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RESEARCH ON MICROSTRUCTURE AND MECHANICAL PROPERTIES OF Al-Zn-Mg-Cu ALLOY WHEN MODIFIED BY La, Ce AND THERMO-MECHANICAL TREATMENT

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Abstract

Influence of rare-earth (La, Ce) and thermo-mechanical treatment on microstructure and mechanical properties of Al-Zn-Mg-Cu alloy are presented in this article. After casting, the alloy which was modified by La, Ce, the grain size of samples obtained around 40–50 µm compared to that of without about 65 µm; and after homogeneous, the grain sizes is about 30 µm. After the cold deformation process, the distance between plates is 10 µm. By EDS after casting, the samples have tended to more La, Ce elements at the grain boundary, after homogeneous, the uniformation distribution of rare-earths was presented by mapping of EDS's results. In addition, after rolling and heat treatment, the elements were found on the grain boundary and matrix. After recrystallization annealing, the grain size is around 10 µm with the modification sample. The grain size was reduced by two processes of modification as well as thermal-mechanical treatment is a condition for increasing the ductility of the studied alloy. Further, as a result of ability deformation from the tensile test, these results demonstrate that the tensile test obtained 140 % when adding La, Ce contents into the alloy combine with thermal-mechanical treatment. The combined used of La, Ce and thermal-mechanical treatment have increased the ductility of Al-Zn-Mg-Cu alloy.

Keywords: modification, thermal-mechanical, deformation, grain size, homogeneous, recrystallization annealing, ductility.

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1. Introduction

Superplastic materials were produced as a huge success in the mechanical industry. There are many certainly prominent advantages compared to conventional metals such as the ability to fabricate the thinner parts with the thick more uniformity, the shape more complicated, to save material source, reduction of mass, and increasing the effective use of parts in the case of use and production as well, in details like stamping, metal blowing, and press process. All of them bring to save the cost of tools, producing the durable products with more accurately and highly effective economy.

Materials considered are superplastic in the detail conditions, which is speed and temperature deformation that must be obtained over 200 % in elongation. Aluminum alloys have different characteristics once change alloy components. Therefore, the denaturation is dissimilar with each this type of alloy. The denaturation included two aims, which are to make the fine grain in both matrix aluminum and alloy. Most aluminum alloys do not modification possess in the freezing process but instead formulation of clearly different three area microstructure in which including the outer shell of fine, and then the cylinder crystal layer and in the center of ingots contain the rough crystals. The level of coarse grain and the length of cylinder crystals depend on pouring temperature, the gradient in mold, and the number of nucleations formed in the crystallization process. In the view of principle, then adding the element alloy into liquid aluminum, the grain size is decreased following the rule of the solubility rate: the chemicals have a higher solubility rate, the more grain size is decreased. Alloys that contained many elements a great solubility rate such as Cu, Mg, Zn will easily denature small grain as much as that of alloy included Si content. For instance, by using a modification for Al-Si-Mg-Cu alloy, it can obtain the grain with fine, but opposed to Al-Si with a high amount of Si content, additionally Si is hard to denature [1–3].

In the views of thermodynamic, the recrystallization process included two stages: nucleation and growth of nuclei. The speed of these two phases is critical parameter in the formation of microstructure and properties of alloys. The main purpose of denaturation is to change these two factors. The modification absorption will produce a absorb layer which is cover the established crystals to protect these crystals grow up. The denaturation which produces the foreign particles: when it is doped into liquid aluminum then will react with the available elements that presented in the alloy to form other compounds such as oxide, sulfite. These compounds have a crystal lattice similar to the crystallization phase and consequently, it will become the crystallization nucleus for itself. B, Ti, Zr, C, Sr elements are often used to denature the solid-solution alloys. Once crystallized, these elements were made with the aluminum which forms the phases AlB_2, Al_3Ti, and Al_3Zr these phases are nuclei of crystallization. These compositions do not only increase the strength but also robust the abrasion properties under stress that relied on complicated phases in the grain boundaries. Moreover, it is also employed under intermediate alloy formation [4].

Besides the refining modification applied to make small grain size, it also has the following benefits below: the additional elements for ingots sample in the ultimate stage of the crystallization process were mainly supported based on the presence of enormous nucleation centers causing by refining substances. The coarse porosities were highly increased while the micro-porosity tended to more constant distribution. Additionally, in the crystallization process, there are any gas porosities are formed will be more evenly spread in ingot samples with the fine refining denaturation. The trends of heat cracks in the crystallization process were declined to the lowest peak with the separate areas that growth inability. Consequently, the separation and cracks of grain boundaries will rarely happen. The fine structure will produce more value for the surface layer in anodized. The rough grain of ingot samples is very hard to engineer mechanical and generally, the surfaces are much less worth compared to the fine ingot sample as doing so [5, 6].

The fine grain structure will parently robust the mechanical properties of ingots. However, the alloys contain over 5 % Si which the mechanical traits are slightly enhanced under conditions that ingots must be guarantee to solid or have a certain number of intermetallic phases, such as $FeSiAl_5$ at a certain extent [4, 7].

The generation of super-conductivities with the fine microstructure was improved in both the strength and ductility and especially, strongly increasing the toughness thus it is hard to cause damaging brittle. Once the size is smaller causing the deformation rate is higher, and the temperature of the super-plastic effect is smaller

Due to the small grain size causing the total area of grain boundaries is larger than hindered the strong slip (increasing the strength). When the deformation at high temperature, the bond between grains becomes weaker along with high-density grains (the number of grains was adapted with the slip) that making the slip was caused more facile at the grain boundary. The slip-on grain boundary is defined as a deformation of the polycrystalline materials that is recognized as a reciprocal movement between adjacent particles, they replace the nearby particles leading to increase the particle density follow to the tensile direction and decrease on the horizontal area-this alternative process is characteristic for super-conductivity deformation that seldom found in the normal conditions. The slip process of the particle can cause their movement along to the boundary or in a boundary area [8, 9].

When the metal is normally deformed with a large particle size, the elongation of the test sample is often not uniform, results in the elongation of particles and as can be found on the cross-section of the sample, the number of particles has no decrease. In the view of high plastic deformation, the shape of each particle has less change. If some particles are elongated, the elongation is much smaller compared with the relative elongation of tensile samples. Once the shape of the particles does not change or little change in the high plastic deformation rate this means that there is a substitution of nearby particles, leading to an increase in the grain density in the direction of tensile of sample and decrease on across area which leads to the large of deformation. In the mechanisms of the super-plastic effect, there are two mechanisms as a movement of deviation in grain and diffusion process. These two are interrelated and influence to super-plastic process of study alloy [8, 10–12].

Regarding the model of Ball-Hutchinson [13], the mechanism of grain boundary slip is the gradual creep and displacement of the deflection. At the recrystallization temperature, the deviation was stopped at the grain boundary due to the regulation of deformation rate from the lack of stress concentration. From the insufficient in both stress concentrations and sliding process of deflection are principles for control of deformation rate at high temperature. Mechanical – heat treating is a stage that combined heat treatment and plastic deformation, but impossible to automatically coordinated between deformation, annealing, and cooling processes. If the plastic deformation were conducted subsequent heat treating, this means that is not mechanical-heat treating but is a normal heat treatment plus mechanical pressure. For example, in the case of real plastic deformation that is cool rolling after aging, it is probably results in stiffness feature which leads to increase the strength properties but without effect to the construction of microstructure as phase transformation. The plastic deformation that was performed prior to the heat-treating process does not affect the formation of the last microstructure; hence this combination is also not a type of thermochemical treatment [14–17].

Recrystallization annealing is defined as the main process the ability to regulate the grain size in metal materials after deformation. In addition, after the recrystallization process has finished, the new microstructure formation is not caused by phase transformation, the main cause is the formation of the newly multi-edge particles in metals that cooled deformation. With the Al-Zn-Mg-Cu alloys, the re-crystallization temperature is elected in the range of $230 \div 380 \pm 50$ °C, and ($0.5 \div 2$ h) in time.

This paper presents the results on the influence of La, Ce and thermo-mechanical treatment on the microstructure and mechanical properties of Al-Zn-Mg-Cu alloy. The article also presents the analysis of structure change; phase transformation of this alloy in different states.

2. Material and Methods

2. 1. Materials

The weight of aluminum billet is about 1–1.2 kg. After cutting, the workpiece will be cleaned of dust and dried at a temperature of 200 °C to remove moisture remaining on the surface of the workpiece. Addition of pure metal is required to achieve the desired batch composition. The mixing ratio has been calculated in advance. Pure Zn and Mg are chopped, dried together with aluminum billets before being used for melting.

The modification elements used here are rare earths with the main ingredients being La and Ce. After melting, the alloys get in the mould and analyzed chemical composition. The chemical composition was present in **Table 1.** The sample hasn't rare earth which is denoted M1. The sample has rare earth which is denoted M2.

Table 1

Chemical composition

Sample	Zn	Mg	Cu	Si	Fe	Mn	Cr	La	Ce	Al
M1	5.4	2.2	1.33	≤0.34	≤0.19	0.165	0.018	–	–	Bal.
M2	6.1	2.2	1.74	≤0.34	≤0.19	0.165	0.018	0.155	0.21	Bal.

2. 2. Experimental procedures

The ingots are conducted homogeneous annealing at 480 °C within 16 h to ensure removal of casting microstructure. After the homogeneous process, the patterns are rolled from the thickness of 4 mm to 2 mm. Next, all the samples was recrystallization annealing at 400 °C in 1 h 15.

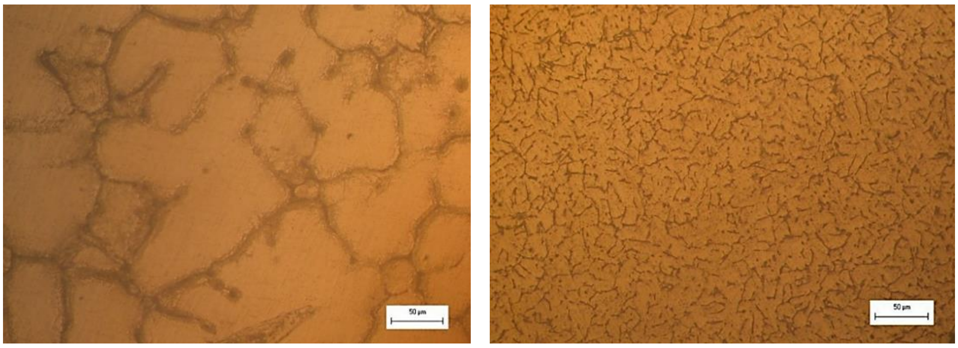
The microstructure of these patterns is investigated by using optical microscopic (Axiovert-25A), and FE-SEM (Jeol JSM-7600F).

The tensile test is applied on the patterns that determined deformation degree at 400 °C, the results were extracted in elongation value. About this experimental, the samples were made by Devotrans (FU/R).

3. Result and Discussion

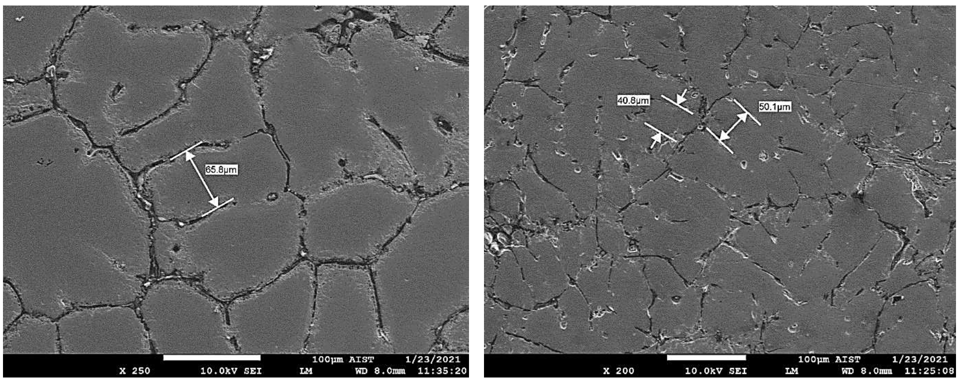
From the micro-optical analysis results (**Fig. 1**): the samples with La, Ce show a smaller grain size compared to the samples without one as rare-earth. The finding on grain size of the rare-earth samples has eight levels, while the samples without rare-earth composition possess at six-level (as the ASM standard). Thus, once the alloys are added to the rare-earth substance, the grain size witnessed a considerable decrease compared to that of without rare-earth compound. This is can be explained by the role of rare-earth into the fine grain of Al-Zn-Mg-Cu alloy. Making the grain allocation more uniform than without La, Ce and no longer exist the high gap of grain size. By using the optical-microscope analysis cannot find the structure and intermetallic phases of rare-earth with elements in alloy's study.

In the **Fig. 2**, while the distance between tree branches of samples without La, Ce was approximately 65 µm, when the samples have rare-earth, the grain size was 10–50 µm, it is worth noting that due to the role of the RE phase that is decreased the size of tree branches.



a *b*

Fig. 1. Microstructure of sample after casting: *a* – M1 sample; *b* – M2 sample



a *b*

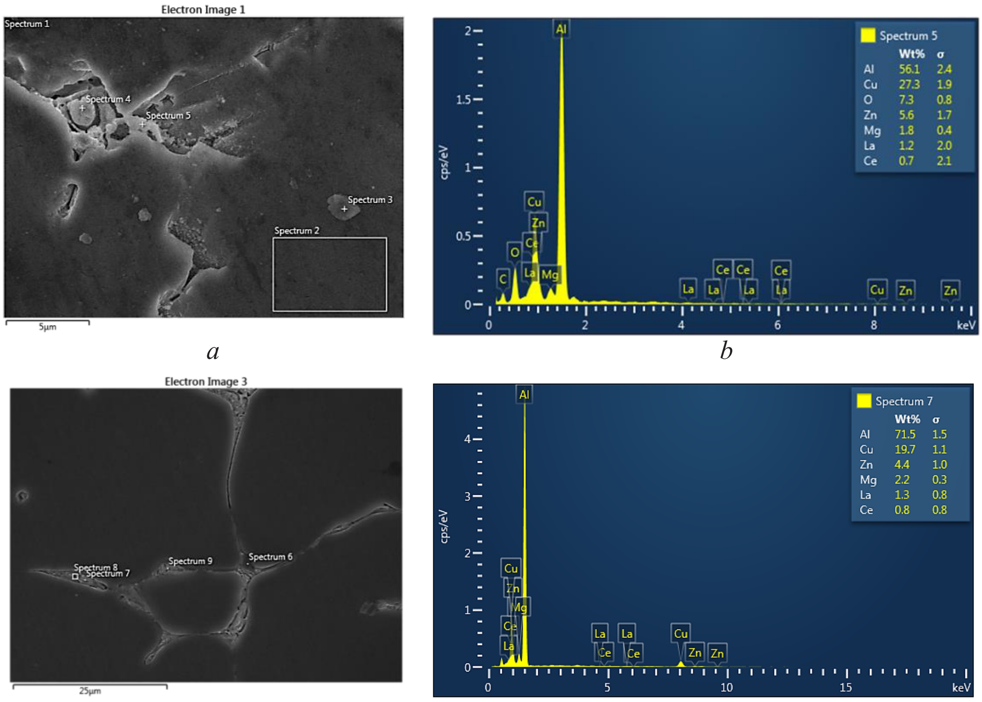
Fig. 2. Microstructure of sample after casting: *a* – M1 sample; *b* – M2 sample

From optical-microscope result of alloys, patterns exhibited the brown color of matrix phase as Al phases, and the black color parts predicted as the intermetallic of Zn, Mg, Cu metals and possible to presence of race earth compounds mixed into the black color phases.

That provement above was confirmed by SEM and EDS analysis at the grain boundary of post-casting as follows below.

Fig. 3 shows the typical peaks of consisting La and Ce in alloys at the grain boundary, manifesting characteristics peaks at 5 and 7. The alloys gathered at the grain boundary that is linked to other elements to form intermetallic phases which is stop the growth of grains after adding the rare earth compound.

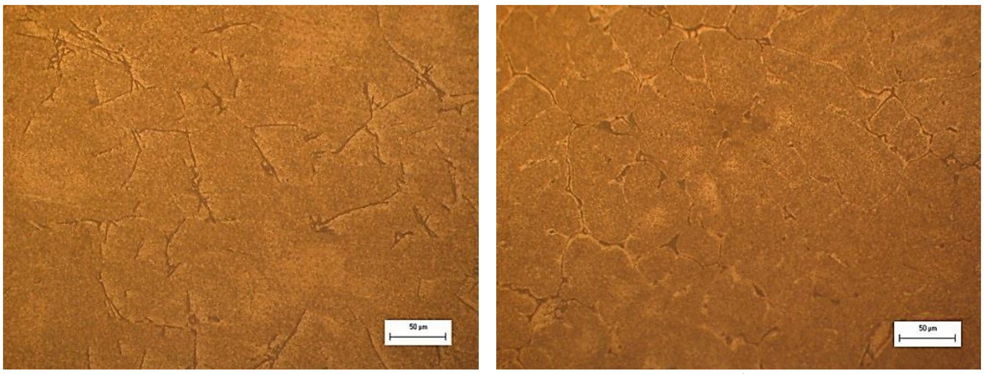
After homogeneous annealing at 480 °C within 16 h it is shown that the grain size has higher than grain size of casting samples, as shown in **Fig. 4**. In the terms of alloys that consisting of La and Ce, the grain level is 7 after homogeneous, while without La, Ce is just over 5. It is worth noting that the microstructure at casting state was eliminated after homogeneous annealing.



a *b*

c *d*

Fig. 3. EDS of M2 sample: *a* – SEM of spectrum 5; *b* – EDS of spectrum 5; *c* – SEM of spectrum 7; *d* – EDS of spectrum 7



a *b*

Fig. 4. Microstructure of samples after homogeneous annealing (OM): *a* – M1 sample; *b* – M2 sample

According to the alloys without La, Ce, the grains are not uniform and relatively large at above 57 µm in **Fig. 5, *b***, and some grains at 20–30 µm range in **Fig. 5, *a***. By contrast, with the alloys consisted La, Ce, there is a medium grain size of 30 µm at the lowest resolution and relatively uniform grain sizes in **Fig. 5, *c, d***. Moreover, the higher resolution shows the uniformation and relatively small grains.

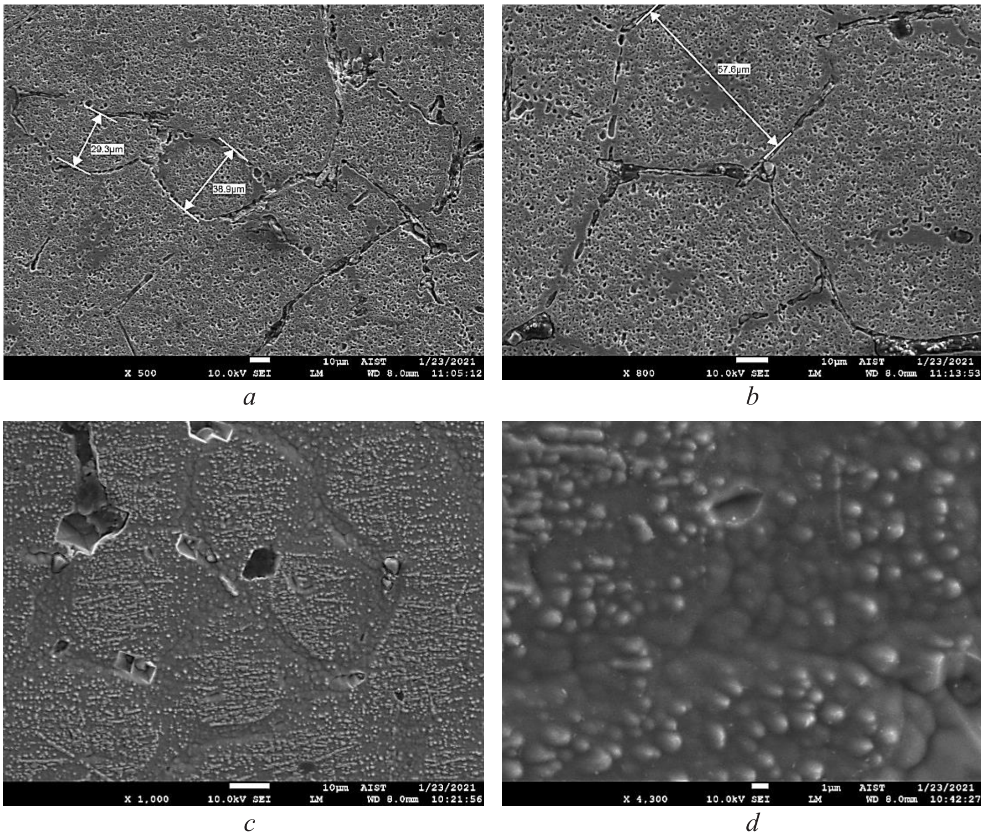


Fig. 5. Microstructure of samples after homogeneous (SEM): *a* – M1 sample (×500); *b* – M1 sample (×800); *c* – M2 sample (×1000); *d* – M2 sample (×4300)

Fig. 6 shows the very uniform of elements on the whole area after homogeneous annealing, which compared with the casting sample. There is La, Ce concentration at the grain boundary. Thereby, the elements were uniformly solid solution on the surface of homogeneous samples.

In **Fig. 7**, the grain size is smaller after homogeneous annealing; with the samples with La, Ce that is evenly distributed of microstructure and without black color phases compared with samples which not included La, Ce. These characteristics are consistent with the analytical results which will be discussed later.

After deformation shows the sample with La, Ce is the smaller distance of vestigial deformation than the sample without La, Ce, as shown in **Fig. 8**. The measurement of EDS lines was indicated the main elements in an alloy that are still existed the uniformity of these elements (**Fig. 9**).

From **Fig. 10**, samples M1 and M2 after deformation and annealing recrystallize, the grain size becomes more uniform. However, with sample M2 the average grain size of the sample was 9.4 µm while with sample M1 the average particle size was above 15 µm. Thus, it can be observed that the influence of La and Ce will greatly affect the grain size. With such a change in particle size, the mechanical properties of the sample will be affected.

Table 2 shows the different degrees at the highest deformation level of the samples with and without La, Ce. While the elongation value of M1 is 11.58 %, the sample with 4 % modification (M2) is 31.92 % that consistent with an increase of 63 %. This may be attributed that although the grain size is not largely decreased the deformation level is high effective. The tensile test result indicates that the effective denaturation process contributed to a significant increase in the deformation degree of alloys.

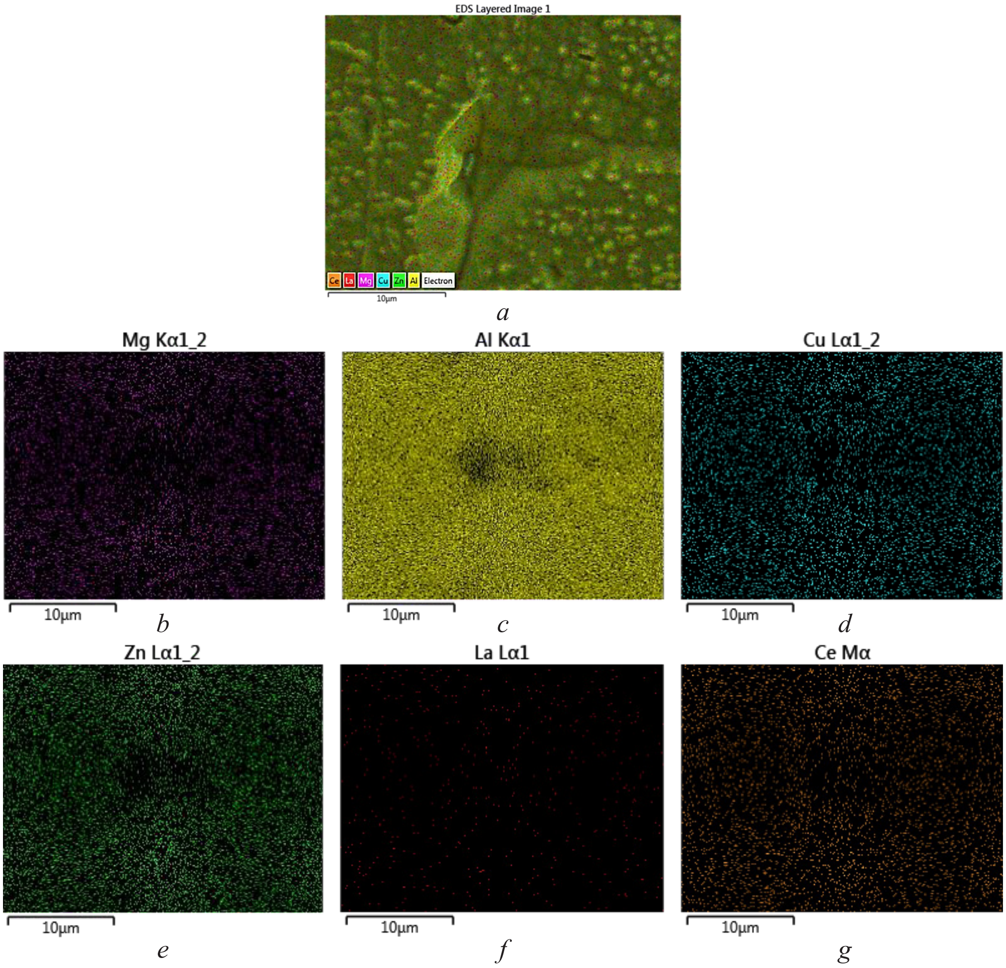


Fig. 6. Element spectrum of M2 sample after homogeneous annealing by Mapping:
a – mapping of all elements; *b* – mapping of Mg; *c* – mapping of Al; *d* – mapping of Cu;
e – mapping of Zn; *f* – mapping of La; *g* – mapping of Ce

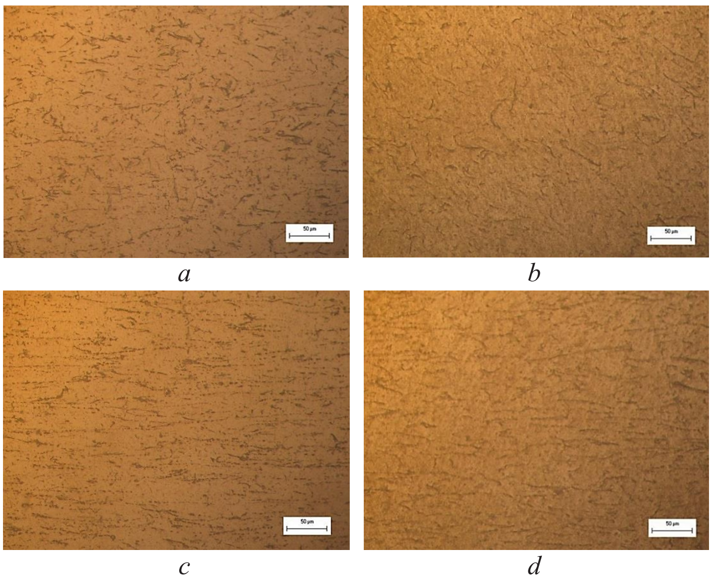
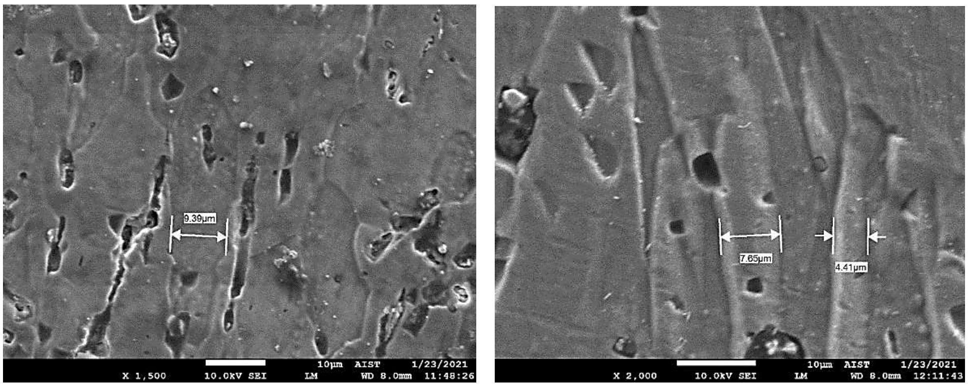
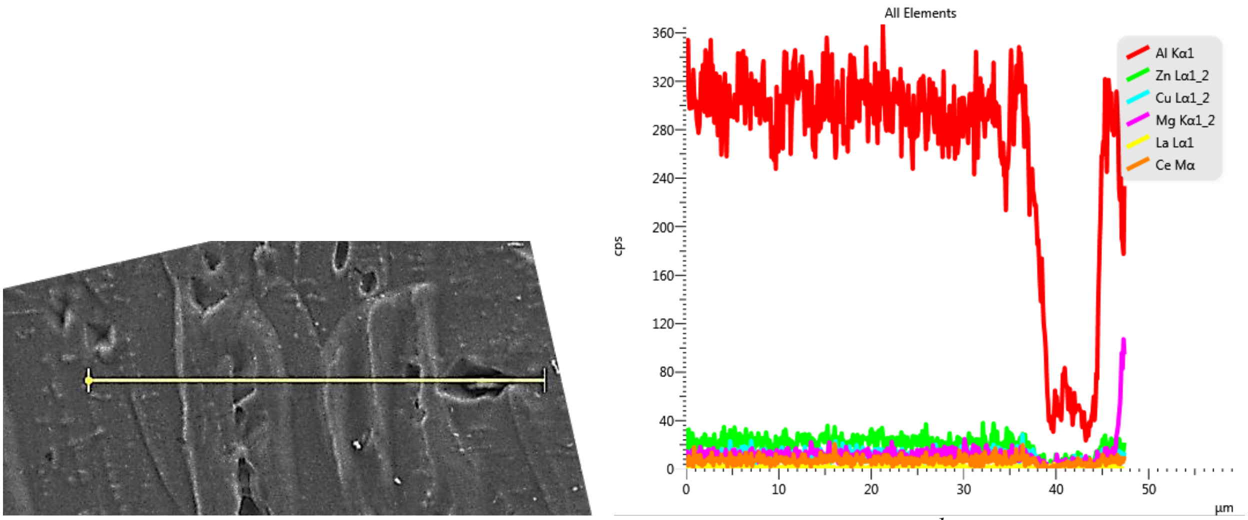


Fig. 7. Microstructure of samples after deformation: *a* – M1 sample (Perpendicular to rolling direction); *b* – M2 sample (Perpendicular to rolling direction); *c* – M1 sample (rolling direction); *d* – M2 sample (rolling direction)



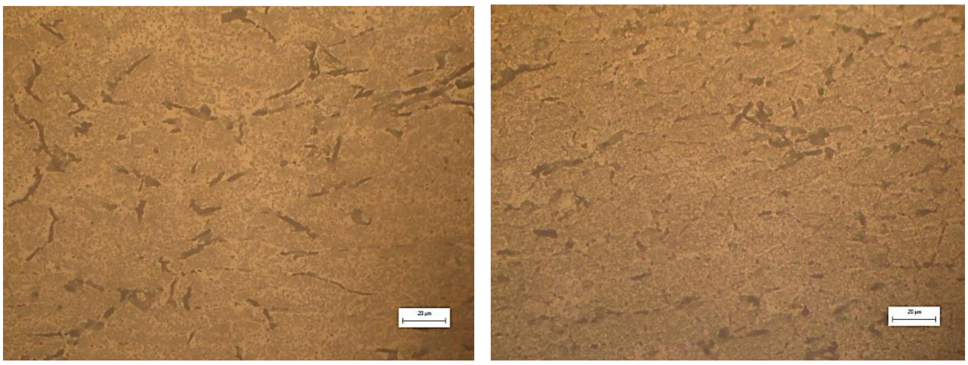
a *b*

Fig. 8. Microstructure of samples after deformation (SEM):
a – M1 sample; *b* – M2 sample



a *b*

Fig. 9. EDS lines of samples after deformation: *a* – SEM of EDS line; *b* – EDS line



a *b*

Fig. 10. Microstructure of sample after deformation and recrystallizion annealing:
a – M1 sample; *b* – M2 sample

In **Fig. 11** based on the results of samples, which are deformation combined with recrystallization annealing alloys, are higher in elongation than the samples with La, Ce, in detail at 140 %. These results are consistent with the research presented above. In additionally, the grain size of deformation combined with recrystallization heating alloys becomes smaller once compared with the samples that no both denaturation and deformation.

Table 2
The result about ductility and strength of samples

Samples	Ductility ε (%)	Limit strength σ_b (MPa)
M1	11.58	39.5
M2	31.92	58.3
M1-deformation (M1-1)	74.9	48.1
M1-deformation-recrystallization annealing at 400 °C in 1 h 15 (M1-2)	82.4	78.1
M2-deformation (M2-1)	121	47.8
M1-deformation-recrystallization annealing at 400 °C in 1 h 15 (M2-2)	140	104

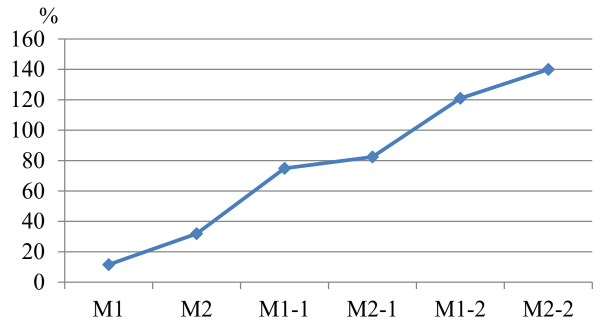


Fig. 11. Diagram of ductility (%)

In **Fig. 12** analysis of strength limit results shows that: the strength limit have the highest value when the M2 is deformed and recrystallization annealing. This value is consistent with the results about microstructure.

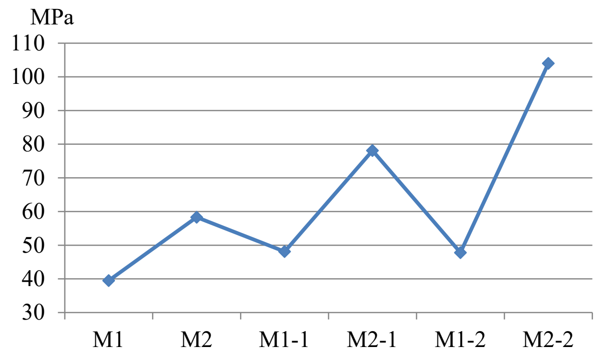


Fig. 12. Diagram of limit strength (MPa)

Although the study has also determined the process of increasing the ductility and strength of Al-Zn alloys. However, in this work, the role of the intermetallic phases on reducing grain size after casting as well as after deformation and heat treatment has not been presented. The article also has not shown the mechanism of the grain reduction after deformation and heat treatment; the optimal thermo-mechanical treatment for Al-Zn alloy has not been determined. These are issues that will be presented in future studies.

4. Conclusions

In this study, the article are determined the transformation of the morphology and structure of Al-Zn once with and without La, Ce elements. Moreover, the change of microstructure of alloys is also presented in this work. The alloy is finely grained with La and Ce combined with homogeneous annealing, the elements are evenly dispersed in the alloy's structure. The grain size of the alloy after thermo-mechanical treatment reaches less than 10 µm, which ensures the ductility of the studied alloy. This results in mechanical properties after homogeneous heating, deformation, and recrystallization annealing indicate that the increase in ductility of the study's alloy.

The article also showed that the maximum elongation of the alloy after thermo-mechanical treatment reached 140 %; the alloy strength value is 104 MPa after deformation and recrystallization annealing at 400 °C for 1 h 15. In addition, the alloys based on La, Ce are also attributed to an increasing deformation.

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