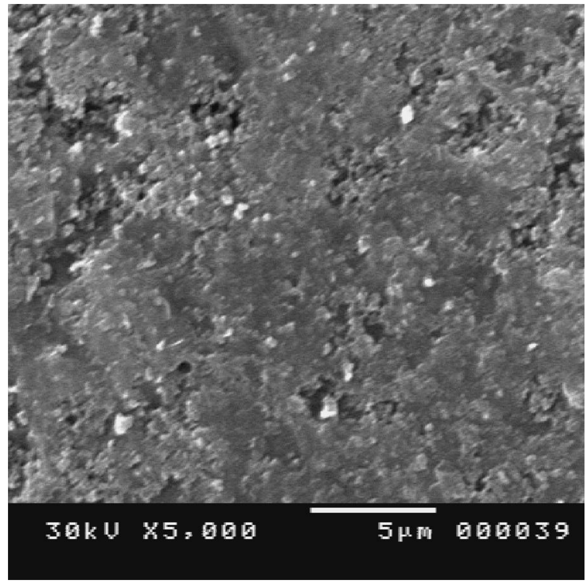


(d) 5h

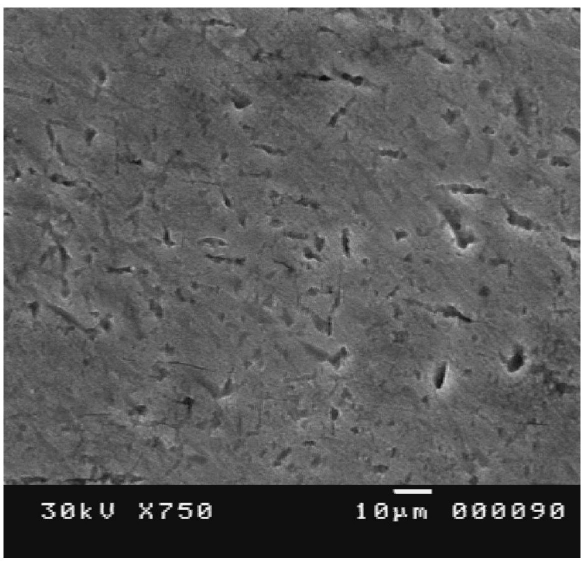
Fig.2a-d: SEM micrographs of the Al-Mg-Si alloy without Ce with different homogenization time (a) 0.5h (b) 1h (c) 3h (d) 5h.at 450C^{o}.



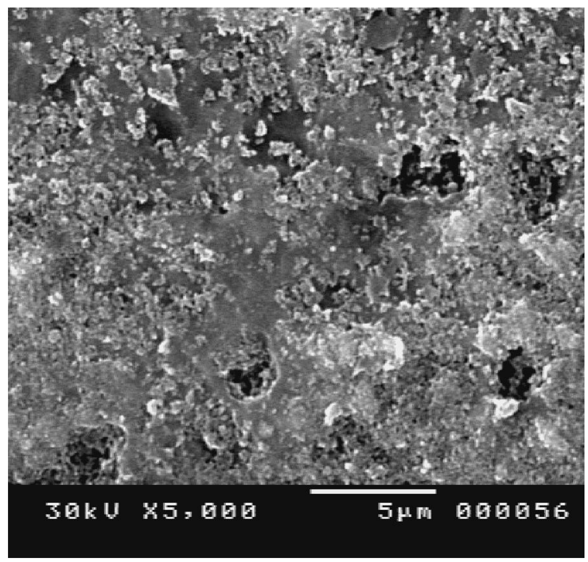
(a) 0.5h



(b) 1h



(C) 3h



(d) 5h

Fig.3 a-d: SEM micrographs of the Al-Mg-Si alloy 0.024 wt% Ce with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C.

Cerium has low resistivity coefficients and atomic radii that are relatively different from that of aluminum. These characteristics cause solute element to react with crystal defects such as dislocations and grain boundaries and enhance the mechanical properties of the base metal favorably [16]. It has also been reported that Ce reduced the interdendritic spacing of the alloy which can resist the movement of dislocation. In addition, the strengthening effects of Ce atoms segregated at the grain boundary have the contribution of keeping the highest tensile properties. That is, to say, the main reason that makes the segregation of Ce atom at grain boundary increases the sliding resistance of the grain boundary increase the mechanical properties. On the other hand, it is found that an excess of Ce > 0.054 wt% can reduce its useful effect [17].

All the alloys investigated which contain Ce have the highest strength accompanied by low ductility compared to alloy without cerium. The low ductility is due to the addition of cerium, which dissolves in the aluminum matrix and contributes to the formation of insoluble inter-metallic phase is after solidification [18].

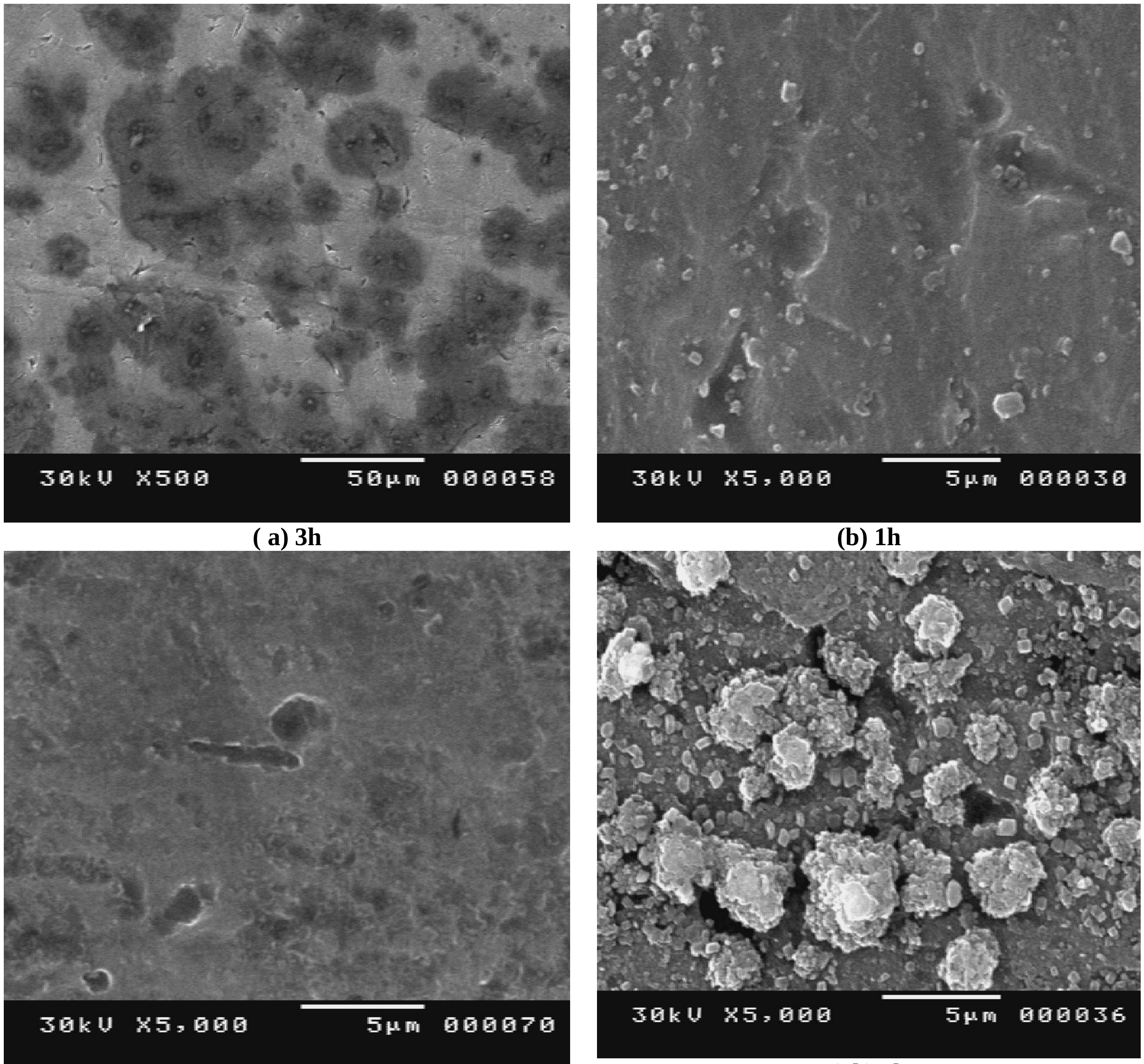
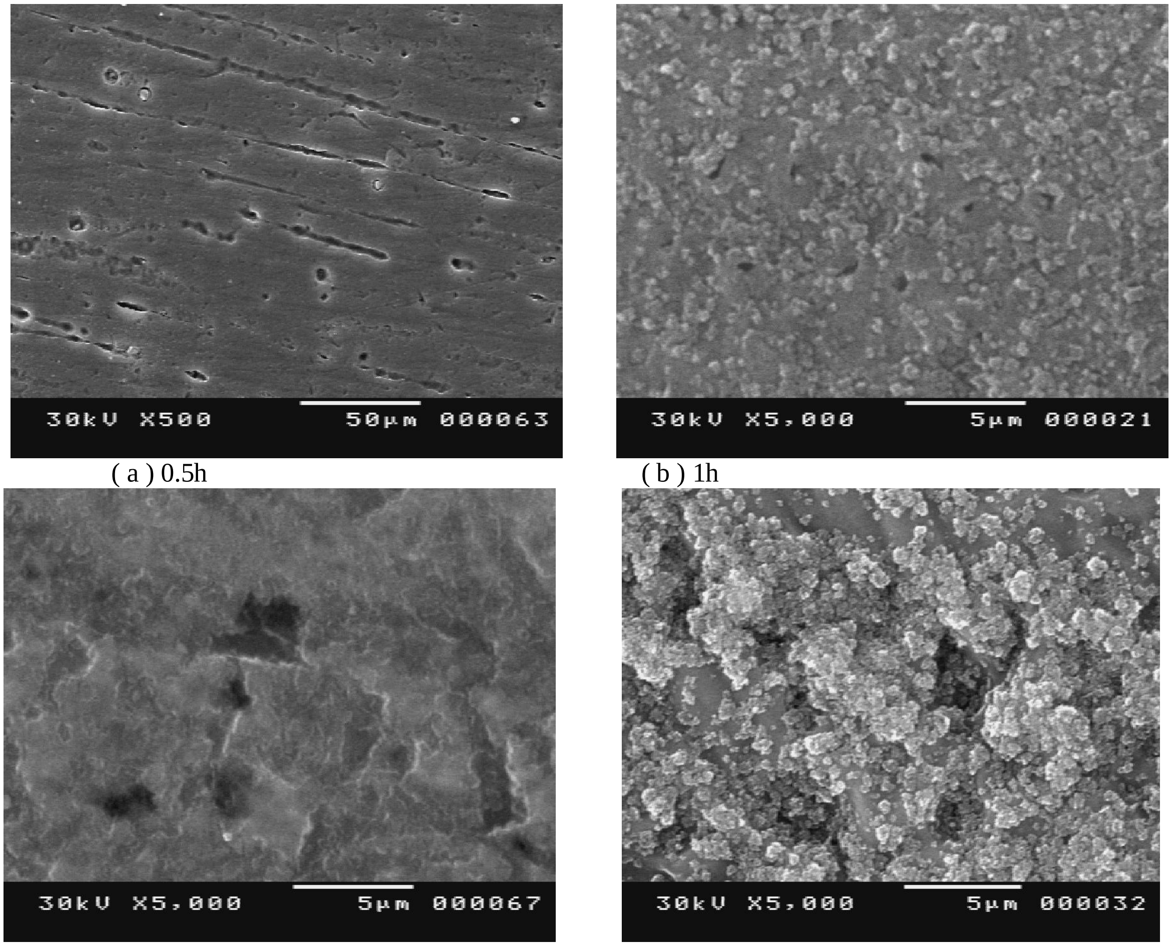


Fig. 4a-d: SEM micrographs of the Al-Mg-Si alloy 0.043 wt% Ce with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C

The influences of solutionizing time on the mechanical properties tensile strength, hardness and ductility of the Al-Mg-Si alloy at different Ce content in the range 0.0 wt% - 0.194 wt% are presented in Fig.10, a-c. It can be clearly observed from the figures that, after the homogenization temperature, the changing tendency of tensile testing is different to that at room temperature (unhomogenized alloy). It was observed that solutionizing time causes decreasing on the mechanical strength although the positive influences may be observed on ductility of these alloys depending on the solutionizing time of each composition as seen in Fig.10, a, b, and c. The ductility reaches its maximum value at 3 hours for 0.024, 0.043, 0.054 and 0.194 wt% Ce alloys and at 0.5 hour for (0.0, 0.133 and 0.166 wt% Ce) alloys. Beyond this time the ductility decreases gradually with annealing time as shown in Fig.10, c.



(a) 0.5h (b) 1h

(c) 3h (d) 5h

Fig. 5a-d: SEM micrographs of the Al-Mg-Si alloy 0.054Ce wt% with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C

All alloys homogenized and quenched exhibited reduction in both the tensile strength and hardness but the elongation associated with the measuring of homogenized alloys is higher than the unhomogenized . The observed improvement in ductility and the reduction of strength depends on the solutionizing time or soaking time which is applied at the specified heat-treating temperature to provide the chance for the dissolution of undissolved soluble phases and to achieve a homogeneous microstructure [18]. It was found that the mechanical properties change as a function of time as a result of morphology change as shown from SEM photographs. The micro structural development changes for all alloys during the phase transformation of β to α. The rod-shaped particles (β) are gradually replaced by a new uniform dispersion of particles. The solutionizing time, however, is not a constant but affect on particle size, particle geometry, and depend on overall composition [19]. So, it is important to keep the grain size largely unchanged before anneal, because the size of the grains affects the plastic deformation of materials. Shortened anneal time 3 hours for (0.024, 0.043, 0.054 and 0.194 wt% Ce) alloys at 0.5 hour for (0.0, 0.133 and 0.166 wt% Ce) alloys and at annealing temperature 540ºC were efforts to minimize the grain size as in Table (1). Further increasing in the homogenization time, the thickness of the particles increase as a result of coarsening mechanism, which may have a negative effect on the ductility, because the dislocations only flow around particles smaller than a critical size. It is found that the rate of the transformation is affected by the particles size of the alloy; if the particles are spheroidal and smaller, the dislocation may loop around the particles, thereby increase the ductility, and the material is easier to extrude [20].

The calculated values of resistivity are summarized in Table (2), it can be observed that the resistivity increases but the variation is low for the unhomogenized samples.

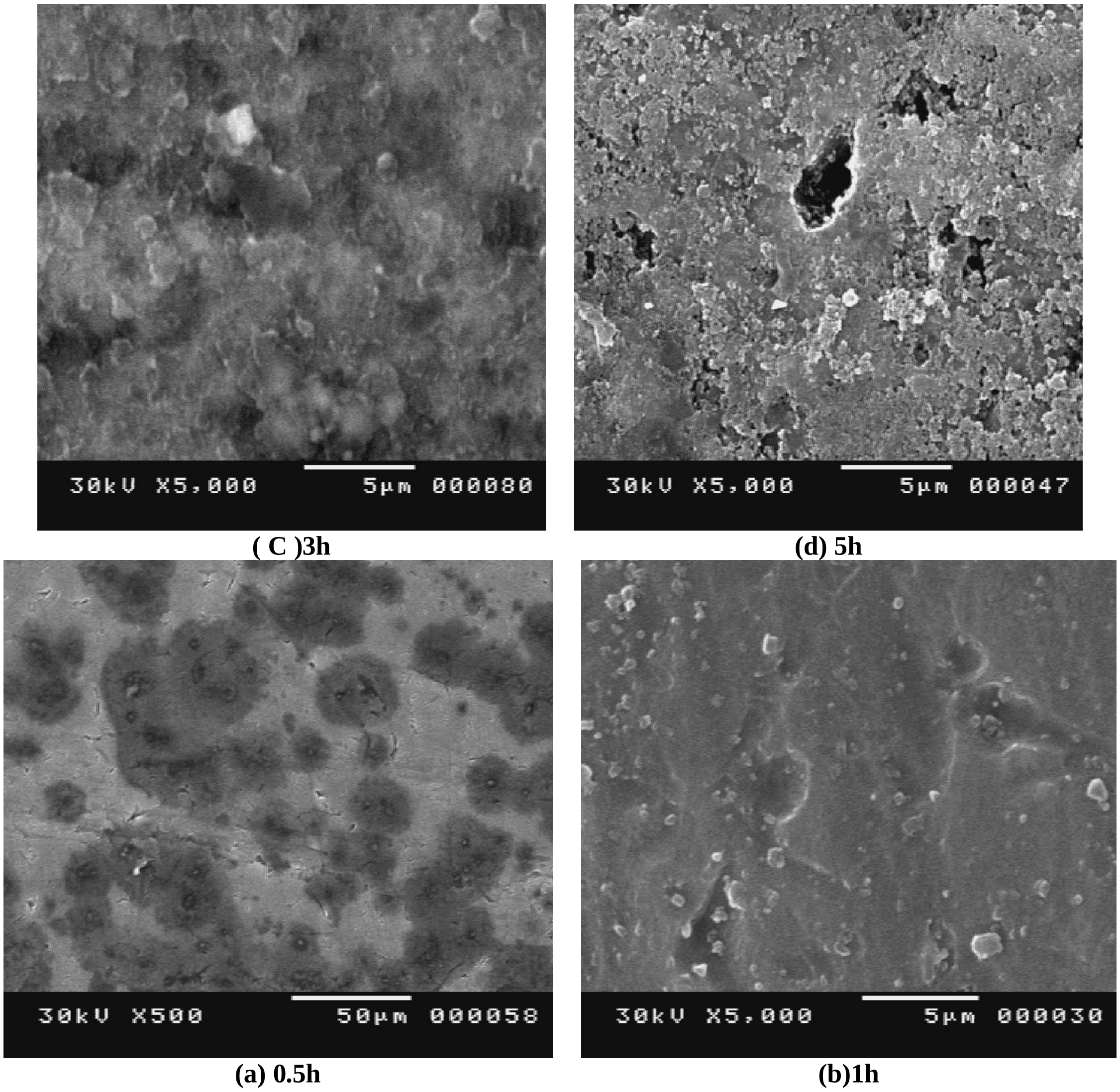
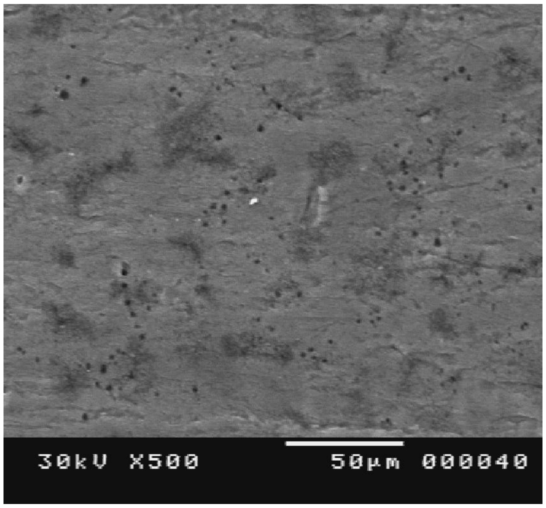
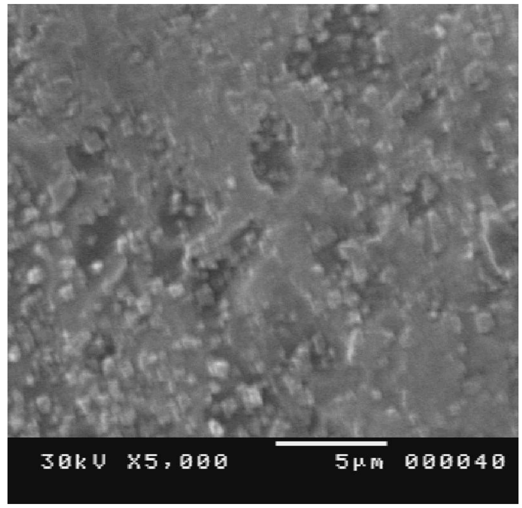


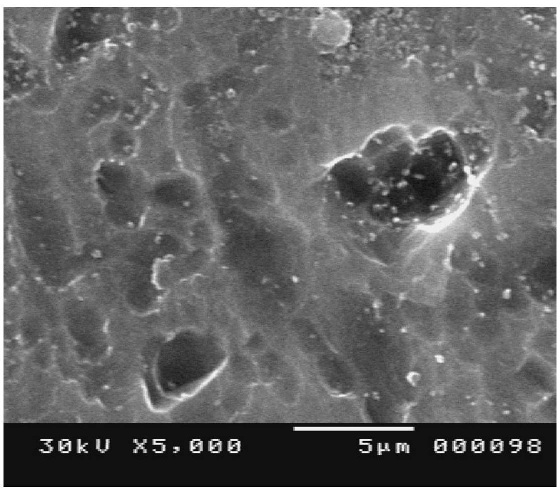
Fig.6 a-d: SEM micrographs of the Al-Mg-Si alloy 0.133 wt% Ce with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C



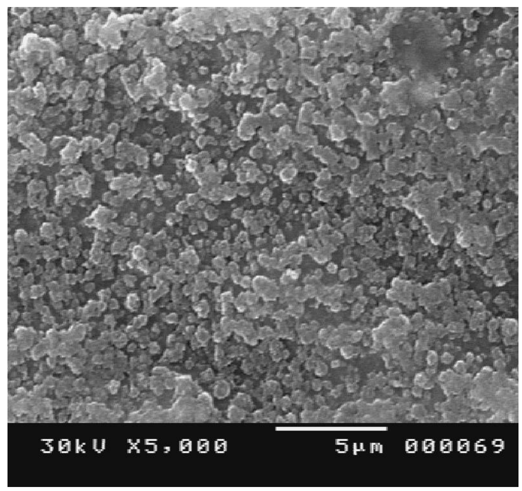
(a) 0.5h



(b) 1h

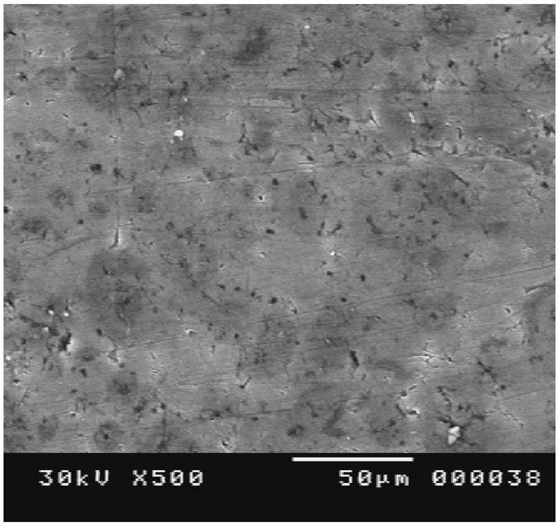


(C) 3h

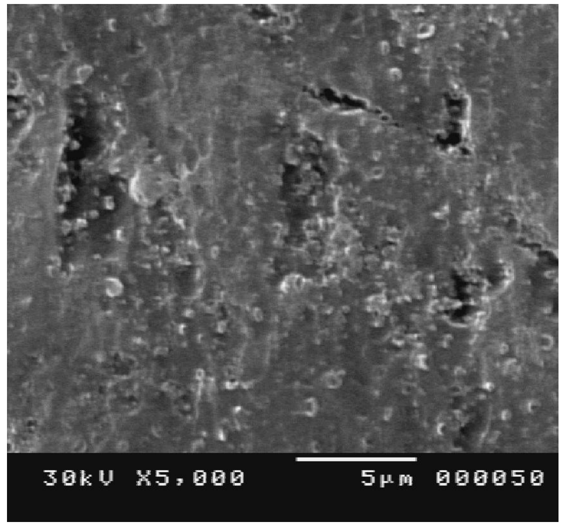


(d) 5h

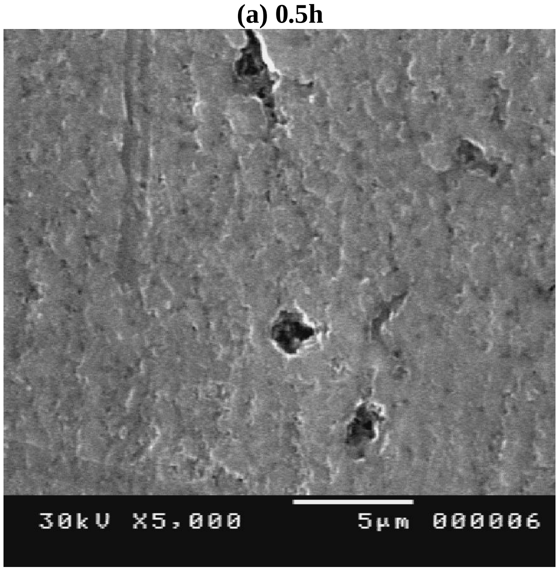
Fig.7 a-d: SEM micrographs of the Al-Mg-Si alloy with 0.166 wt% Ce with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C.



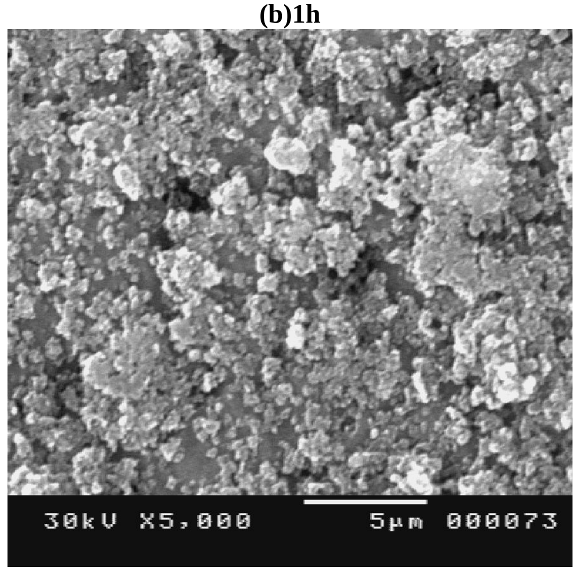
(a) 0.5h



(b)1h



(C)3h



(d) 5h

Fig.8 a-d: SEM micrographs of the Al-Mg-Si alloy with 0.194 wt% Ce with different homogenization time ; (a) 0.5h (b) 1h (c) 3h (d) 5h. at 450^{o}C

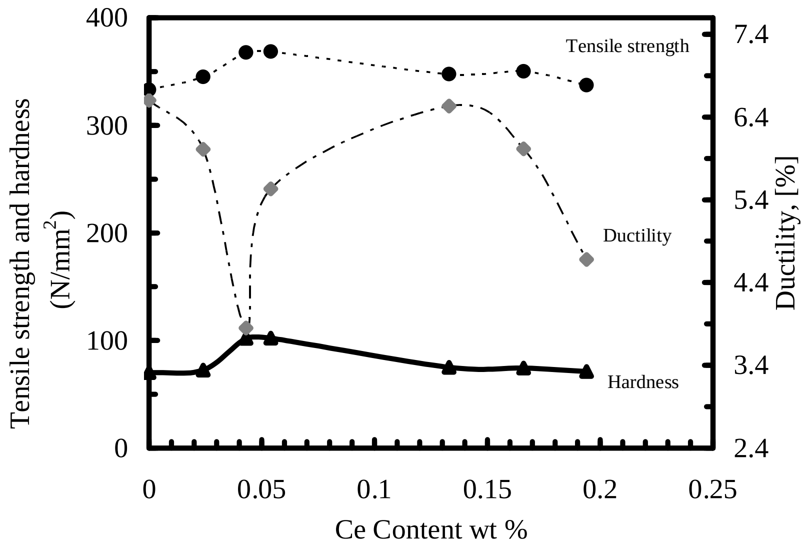


Fig.9: Variation of tensile strength, hardness and ductility as function of cerium content

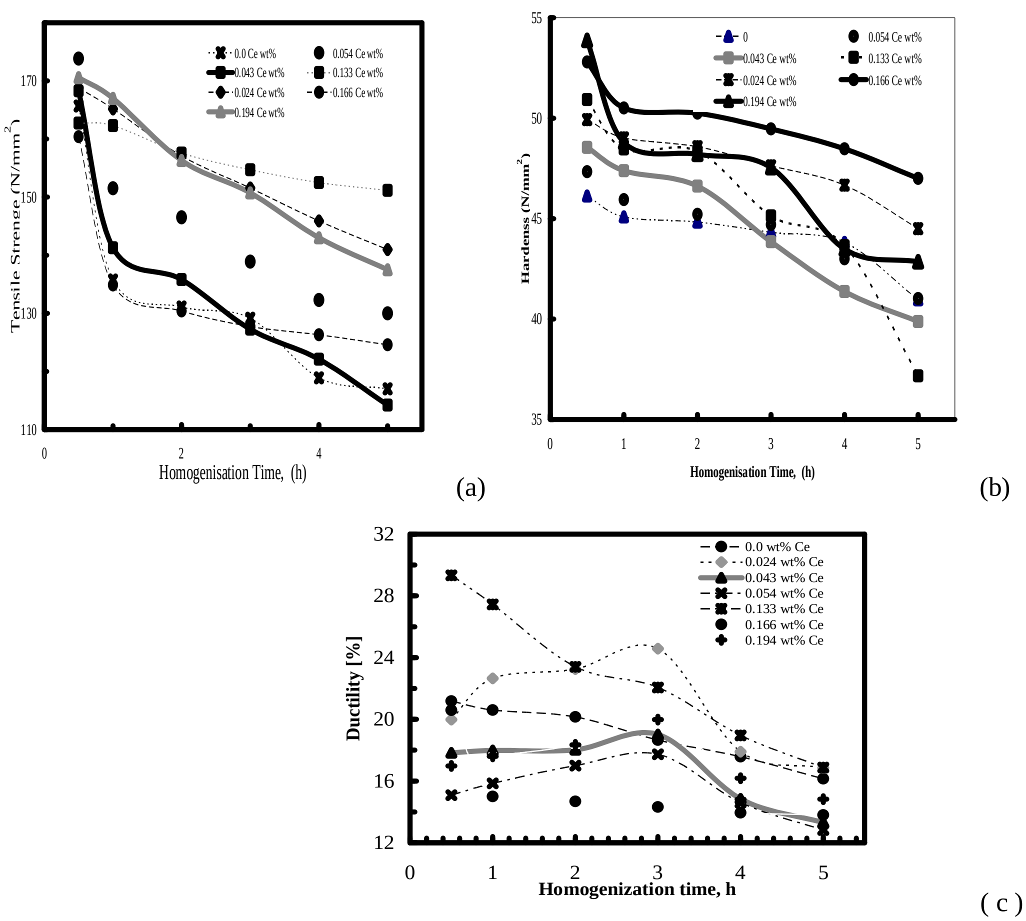


Fig.10 a, b and c: Variation of mechanical properties of Al-Mg-Si alloys; (a) tensile strength, (b) hardness and (c) ductility for different Ce contents as a function solution treatment time at 540°

The increasing in resistivity can be interpreted as; the resistivity is a consequence of disturbances in the atomic periodicity in a crystal structure according to the Block model [21]. These disturbances can be due to atomic vibrations thermal agitation, other electrons, defects in the crystals such as vacancies dislocations or grain boundaries, or substitution of impurity atom in the pure metal lattice sites. The most important of these effects is from the alloying elements in solid solution. The elements such as : Fe, Co, Ni, Sb, La, Ce, Cd, TH, mischmetal "least detrimental to conductivity" forms eutectic phase diagrams at the aluminum-rich end. The eutectic reactions expressed by L à α + β. This is in agreement with the microstructures observed in SEM investigation. The group of elements least detrimental to conductivity form inter-metallic compounds which remain out of solution. The compounds precipitate in the grain boundaries and leave the alpha aluminum almost pure. This helps the alpha to become pure where, in this pure state, it takes the major burden of the electron transport and thus hardly increases the resistivity [22, 23].

Elements in the solution can only be least detrimental to conductivity if they have similar electronic structure to that of aluminum. The rare earth elements, such as cerium have similar electronic configuration, in the solution in an aluminum matrix, produce a very low differential change in the resistivity of aluminum this gives the best bonding with the largest free energy [24]

Table.1-a: Variation of tensile strength, hardness and ductility of Al-Mg-Si alloys for different Ce contents

Ce content (wt%)	Tensile strength	Ductility	Hardness
0	333	6.603	70.18
0.024	345	6.01	72.38
0.043	367.67	3.852	102
0.054	368.65	5.532	102.28
0.133	347.56	6.533	75.133
0.166	350.23	6.015	74.4
0.194	337.436	4.68	71.27

Table1- b:Variation of tensile strength of Al-Mg-Si alloys for different Ce contents as a function of solution treatment time at 540^{o}C

Time	0.0 wt%	0.024 wt%	0.043 wt%	0.054 wt%	0.133 wt%	0.166 wt%	0.194 wt%
0.5	165.666	168.782	168.27	173.828	162.73	160.398	170.55
1	135.74	165.175	141.272	151.51	162.3	134.878	166.988
2	131.1	157.1	135.808	146.513	157.57	130.4	156.293
3	129.16	151.57	127.268	138.883	154.7	127.718	150.72
4	118.9	145.9	122.1	132.28	152.5	126.3	143.01
5	117.015	140.97	114.2	130	151.158	124.58	137.5

Table.1-c: Variation of hardness of Al-Mg-Si alloys for different Ce contents as a function of solution treatment time at 540^{o}C

Time	0.0 wt%	0.024 wt%	0.043 wt %	0.054 wt%	0.133 wt%	0.166 wt%	0.194 wt %
0.5	46.125	49.93	48.543	47.34	50.93	52.8	53.875
1	45.09	49.019	47.386	45.95	48.466	50.507	48.8
2	44.83	48.583	46.614	45.21	48.36	50.25	48.2
3	44.32	47.628	43.85	44.7	45.136	49.467	47.54
4	43.817	46.666	41.37	43	43.657	48.474	43.49
5	40.96	44.5	39.875	41.025	37.17	47	42.85

Also, the solubility is controlled mainly by the ratio of atomic size solvent and solute. If the difference in atomic radii between two elements is less than 15% then there is a large chance of making extended solid solution. In case the difference is greater than 15% solubility is always low [22]. Thus with unfavorable size factor the alloy element will be out of solution generally as a compound. Elements out of solution form small separate particles of low conductivity, but also occupy a very small volume percent of the alloys, and thus have maximum effect on the conductivity. Ce is an example that elements least detrimental to conductivity generally have atomic radii differing widely from that of aluminum.

The low alloy content, the resistivity can be kept reasonably.

For the homogenized samples as seen from Table (2), the general behavior observed is an increasing in resistivity with annealing time. It has been known for a long time that, the resistivity increases nearly linearly with concentration of the alloying elements in solid solution. This explains the increasing in the resistivity at higher homogenization time [26]. The quench itself often produce lattice strain and this is usually considerably increased the electrical resistance [27] as pointed in Table (2).

Table 2: Resistivity (x10^{-6}Ω.cm) for Al-Mg-Si alloys with different Ce content as- received and after solution heat treatment at 540C for different duration time

Conc. wt % / Time .(h)	00.000 Wt%	0.024 wt%	0.043 wt%	0.054 wt%	0.133 wt%	0.166 wt%	0.194 wt%
00	3.49	3.56	3.71	3.46	4.825	3.51	3.50
0.5	3.77	3.63	3.86	3.49	3.72	3.69	3.52
1	3.58	3.69	3.96	3.62	3.51	3.62	3.50
2	3.54	3.84	3.56	3.63	3.90	3.75	3.52
3	3.70	3.53	3.47	3.73	3.49	3.63	3.53
4	3.45	3.71	4.59	3.38	3.72	4.00	3.55
5	3.44	3.83	3.98	4.83	3.76	3.55	3.54

4. Conclusion:

SEM shows that Al- Mg- Si alloy (free Ce) has two kinds of morphologies, Mg_2Si along grain boundaries and round particles that distribute both inside and at grain boundaries. The addition of Ce, a new rod-shaped have an obvious tendency to distribute at grain boundaries and grain refinement the alloys.

All the alloys investigated which contain Ce having the highest strength companied by low ductility compared to alloy free Ce. The improvement of mechanical properties is ascribed to the grain refining. On the other hand, the tensile strength and hardness show a more increase with a suitable Ce addition in the range (0.043 to 0.054 wt%) which have major grain boundaries and the best grain refining, excess of Ce > 0.054 wt% reduce its useful effect. Meanwhile the lowest ductility is due to the formation of insoluble inter-metallic compound in the aluminum matrix.

It is found that Ce form eutectic reaction at the aluminum end of the phase diagram and has electronic structure similar to that of in solution, in an aluminum matrix that due to hardly increases resistivity. Although, the large difference in the atomic radii between Al and Ce but a very low contents are less damaging to conductivity. The quench itself often produces lattice strain and this is usually considerably slightly increased the electrical resistance.

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5. References

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