

Effect of heat treatment conditions on microstructure and wear behaviour of Al₄Cu₂Ni₂Mg alloy

Erdem KARAKULAK, Muzaffer ZEREN, Ridvan YAMANOĞLU

Metallurgical and Materials Engineering Department, Kocaeli University, 41380 Kocaeli, Turkey

Received 29 August 2012; accepted 5 November 2012

Abstract: Al₄Cu₂Ni₂Mg alloy is an age-hardenable aluminum alloy. The effect of different solution and aging heat treatment conditions on the microstructure, hardness and wear resistance of the alloy was studied. The cast specimens were solution treated and then artificially aged. Optical microscopy and scanning electron microscopy were used to investigate the microstructures of the specimens. The hardness and wear tests were applied to understanding the effects of heat treatment. After aging for 8 h, the hardness of the alloy increases from HV₁₀ 96.5 to 151.1. Aging treatment for a longer duration causes a drop in the hardness because of over aging. Increasing the hardness of the alloy increases the wear resistance. As a result of all tests, solution heat treatment at 540 °C for 8 h and aging at 190 °C for 8 h were chosen for optimum heat treatment conditions for this alloy.

Key words: Al–Cu–Ni–Mg alloy; piston alloy; artificial aging; hardness; wear

1 Introduction

Aluminum alloys have a wide diversity of industrial applications because of their high specific strength, low density and corrosion resistance. Therefore, these alloys motivate considerable interest to the transportation and aviation industries [1–7]. Decreasing the mass of the construction in transportation industries improves fuel economy in internal combustion engines as well as reduces the CO₂ emissions to the atmosphere. Because of these facts, grey cast iron has been substituted by aluminum alloys especially in the engines of the automobiles [8–10]. Some parts like engine blocks and cylinders are already being produced using aluminum alloys by most of the automobile producers.

Al–Cu alloys are normally used in making cast elements designed for structural components. Copper addition increases the strength of the alloy [11]. Copper is a potent precipitation agent in aluminum. Cu addition up to about 5% (mass fraction) leads to alloys with high strength and good toughness [12]. The Al₂Cu precipitates which exist after aging process and are named as θ phase provide these superior mechanical properties [13]. The solubility of copper in aluminum increases with increasing temperature. At elevated temperatures Al₂Cu

phase dissolves and copper atoms enter into aluminum unit cells to create a solid solution. The dissolution of Al₂Cu precipitates in the alloy leads to a dramatical drop in the strength of the material. To avoid this problem precipitates with different chemical compositions which can withstand elevated temperatures are needed. Most important intermetallic phases which can be stable at high temperatures contain Ni and Mg. The chemical compositions of these intermetallics were reported as Al₆Cu₃Ni, Al₂CuMg, Al₇Cu₄Ni, Al₁₂Cu₂₃Ni, Al₃CuNi and Al₃Ni by the previous researchers [13–18].

The control of the microstructural properties like size, chemical compositions and distribution of these precipitates has an important role in the properties of the final product [19]. The difference in the chemical compositions of the intermetallic phases is caused by the chemical composition of the alloy and the thermal history of the material. The production process parameters such as casting conditions and applied heat treatments have major effects on the composition and distribution of the intermetallic phases. Since the presence and properties of the precipitates affect the properties of the final product, the heat treatments have a critical importance to the properties of materials.

The aim of this study is to understand the effect of aging treatment on the microstructure and wear

behaviour of Al₄Cu₂Ni₂Mg alloy which is a candidate material for parts operating at high temperatures in automotive, marine and aircraft industries due to its excellent properties [20].

2 Experimental

Pure metals were used to produce Al₄Cu₂Ni₂Mg cast alloy. Casting process was carried out using an induction furnace with a graphite crucible. Molten metal was poured in a metal mould. Optical emission spectrometer (OES) was used to obtain the chemical composition of the alloy. The composition of the cast alloy is given in Table 1. To understand the effect of heat treatment duration, different heat treatments were applied to the specimens. The solution treatments were carried out at 540 °C for 4, 6 and 8 h respectively. The specimens were quenched in room temperature water after solution treatments. Following quenching the specimens were aged at 190 °C for durations from 6 to 10 h.

Optical microscopy and scanning electron microscopy (SEM) were used to investigate the microstructures of the specimens. The specimens were metallographically prepared with usual manner with a final polishing with 3 µm diamond. Keller's reagent was

used for etching to reveal the microstructure of the specimens.

Future-Tech Vickers hardness tester was used to understand the effect of heat treatments on the hardness of the alloy. All given hardness values are the average of 5 measurements. All sepecimens were metallographically prepared before wear tests. Wear tests were conducted on a Nanovea pin-on-disc type tribometer at room temperature, using 10 N normal load and AISI 52100 steel ball of 5 mm in diameter was used as a counter surface. Sliding distance was kept constant at 1 km for all tests. The specimens were cleaned with alcohol and then dried with a hot air blower before and after tests. The wear loss data of the cleaned samples were obtained using a microbalance with 0.1 mg resolution.

3 Results

3.1 Microstructure

Micrographs of as-cast Al₄Cu₂Ni₂Mg alloy are given in Fig. 1. Figure 1 shows that the Al–Cu eutectic phase surrounds the α (Al) dendrites. When Al₂Cu precipitates are coarse and placed along grain boundaries, the effect of copper on the hardness and strength of the alloy is very limited. Also the fragility of hard Al–Cu precipitates in grain boundaries affects the toughness of the alloy negatively. Besides Al₂Cu, some other intermetallic phases were reported in the microstructure of the alloy. These intermetallic phases were formed by other alloying elements (Mg, Ni) during solidification. It was expected to dissolve these phases in α (Al) solid solution during solution treatment.

Table 1 Chemical composition of Al₄Cu₂Ni₂Mg alloy used in experiments (mass fraction, %)

Cu	Ni	Mg	Si	Ti	Al
4.21	1.98	1.55	0.6	0.01	Bal.

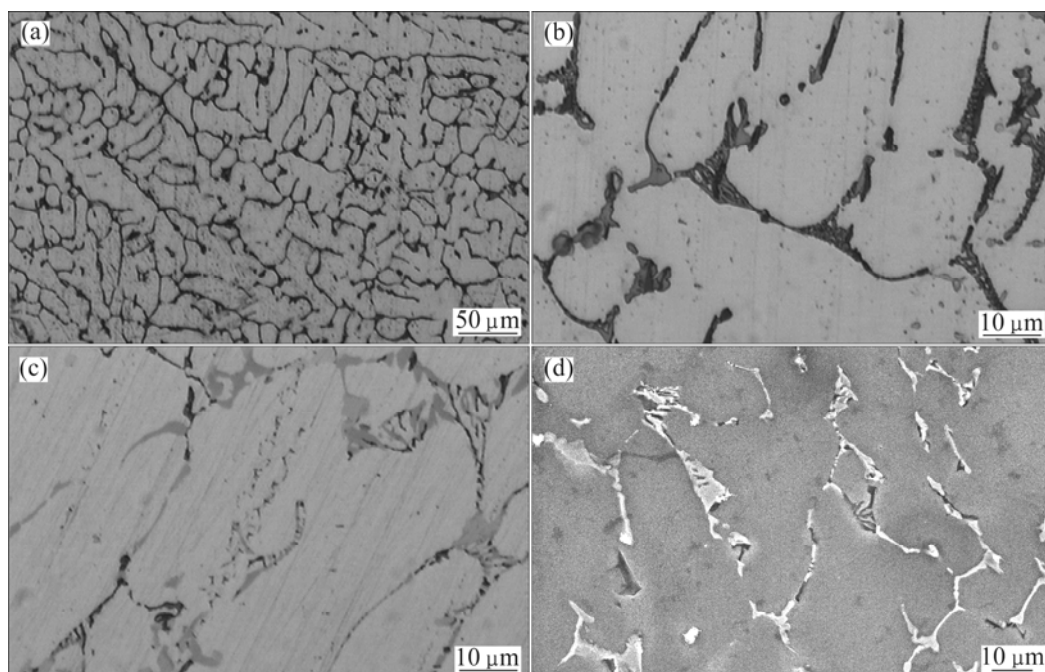


Fig. 1 OM (a, b, c) and SEM (d) images of as-cast Al₄Cu₂Ni₂Mg alloy

Figure 2 shows the microstructures of the specimens solution treated for different durations. It is obvious that increasing solution heat treatment duration causes the dissolution of more copper atoms in $\alpha(\text{Al})$ grains. In the microstructure of the specimen treated for 4 h some undissolved Al_2Cu phases in the grain boundaries can be seen. The amount of undissolved Al_2Cu phase is decreased in the specimens solution treated for 6 and 8 h. There are still some undissolved Al_2Cu at the grain boundaries. But prolonging solution treatment durations may cause grain growth. The increase in the size of $\alpha(\text{Al})$ grains causes a decrease in the mechanical properties of the alloy. Some undissolved phases were reported in the solution treated microstructures of the alloys. These

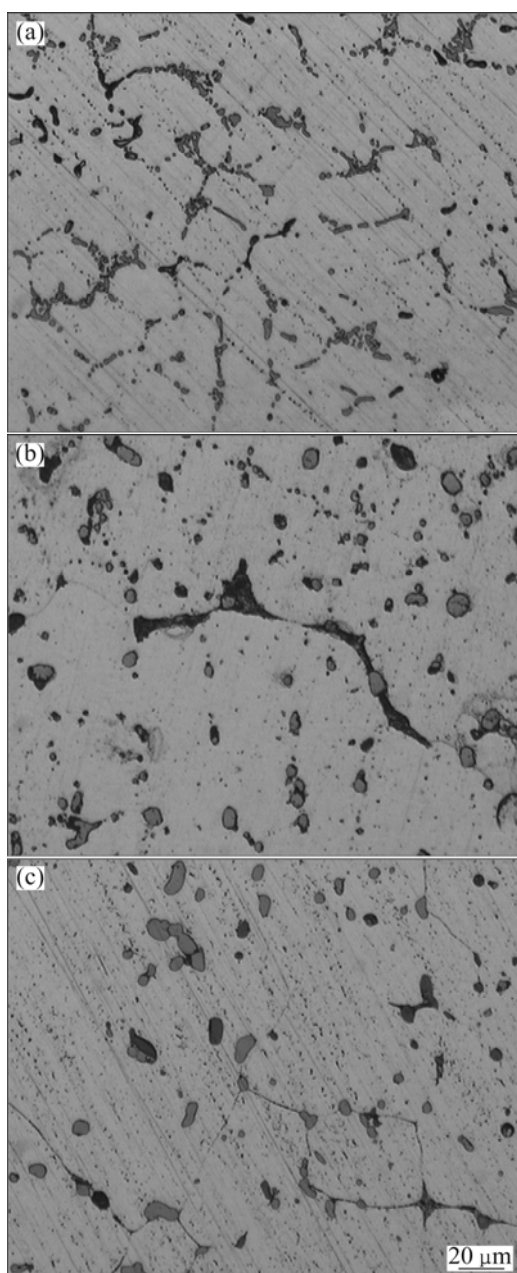


Fig. 2 Microstructures of specimens solution heat treated at 540 °C for different durations: (a) 4 h; (b) 6 h; (c) 8 h

phases are stable at elevated temperatures. Instead of being dissolved in aluminum grains, their morphology becomes spherical during heat treatment. The chemical compositions of the undissolved intermetallic phases were determined by EDS analysis and are given in the relevant section of this study.

Figure 3 shows the microstructures of the specimens aged for different durations at 190 °C after 8 h of solution treatment. Aging causes precipitation of small Al_2Cu phases from the supersaturated $\alpha(\text{Al})$ grains. These precipitates interact with the existing dislocations in the material. This interaction impedes dislocation movement and increases the hardness and strength of the alloy. In the specimen aged for 6 h small precipitates can be seen on the microstructure. Increasing aging time to 8 h causes more small precipitates. But when the specimens are aged for 10 h the number of the precipitates decreases and the size of the precipitates increases. This phenomenon is called over aging, resulting in small number of coarse precipitates. Decreasing the number of precipitates leads to less obstacles for dislocations and causes a dramatical drop in hardness and strength of the material.

3.2 EDX analysis

Energy dispersive X-ray (EDX) analysis was used to determine the chemical compositions of different intermetallic phases in the as-cast and heat treated samples.

Figure 4 shows the SEM image and EDX spectrum taken from the as-cast specimen. The chemical compositions of the phases in the microstructure of as-cast sample are given in Table 2. The results show that $\alpha(\text{Al})$ matrix can solve nearly all of the Mg in the alloy but can only solve a limited amount of Cu. There is no nickel solved in $\alpha(\text{Al})$ matrix, because the solubility of nickel in aluminum is nearly zero at room temperature [21]. Nickel atoms form intermetallic phases with other alloying elements and exist especially at the grain boundaries of $\alpha(\text{Al})$ dendrites. The morphology and size of these intermetallics are related to the chemical composition of the alloy, solidification rate and heat treatments applied to the alloy. It is reported that phases with high nickel content have angular morphology, while the phases with high content of copper have spherical morphology.

Figure 5 shows SEM image of the specimen solution treated for 8 h and Table 3 shows the EDX analysis results of the phases in the microstructure. Most of the phases at the grain boundaries are dissolved after solution treatment. But some of the phases remain unaffected after solution heat treatment. The EDX analysis shows these phases having high nickel content.

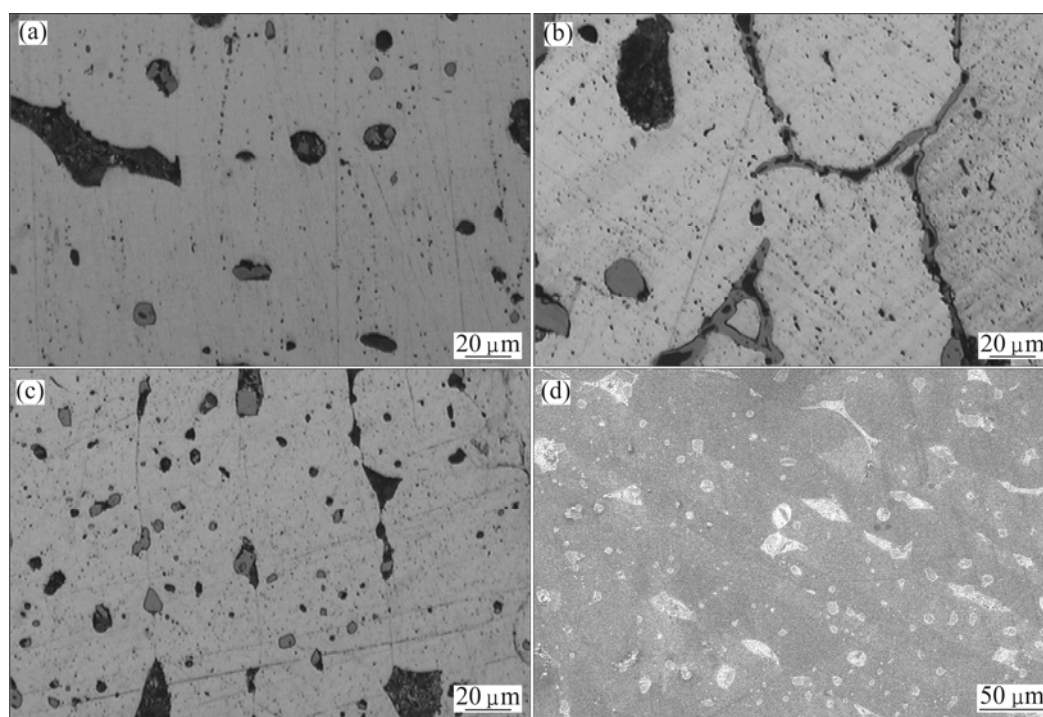


Fig. 3 Microstructures of 8 h-solution treated specimens after aging at 190 °C for different durations: (a) 6 h, OM; (b) 8 h, OM; (c) 10 h, OM; (d) 10 h, SEM

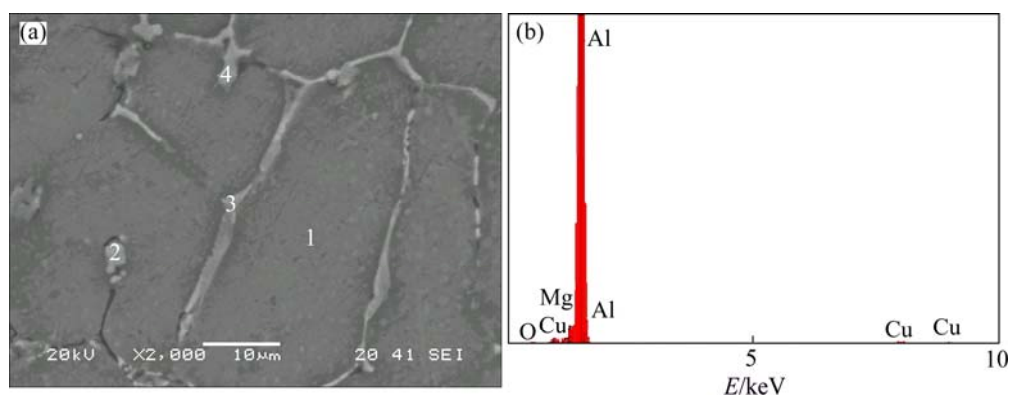


Fig. 4 SEM image (a) of specimen in as cast condition and EDX result (b) taken from matrix

Table 2 EDX analysis of phases in microstructure of as-cast specimen

Position in Fig. 4	Chemical composition in mass fraction/%					
	Cu	Mg	Ni	Fe	Si	Al
1	0.93	1.41	—	—	—	Bal.
2	32.00	—	11.72	—	—	Bal.
3	5.58	4.62	14.15	2.01	3.27	Bal.
4	6.52	0.80	0.95	0.23	1.59	Bal.

Figure 6 shows SEM image of the specimen aged for 6 h after solution treatment for 8 h. The EDX analysis results for this sample is given in Table 4. It can be seen

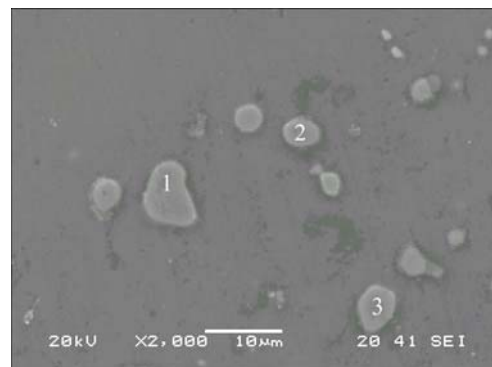
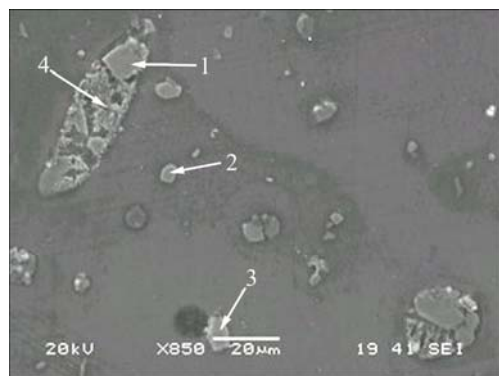


Fig. 5 SEM image of specimen after solution heat treatment for 8 h

Table 3 EDX analysis of phases after solution heat treatment for 8 h

Position in Fig. 5	Chemical composition in mass fraction/%			
	Cu	Ni	Si	Al
1	23.65	28.28	–	Bal.
2	25.05	27.71	–	Bal.
3	22.29	22.26	1.73	Bal.

**Fig. 6** SEM image of specimen aged for 6 h after solution treatment for 8 h**Table 4** EDX analysis of phases in microstructure of aged specimen for 6 h after solution treatment for 8 h

Position in Fig. 6	Chemical composition in mass fraction/%				
	Cu	Mg	Ni	Si	Al
1	25.10	–	28.83	–	Bal.
2	24.72	–	28.27	–	Bal.
3	22.60	–	28.43	–	Bal.
4	15.46	0.76	3.14	4.70	Bal.

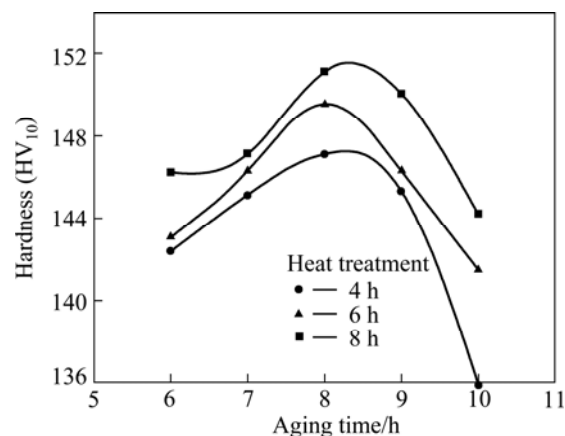
that aging treatment has no effect on Ni-containing phases. But it was reported that Cu-containing phases are located near the high nickel-containing intermetallics (Phases numbered 1 and 4 in Fig. 6). It may be concluded that Ni-containing phases act as nucleation sites for Al_2Cu phases but more research is needed to understand and prove this approach.

3.3 Hardness measurements

The hardness of the specimens was determined after different aging treatments. The hardness values of the specimens after different solution and aging treatments are given in Fig. 7. The hardness value of the as-cast sample was measured as HV_{10} 96.5.

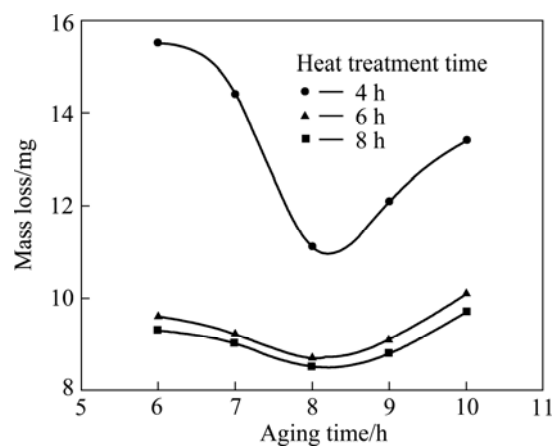
The reported hardness values are compatible with the microstructural findings. With increasing aging duration the hardness of the material increases to a maximum value and then starts to decrease because of over aging. When the specimens with the same aging duration were compared, the specimens with longer solution treatment durations have higher hardness values. This is a result of the increase in the number of dissolved

copper atoms with increasing solution treatment duration. Increasing the number of dissolved copper atoms causes the formation of more precipitates during aging treatment.

**Fig. 7** Hardness variations of specimens solution heat treated for 4, 6 and 8 h with aging time

3.4 Wear tests

Wear behavior of $Al4Cu2Ni2Mg$ alloy is critical because this material is used to produce engine pistons. Variation of the wear loss data with aging time is given in Fig. 8. As can be seen that the wear test results are in good harmony with the hardness test results. With increasing hardness the wear rate of the material decreases. When the specimen is over aged, the hardness starts to decrease and the wear loss of the alloy increases. Increasing solution heat treatment duration from 4 to 6 h has enormous effect on the wear rate of the specimens for all aging durations. Further increasing solution treatment duration from 6 to 8 h also has a positive effect on the wear rate but the decrease in the wear loss is slight. Although the difference in the mass loss of the alloys is slight in the experiments, these results will have great effect on the service life of the alloy.

**Fig. 8** Effect of aging time on mass loss of specimen heat treated for different time

During aging treatment, precipitates with high hardness form. These precipitates increase the hardness of the alloy. With increasing the hardness of the alloy the wear loss during wear test decreases.

4 Conclusions

1) The hardness of the alloy increases with increasing aging duration to a maximum value and then decreases because of over aging. The maximum hardness value after heat treatments was reported as HV₁₀ 151.1. The hardness of the as-cast sample was HV₁₀ 96.5. The increase in the hardness of the alloy is more than 50%.

2) The wear test results are compatible with the hardness test results. With increasing the aging duration the wear loss decreases to a minimum value and then starts to increase. The increase in the wear resistance of the alloy is caused by the precipitates that increase the hardness of the material.

3) It is reported that nickel-containing phases may act as nucleation sites for the copper-containing precipitates during aging treatments. More studies are needed to understand this mechanism.

4) With all the data obtained during the experiments, solution treatment for 8 h and aging for 8 h are chosen as optimal heat treatment conditions for Al₄Cu₂Ni₂Mg alloy.

Acknowledgement

The authors thank to the Scientific Research Projects Unit of Kocaeli University and Anel Doğa Entegre Geri Dönüşüm Endüstri A.Ş. for their supports.

References

- [1] TANG H, CHENG Z, LIU J, MA X. Preparation of a high strength Al–Cu–Mg alloy by mechanical alloying and press-forming [J]. *Materials Science and Engineering A*, 2006, 550: 51–54.
- [2] YAZDIAN N, KARIMZADEH F, TAVOOSI M. Microstructural evolution of nanostructure 7075 aluminum alloy during isothermal annealing [J]. *Journal of Alloys and Compounds*, 2010, 493: 137–141.
- [3] DINAHARAH I, MURUGAN N. Dry sliding wear behavior of AA6061/ZrB₂ in-situ composite [J]. *Transactions of Nonferrous Metals Society of China*, 2012, 22: 810–818.
- [4] PRABHU B, SURYANARAYANA C, AN L, VAIDYANATHAN R. Synthesis and characterization of high volume fraction Al–Al₂O₃ nanocomposite powders by high-energy milling [J]. *Materials Science and Engineering A*, 2006, 425: 192–200.
- [5] TORRALBA J M, VELASCO F, COSTA C E, VERGARA I, CACERES D. Mechanical behaviour of the interphase between matrix and reinforcement of Al 2014 matrix composites reinforced with (Ni₃Al)_p [J]. *Composites Part A: Applied Science and Manufacturing*, 2002, 33: 427–434.
- [6] MONDAL D P, JHA N, BADKUL A, DAS S. Effect of Al–TiB master alloy addition on microstructure, wear and compressive deformation behaviour of aluminum alloys [J]. *Transactions of Nonferrous Metals Society of China*, 2012, 22: 1001–1011.
- [7] GUPTA M, SRIVATSAN T S. Interrelationship between matrix microhardness and ultimate tensile strength of discontinuous particulate-reinforced aluminum alloy composites [J]. *Materials Letters*, 2001, 51: 255–261.
- [8] LOZANO D E, MERCADO-SOLIS R D, PEREZ A J, TALAMENTES J, MORALES F, HERNANDEZ-RODRIGUEZ M A L. Tribological behaviour of cast hypereutectic Al–Si–Cu alloy subjected to sliding wear [J]. *Wear*, 2009, 267: 545–549.
- [9] TAGHIABADI R, GHASEMI H M, SHABESTARI S G. Effect of iron-rich intermetallics on the sliding wear behavior of Al–Si alloys [J]. *Materials Science and Engineering A*, 2008, 490: 162–170.
- [10] ZEREN M. The effect of heat-treatment on aluminum-based piston alloys [J]. *Materials and Design*, 2007, 28: 2511–2517.
- [11] WIERZBINISKA M, SIENIAWSKI J. Microstructural changes to AlCu₆Ni₁ alloy after prolonged annealing at elevated temperature [J]. *Journal of Microscopy*, 2010, 237: 516–520.
- [12] KIM Y, BUCHHEIT R G. A characterization of the inhibiting effect of Cu on metastable pitting in dilute Al–Cu solid solution alloys [J]. *Electrochimica Acta*, 2007, 52: 2437–2446.
- [13] WIERZBINISKA M, SIENIAWSKI J. The influence of long-lasting annealing on microstructure of AlCu₄Ni₂Mg₂ alloy [J]. *Journal of Achievements in Materials and Manufacturing Engineering*, 2009, 34: 122–129.
- [14] WANG F, XIONG B Q, ZHANG Y A, LI Z H, LI P Y. Microstructural characterization of an Al–Cu–Mg alloy containing Fe and Ni [J]. *Journal of Alloys and Compounds*, 2009, 487: 445–449.
- [15] MROWKA-NOWOTNIK G. Examination of intermetallic phases in AlCu₄Ni₂Mg₂ aluminum alloy in T6 condition [J]. *Archives of Metallurgy and Materials*, 2010, 55: 489–497.
- [16] MROWKA-NOWOTNIK G. Intermetallic phase particles in cast AlSi₅Cu₁Mg and AlCu₄Ni₂Mg₂ aluminum alloys [J]. *Archives of Materials Science and Engineering*, 2009, 38: 69–77.
- [17] ZOLOTOREVSKY V S, BELOV N A, GLAZOFF M V. *Casting aluminum alloys* [M]. Oxford: Elsevier Publishers, 2007: 52.
- [18] BELOV N A, ESKIN D G, AVXENTIEVA N N. Constituent phase diagrams of the Al–Cu–Fe–Mg–Ni–Si system and their application to the analysis of aluminium piston alloys [J]. *Acta Materialia*, 2005, 53: 4709–4722.
- [19] CHEN C L, RICHTER A, THOMSON R C. Mechanical properties of intermetallic phases in multi-component Al–Si alloys using nanoindentation [J]. *Intermetallics*, 2009, 17: 634–641.
- [20] AHMAD Z. *Recent trends in processing and degradation of aluminium alloys* [M]. Rijeka: InTech Publisher, 2011: 19.
- [21] HUMPHREYS F J, CHAN H M. Discontinuous and continuous annealing phenomena in aluminium–nickel alloy [J]. *Materials Science and Technology*, 1996, 12: 143–148.

热处理条件对 $\text{Al}_4\text{Cu}_2\text{Ni}_2\text{Mg}$ 合金 组织和磨损性能的影响

Erdem KARAKULAK, Muzaffer ZEREN, Ridvan YAMANOĞLU

Metallurgical and Materials Engineering Department, Kocaeli University, 41380 Kocaeli, Turkey

摘 要: $\text{Al}_4\text{Cu}_2\text{Ni}_2\text{Mg}$ 合金是一种可时效硬化合金。研究不同固溶和时效热处理工艺参数对 $\text{Al}_4\text{Cu}_2\text{Ni}_2\text{Mg}$ 合金显微组织、硬度和耐磨性能的影响。采用光学显微镜和扫描电镜对样品的显微组织进行表征。通过硬度测试和磨损试验研究热处理工艺对材料性能的影响。结果表明,合金经过 8 h 时效后,硬度值从未经时效处理的 HV_{10} 96.5 增加到 HV_{10} 151.1,继续延长时效时间会导致合金的硬度值下降。随着合金硬度的提高,其耐磨性能得到提升。综合实验结果表明,在 540 °C 固溶处理 8 h,然后在 190 °C 时效处理 8 h,为该合金的最优热处理条件。

关键词: Al–Cu–Ni–Mg 合金; 活塞合金; 人工时效; 硬度; 磨损

(Edited by Sai-qian YUAN)