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TEMPERATURE OF PRECIPITATION HARDENING AND ITS EFFECT ON MECHANICAL AND PHYSICAL PROPERTIES OF Zr CONTAINING AlSi5Cu2Mg ALLOY

The paper examines the effect of precipitation hardening temperature on selected properties of AlSi5Cu2Mg alloy alloyed by 0.20 wt.% of Zr. The newly developed AlSi5Cu2Mg alloy intended for cylinder head castings is specific due to its limited Ti content, which prevents the use of standard Al-Ti-B type grain refiners. Zr added in the form of AlZr20 master alloy acts as a grain refiner. The grain refinement effect of Zr positively affects the mechanical properties. However, the physical properties defining the lifetime of cylinder head castings are not affected by the presence of Zr-rich phases. For this reason, the research focuses on the proposal of the optimal T6 heat treatment procedure in order to positively influence the physical and mechanical properties of the AlSi5Cu2Mg alloy. For the research, four T6 thermal regimes with graduated aging temperatures by 20°C from 180 to 240°C ± 5°C were selected. The results showed that increasing aging temperature positively affects physical properties, especially thermal conductivity, and mechanical properties of R_m, $R_{p0.2}$, and HBW. On the other hand, with increasing aging temperature up to 220°C ± 5°C, a negative decrease in ductility was achieved. Optimum ductility of, especially, AlSi5Cu2Mg alloy with 0.20 wt.% Zr was achieved by the T6-240 thermal regime. Optimal combination of thermal conductivity and mechanical properties of the AlSi5Cu2Mg alloy with 0.20 wt.% Zr was achieved by the T6-240 heat treatment due to the requirements placed on cylinder head castings.

Keywords: Mechanical properties; zirconium; microstructure; corrosion; AlSi5Cu2Mg

1. Introduction

Due to the advantageous combination of foundry properties, Al-Si-Cu-Mg alloys are used in the production of highly stressed automotive cylinder head components. Cylinder heads are complex castings that determine engine performance and fuel consumption. In operating conditions, cylinder heads are exposed to mechanical stress, particularly to a wide range of operating temperatures above 200°C. However, conventional Al-Si-Cu-Mg alloys intended for cylinder head castings are mechanically stable up to a temperature of approximately 200°C, the subsequent decrease in mechanical properties affects the functionality and service life of cylinder head castings. The effort of car manufacturers to increase the specific performance of the engine and simultaneously reduce the production of emissions leads to an increase in the temperature of the combustion process, and thus to a direct increase in the operating temperatures of cylinder heads. From this point of view, it is necessary to focus on the development of new aluminum alloys for cylinder head castings [1-3].

One of the possibilities of effectively increasing the properties of Al-Si-Cu-Mg alloys is the alloying of alloys through transition metals such as Zr, Mo, Mn, or Ni. In general, Al-Si alloys intended for use at elevated temperatures must contain an alloying element capable of forming strengthening intermetallic phases, have low solubility in α (Al) solid solution under operating conditions and low diffusivity in α (Al) solid solution. Based on the literature survey, it can be proven that the above-mentioned conditions are met by the transitional element Zr. Zr creates temperature-stable intermetallic phases based on Al_3Zr. The strength of Al-Si alloys with Zr addition achieved by precipitation hardening can be significantly affected by the limited concentration of Zr in the solid solution during solidification. A key prerequisite for preserving the strength properties of Al-Si alloys with the addition of Zr at elevated temperatures is the slow diffusion kinetics of the alloying element Zr. Zr also has a grain refinement effect on Al-Si alloys. Al_3Zr intermetallic phases act as nucleation nuclei for α (Al), thereby influencing the grain size of the primary α (Al) phase. Grain size is an important parameter that mainly affects the mechanical properties of Al-Si alloys.

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Based on the literature survey, it was demonstrated that current research is focused on the development of new Zr-alloyed Al-Si aluminum alloys, whose potential lies in the temperature stability of Zr-rich phases [4-8].

Another significant problem with cylinder heads is heat accumulation due to the complex geometry of the casting. The physical properties of Al-Si alloys greatly affect the functionality and service life of cylinder head castings. The complex geometry of cylinder head castings creates an uneven temperature field. A literature survey has shown that Zr alloying/grain refinement does not have a significant effect on the physical properties of Al-Si alloys. One of the ways to effectively increase the physical properties of Al-Si alloys is to use an optimal heat treatment mode. The optimal mode of heat treatment creates a more uniform temperature field due to the increase in the physical properties of Al-Si alloys [8-9].

The aim of the paper was to analyze the possibilities of increasing the physical and mechanical properties of the AlSi5Cu2Mg alloy with 0.20 wt.% Zr through T6 heat treatment. The AlSi5Cu2Mg alloy, which is used in the production of cylinder head castings, is notable for its limited Ti content. From this point of view, using standard grain refiners of the Al-Ti-B type was impossible. Based on the literature survey, Zr was chosen as an alternative grain refiner, with the aim of achieving stable mechanical properties of AlSi5Cu2Mg at operating temperatures above 200°C [4]. The work analyzed the effect of T6 with a gradual precipitation hardening temperature increase by 20°C from 180 to 240°C (±5°C) on the mechanical and physical properties of AlSi5Cu2MgZr0.2. In general, the correlation between the T6 heat treatment and the physical properties of the Al-Si-Cu-Mg alloy was investigated in the work. With the main goal of increasing the quality and functionality of cylinder head castings due to the creation of a more uniform temperature field.

2. Material and methods

For the experiment, a hypoeutectic AlSi5Cu2Mg alloy with an addition of 0.20 wt.% Zr was used. Zr introduced into the melt in the form of a master alloy AlZr20 was used as an alloying element/grain refiner. The grain refinement effect of Zr is characterized by the formation of intermetallic phases rich in Zr, which act as potential nucleation seeds for the α (Al) phase. The Zr content for the AlSi5Cu2Mg alloy was chosen based on Wang´s studies, according to which the optimal refining effect of the α (Al) phase is demonstrated by the addition of 0.20 wt.% Zr [4]. In his studies, Wang further demonstrated that by adding 0.10 to 0.15 wt.% Zr, a negligible refinement of the α (Al) phase occurs. Also, by adding more Zr 0.25 wt.% Wang did not notice a more significant refinement of the α (Al) phase compared to the grain refinement effect of 0.20 wt.% Zr. The chemical composition of the experimental AlSi5Cu2Mg alloy is shown in (TABLE 1). Based on the chemical composition of the AlSi5Cu2Mg alloy, it can be declared that the alloy was supplied by the manufacturer in a pre-modified state, which is evidenced by the presence of Sr [4,10].

The experimental AlSi5Cu2Mg alloy was melted in an electric resistance furnace. Subsequently, at a temperature of 770°C ± 5°C, a master alloy of the AlZr20 type was added to the melt. The experimental samples were cast using gravity casting technology into a metal mold (Fig.1). The casting temperature was set at 740°C ± 5°C. The experimental alloy was not further modified and degassed in the preparation process.

The experimental samples were subsequently subjected to T6 heat treatment with a graduated precipitation hardening temperature by 20°C from 180°C to 240°C (±5°C). The thermal regime consisted of solution annealing 520°C ± 5°C / 5.5 h, rapid cooling in water at a temperature of 70°C ± 5°C and precipitation hardening 180°C – 240°C ± 5°C / 5 h. For the experiment, four T6 modes were selected, marked as:

- T6-180 – precipitation hardening 180°C ± 5°C / 5 h,
- T6-200 – precipitation hardening 200°C ± 5°C / 5 h,
- T6-220 – precipitation hardening 220°C ± 5°C / 5 h,
- T6-240 – precipitation hardening 240°C ± 5°C / 5 h.

Reference values were provided by the experimental AlSi5Cu2Mg alloy in the cast state, which was designated as the reference alloy.

The methodology for determining the thermal conductivity was based on the measurement of the conductivity of the experimental samples using the Sigma Check 2 measuring device.

TABLE 1

Chemical composition of the experimental alloys

Alloy	Si	Cu	Mg	Sr	Ti	Cr	Zr	Mn/Fe	Al
AlSi5Cu2MgZr0.20	5.40	1.85	0.29	0.008	0.01	0.017	0.19	0.09	Bal.

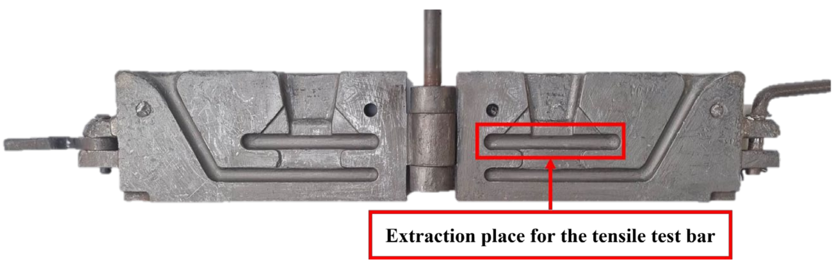

Fig. 1. Metal mold with extraction place for the tensile test bar

By substituting the electrical conductivity values (σ) into the empirical formula (1), the thermal conductivity values (λ) were subsequently calculated:

$$\lambda = 43.29 \cdot \sigma - 13.321\ [\mathrm{W \cdot m^{-1} \cdot K^{-1}}] \qquad (1)$$

The mechanical characteristics (R_m [MPa], $R_{p0.2}$ [MPa], and A_{50} [%]) of the experimental alloys were determined by a static tensile test according to EN ISO 6892-1. The tensile test was performed with a universal tearing device Inspekt desk 50 kN. For each experimental variant (reference alloy, T6-180, T6-200, T6-220, T6-240) a set of 5 test round bars with a shank diameter of 8 mm was made.

The hardness of the experimental alloys was determined by the Brinell hardness test (STN EN ISO 6506-1). The hardness test was performed with a Brinell Innovatest Nexus 3000 hardness tester according to the HBW 5/250/10 regulation. The regulation defines the indentation body as a carbide ball with a diameter of 5 mm, a load value of 250 kp, and a load time of 10 s.

The microstructure of the experimental alloys was evaluated with a Neophot 32 optical microscope and a TESCAN LMU II-line electron microscope with a BRUKER EDX analyzer. The experimental samples were prepared by manual wet grinding, polishing on polishing discs impregnated with diamond paste and moistened with alcohol, and subsequently polished using a fully automatic polishing device. Experimental samples intended for observation with an optical microscope were etched with 0.5% HF for 15 seconds. Subsequently, the experimental samples were subjected to deep etching as a starting point for line electron microscopy REM.

3. Results and discussion

3.1. Physical properties

The values of the thermal and electrical conductivity of AlSi5Cu2MgZr0.2 depending on the heat treatment mode were processed into a graphical dependence Fig. 2. Based on the results, it can be demonstrated that the heat treatment of T6-180 to T6-240 results in an increase in thermal and electrical conductivity compared to the reference alloy. The most significant increase in electrical and thermal conductivity by almost 27% was recorded on the experimental alloy T6-240. The negligible effect of heat treatment on thermophysical properties was demonstrated by increasing precipitation hardening temperature from 180°C to 200°C ± 5°C. The increase in physical properties was attributed to the transformation of the morphology of eutectic Si.

Studies have shown that the shape of the eutectic Si and the size of the secondary phase are decisive factors in the evaluation of the physical properties of Al-Si alloys. The electrical resistance of Si is several orders of magnitude greater than the electrical resistance of Al at room temperature. From this point of view, Si can be considered as an insulator. The electrical resistance of eutectic Si can be significantly influenced by changing the cross-section and size of the secondary phase. The electrical resistance of Al-Si alloys increases with increasing P/S ratio, where P denotes the perimeter of the secondary phase and S denotes the cross-sectional area of the secondary phase. Timple confirmed the correlation between thermal conductivity and the shape and size of eutectic Si or of the P/S ratio of the secondary phase. His conclusions were based on considerations that the P/S ratio of the rectangular cross-section of eutectic Si (lamellas) is lower than the P/S ratio of the circular cross-section of eutectic Si (grains). Timple demonstrated that an unmodified as-cast Al-Si alloy with lamellar eutectic Si exhibits an order of magnitude lower conductivity than an identical heat-treated Al-Si alloy, in which microstructure eutectic Si could be observed in the form of grains (Fig. 3) [11-16].

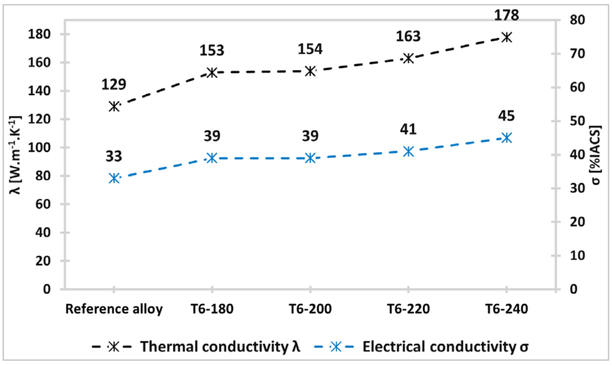

Fig. 2. Impact of varying heat treatment T6 on physical properties of the experimental alloys with Zr addition

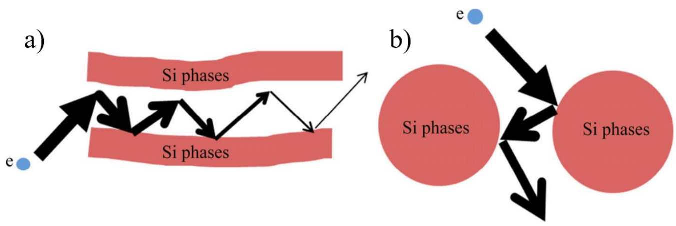

Fig. 3. Transfer of electrons: a) as-cast state, b) after heat treatment [17]

3.2. Mechanical properties

The evaluation of the mechanical properties was based on the comparison of the mechanical properties of the experimental variants T6-180 to T6-240 with the reference alloy. The measured values were processed into a graphical dependence Fig. 4.

Due to the heat treatment of T6-180 to T6-240, there is an increase in the ultimate strength (R_m) compared to the reference alloy in the cast state. The highest increase in R_m by 25% was recorded by the experimental alloy T6-200 compared to the reference alloy. A subsequent increase in precipitation hardening temperature results in a decrease in R_m compared to T6-180 and T6-200. The largest decrease in R_m by 16% was recorded in the experimental alloy T6-240. The agreed yield strength ($R_{p0.2}$) shows a similar course as the strength limit due to the influence of the T6 heat treatment. The maximum value of $R_{p0.2}$ is achieved by applying the thermal regime T6-200, while a 47% increase in $R_{p0.2}$ was recorded compared to the reference alloy.

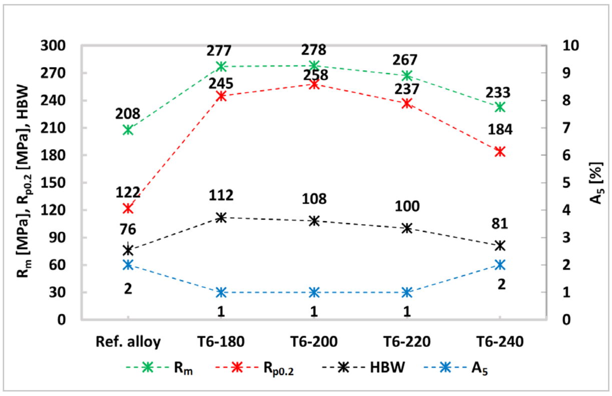

Fig. 4. Impact of varying heat treatment T6 on mechanical properties of the experimental alloys with Zr addition

Increasing the precipitation hardening temperature above 200°C results in a decrease in $R_{p0.2}$. Similar to the evaluation of R_m, the largest decrease in $R_{p0.2}$ compared to T6-200 was observed with the experimental alloy T6-240. The hardness of the experimental alloys increases due to heat treatment. The largest increase in hardness by 32% was recorded by applying the T6-180 thermal mode compared to the as-cast reference alloy. By subsequently increasing the precipitation hardening temperature, the hardness of the experimental alloys decreased. By applying the T6-240 thermal regime, the smallest increase in hardness was recorded compared to the reference alloy of approximately 7%.

The strength limit, agreed yield limit, and hardness of the experimental alloy AlSi5Cu2MgZr0.2 increase due to the presence of strengthening intermetallic phases of Mg_2Si and Al_2Cu. As a result of the higher precipitation hardening temperature (T6-220 and T6-240), strength and hardness decrease due to the slight overaging of the AlSi5Cu2MgZr0.2 aluminum alloy [14,18].

Applying the T6-180, T6-200, and T6-240 thermal modes, a 50% decrease in ductility was recorded compared to the reference alloy. A further increase in precipitation hardening temperature was accompanied by an increase in ductility to the initial value provided by the reference alloy in the as-cast state.

The ductility of alloys intended for cylinder head castings is an important material characteristic that determines the service life and functionality of components working in a wide range of operating temperatures. Alloys commonly used for cylinder head castings have approximately 2% ductility. Heat treatment of the AlSi5Cu2Mg alloy is considered the most effective from that point of view.

Similar to physical properties, mechanical properties could also be influenced by the morphology of eutectic Si. The increase in R_m, $R_{p0.2}$, and HBW as a result of heat treatment T6-180 to T6-220 was accompanied by spheroidization of eutectic Si to a more energetically favorable state of perfectly round grains. Subsequently, the decrease in mechanical properties due to T6-240 could be attributed to the local thickening of eutectic Si. Coarsened eutectic Si particles are characterized by lower strength and hardness [17-20].

3.3. Microstructural analysis

The microstructural analysis of the samples was evaluated by optical microscopy and scanning electron microscopy (SEM) methods. The microstructure of AlSi5Cu2Mg in the as-cast state consists of α-phase, eutectic Si, and intermetallic phases based on Cu and Fe (Fig. 5). Eutectic Si can be observed in the plane of the metallographic cut in the form of imperfectly round grains. This fact indicates that the AlSi5Cu2Mg alloy was delivered from the manufacturer in a pre-modified state. Fe-based intermetallic phases were observed in the form of grey lamellas with split ends. Intermetallic phases rich in Cu can be observed in the plane of the metallographic cut in the form of a ternary eutectic with a compact morphology or isolated particles [21-24].

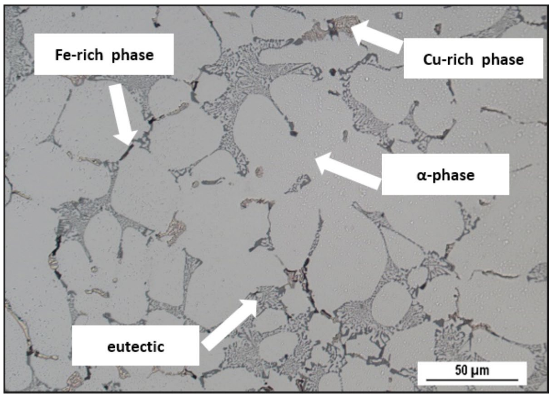

Fig. 5. Microstructure evaluation of reference alloy AlSi5Cu2Mg with Zr addition in the as-cast state (H_2SO_4 etch.)

As a result of the T6 heat treatment, eutectic Si is spheroidized. In Fig. 6a-b, eutectic Si can be observed in the form of perfectly round grains. Increasing the precipitation hardening temperature above 200°C ± 5°C (T6-220 to T6-240) results in a local thickening of the eutectic Si, as can be seen in Fig. 6c-d. Intermetallic phases based on Cu and Fe were observed in the plane of the metallographic cut.

Intermetallic phases rich in Zr can be observed in the plane of the metallographic cut in the form of sharp-edged grains or coarse needles (Fig. 7). By EDX analysis, phases rich in Zr were identified as Al_3Zr and AlSiZr type phases, in the vicinity of which there is an increased concentration of Cu-based intermetallic phases. It follows from the above that Zr creates potential nucleation seeds during the precipitation of Cu phases [4,25].

4. Conclusion

The aim of the work was to analyse the effect of the precipitation hardening temperature on the mechanical and physical properties of the AlSi5Cu2Mg alloy alloyed with 0.2 wt.% Zr, which is intended for cylinder head castings. Based on the obtained results, it can be concluded that:

- as the temperature of precipitation hardening increases, there is an effective increase in thermal and electrical conductivity,

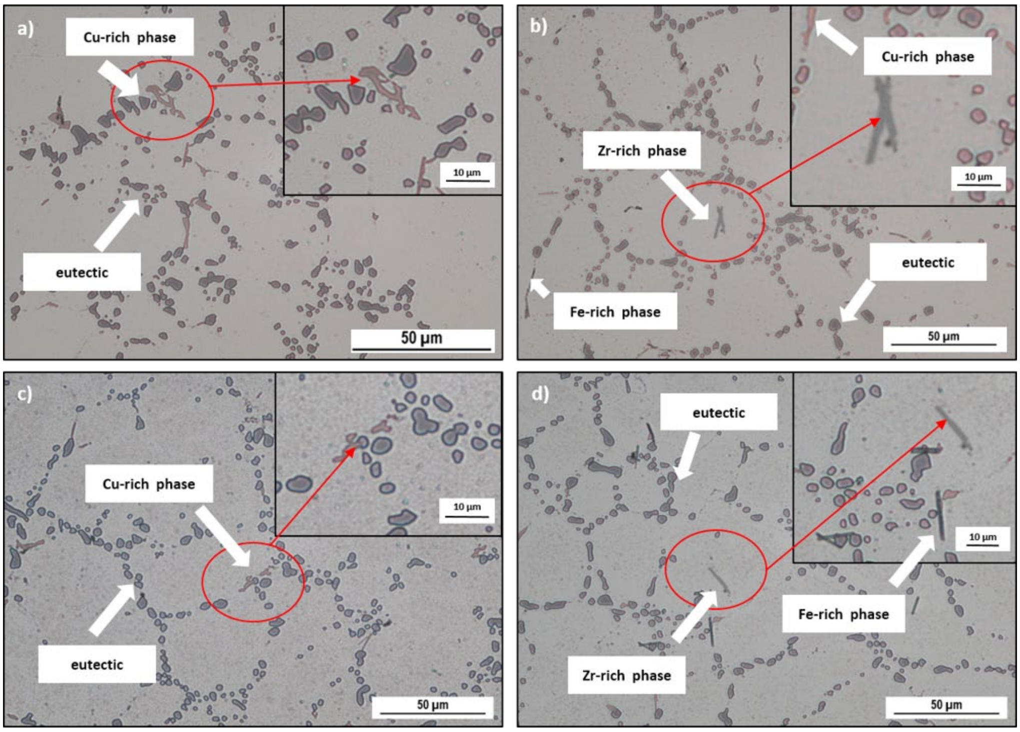

Fig. 6. Microstructure evaluation of experimental alloys after heat treatment T6: a) T6-180, b) T6-200, c) T6-220, d) T6-240 (H_2SO_4 etch.)

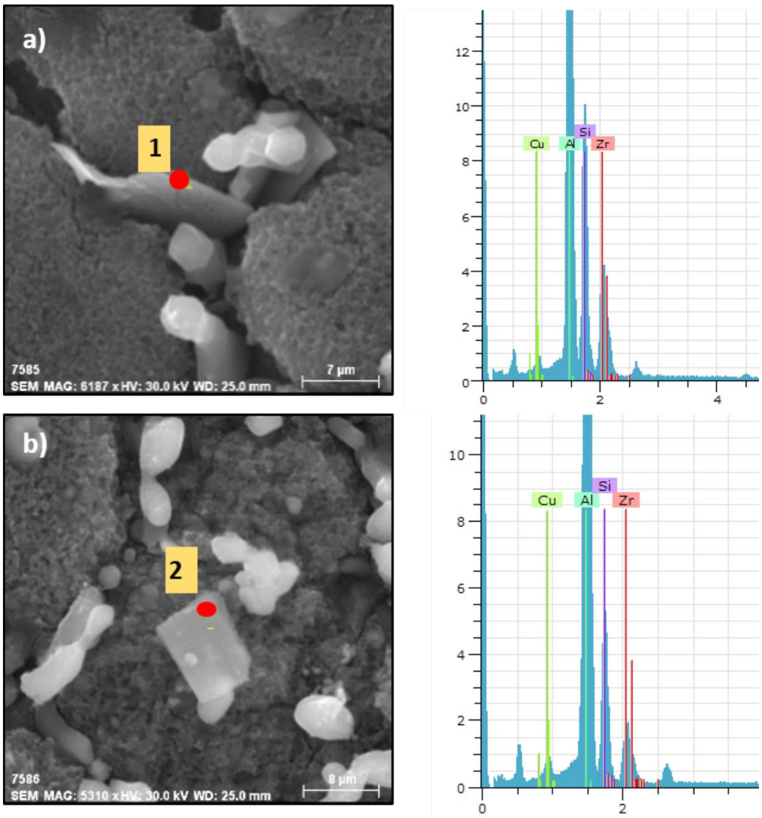

Fig. 7. EDX analysis of Zr intermetallic phases: a) T6-180, b) T6-220

- the increase in thermal and electrical conductivity occurs due to the change in the morphology of eutectic Si to a more energetically favourable state,
- R_m, $R_{p0.2}$ and HBW of the experimental alloys increases up to the aging temperature of 200°C ± 5°C (T6-200), with the subsequent increase in the precipitation hardening temperature, there is a slight decrease in the mechanical properties due to the overaging of the AlSi5Cu2MgZr0.2 alloy,
- the ductility of the experimental alloys decreased by applying the thermal modes T6-180, T6-200, T6-220,

- applying the thermal mode T6-240, the ductility increased to the initial value achieved by reference alloy in the cast state,
- with increasing aging temperature (T6-220, T6-240), local thickening of eutectic Si can be observed,
- increasing precipitation hardening temperature did not affect the morphology of the present intermetallic phases rich in Cu, Fe and Zr.

By applying the T6-240 heat treatment, the most favorable combination of mechanical properties (ductility) and thermal conductivity of the AlSi5Cu2Mg alloy with an addition of 0.2 wt.% Zr, considering the required properties of cylinder head castings. Research has shown that the thermal conductivity of alloys based on Al-Si-Cu-Mg can be influenced to a large extent through heat treatment. By designing an optimal heat treatment mode, it is possible to increase the quality and functionality of cylinder head castings, due to the elimination of faults caused by heat accumulation in a geometrically complex casting.

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REFERENCES

[1] K. Knipling, D. Dunand, D. Seidman, Int. J. Mater. Res. **97**, 246-265 (2022). DOI: https://doi.org/10.1515/ijmr-2006-0042
[2] A.R. Farkoosh, X.G. Chen, M. Pekguleryuz, Mater. Sci. Eng. **620**, 181-189 (2015). DOI: https://doi.org/10.1016/j.msea.2014.10.004
[3] W. Chao, L. Guang-lei, W. Hao, L. Yu-shan, S. Nai-chao, High Temp. Mater. Proc. **37**, 289-298 (2018). DOI: https://doi.org/10.1515/htmp-2016-0199
[4] F. Wang, D. Qui, Z. Liu, J. Taylor, M. Easton, M. Zhang, Acta Mater. **61**, 5636-5645 (2013). DOI: https://doi.org/10.1016/j.actamat.2013.05.044
[5] D. Bolibruchová, L. Širanec, M. Matejka, Mater. **15**, (2022). DOI: https://doi.org/10.3390/ma15144798
[6] M.H. Daneshifar, M.H. Sabzaver, H. Frederiksson, Int. J. Miner. Metall. Mater. **21**, 980-989 (2014). DOI: https://doi.org/10.1007/s12613-014-0999-1
[7] P.D. Staublin, MSc Thesis. (2019). DOI: https://doi.org/10.37099/mtu.dc.etdr/914
[8] M. Rahimin, S. Amirkhanlou, P. Blake, S. Ji, Mat. Sci. Eng. **721**, 328-338 (2018). DOI: https://doi.org/10.1016/j.msea.2018.02.060
[9] Z. Dong, J. Wang, Z. Guanz, P. Ma, P. Zhao, Z. Li, T. Lu, R. Yan, Metals. **11**, (2021). DOI: https://doi.org/10.3390/met11091450
[10] https://www.intechopen.com/chapters/72771 (accessed on 1.5.2022)
[11] W. Weng, H. Nagaumi, X. Sheng, W. Fan, X. Chen, X. Wang, Light Met. 193-198 (2019). DOI: https://doi.org/10.1007//978-3-030-05864-7
[12] M. Timple, N. Wanderka, B.S. Murty, J. Banhart, Acta Mater. **58**, 6600-6608 (2010). DOI: https://doi.org/10.1016/j.actamat2010.08.021
[13] X. Zhang, Y. Zhou, G. Zhong, J. Mater. Sci. **57**, 6428-6444 (2022). DOI: https://doi.org/10.1007/S10853-022-07045-7
[14] E. Vandersluis, C. Ravindran, JOM. **71**, 2072-2077 (2019). DOI: https://doi.org/10.1007/s11837-019-03376-0
[15] P.J. Oliveira, M.L.N.M. Melo, R.S.M. Silva, D.O. Caixeta, Mater. Sci. Forum. **930**, 400-404 (2018). DOI: https://doi.org/10.4028/www.scientific.net/MSF.930.400
[16] E. Vandersluis, C: Ravindran, M. Bamberger, J. Alloys Compd. **867**, (2021), DOI: 10.1016/j.jallcom.2021.159121
[17] E. Vandersluis, P. Emadi, B. Andilab, Metall. Mater. Trans. **51**, 1874-1886 (2020). DOI: https://doi.org/10.1007/S11661-020-05650-2
[18] Ch. Zhong-wei, H. Zhi, J. Wan.qi, Trans. Nonferrous Met. Soc. China. **19**, 410-413 (2009). DOI: https://doi.org/10.1016/j.actamat.2013.05.044
[19] M. Ibrahim, M. Abdelaziz, A. Samuel, Mater. Sci. Eng. **2021**, (2021). DOI: https://doi.org/10.1155/2021/6678280
[20] T.E. Quested, Acta. Mater. **53**, 1323-1334 (2005). DOI: https://doi.org/10.1016/j.actamat.2004.11.024
[21] D. Bolibruchová, L. Richtárech, M.S. Dobosz, K. Major-Gabryś, Archiv. Metall. Mater. **62**, 339-344 (2017). DOI: https://doi.org/10.1515/amm-2017-0051
[22] M. Matejka, D. Bolibruchová, Arch. Foundry Eng. **18**, 25-30 (2018). DOI: https://doi.org/10.24425/123627
[23] E. Tillová, M. Chalupová (2019). ISBN: 978-80-554-0088-4
[24] M. Timple, N. Wanderka, B.S. Murty, J. Banhart, Acta Mater. Ultramicroscopy **111**, 697-700 (2011). DOI: https://doi.org/10.1016/j.ultramic.2010.12.023
[25] M. Timple, N. Wanderka, R. Schlesiger, T. Yamamoto, D. Isheim, G. Schmitz, S. Matsumura, J. Banhart, Ultramicroscopy **132**, 216-221 (2013). DOI: https://doi.org/10.1016/j.ultramic.2012.10.006