



A STUDY OF PHASE TRANSFORMATION IN SHAPE MEMORY ALLOY CuAl9Fe4

Nguyen Duong Nam¹, Vu Anh Tuan^{1,2}, Nguyen Hai Yen¹, Dao Van Lap¹, and Pham Mai Khanh², *

¹Viet Nam Maritime University, Hai Phong City, Viet Nam.

²School of Material Science and Engineering, Ha Noi University of Science and Technology, Ha Noi City, Viet Nam.

*Corresponding author email: khanh.phammai@hust.edu

This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

ARTICLE DETAILS

Article History:

Received 01 February 2019

Accepted 14 March 2019

Available online 28 March 2019

ABSTRACT

Microstructure and phase transformation in shape memory alloy Cu-Al (Fe addition: 9% Al, 4 % Fe) were investigated in this study. It is a material capable of restoring its original shape when impacted by a reasonable temperature or cyclic stress and production cost is not too expensive. In many years, shape memory alloys (SMA) based on Cu - Fe is considered to be very attractive to industrial and biomedical applications such as cardiovascular, orthopedic, surgery and production of spare parts and jigs. Original alloy exists β martensite and only exists $[\alpha^*]_1$ (3R) + $[\beta']_1$ (9R) + $[\gamma^*]_1$ (2H) which is finely dispersed homogenously in microstructure after finishing shape memory transformation. Shape memory rate obtained 6%-24% corresponding to each deformation angle before the experiment. Phase transformation between these two phases during deformation as well as the heating-cooling process which create the basis for the unique properties of the alloy.

KEYWORDS

shape memory alloys, transformation temperatures, shape memory effect.

1. INTRODUCTION

Alloy memory capacity is one of the important features for making machine parts as well as in other fields. In ship shaft systems made of copper alloys, this factor is very important [1,2]. The application of shape memory alloys here will contribute to reducing costs when manufacturing these parts [3,4]. In copper alloys, the phase transformation mechanism takes place including spinodal decomposition mechanism, transformation mechanism of martensite phase, durable chemical phase mechanism. In the above mechanisms, the martensite transformation mechanism will be applied to realize the memory formation for this alloy system [3-5].

There are many copper alloy systems that can be applied in the fabrication of shaft such as nickel copper-tin alloy, aluminum copper alloy alloyed with Fe and Ni ... Among them, aluminum-copper alloy has the transformation of martensite phase; This is an important change to the ship's main shaft [6]. Aluminum alloys are widely used in the automotive and aerospace industry because of their light weight and mechanical properties, which can reduce production costs and are able to work at high temperatures.

However, their thermostability is limited to temperatures not exceeding 200°C. One way to improve thermal stability is alloying with transition elements such as nickel, chromium, iron or manganese. These elements have low diffusion and dissolution in aluminum. Therefore, they stabilize the structure at high temperatures and mechanical properties are still positive. With some current SMAs alloys such as NiTiZr, NiTiHf, NiTiPd, NiAl and NiMnGa SMAs, but NiTiZr, NiAl alloy have high brittleness or NiTiPd alloy has a high working temperature but expensive [7-10].

Aluminum bronzes, ternary alloys includes Cu-Al which are two major elements and addition by the third element which is Fe. Cu-Al-Fe is similar to many other binary and ternary alloys that have martensite transformation from original β disordered, unstable at high temperature. After quenching, it occurs chain of reaction $\beta \rightarrow \beta_2 \rightarrow \beta$, finally, the formation of martensite ordered $\alpha^*1, \beta^*1, \gamma^*1$ according to Al, Fe amount in alloy [11,12].

Shape memory alloy-Symbol: SMA is a material that is capable of restoring its original shape when it is affected by an appropriate thermo-mechanical process. The reason SMA can remember their initial shape was firstly explained by Frederick E. Wang (an expert in crystal physics): changes in the structure of the atomic that form properties of SMAs, he found that

when changing to a certain temperature, there will be a phase transformation in the solid state for this alloy.) Shape memory alloy has two types: non-reversible and reversible type, in this paper, aims to study the reversible shape memory alloy [13-16].

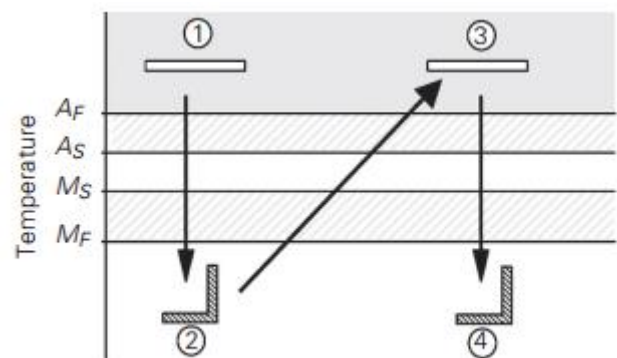


Figure 1: Reversible shape memory effect

The memory pattern is deformed above A_F Temperature (1), Cooling the sample to make the phase change Austenite \rightarrow Martensite, resulting in a change in shape (2), When the temperature rises to A_F other phase transformation (2) \rightarrow (3) here restores the original shape (3). A work other cooling causes the sample to return to a low-temperature shape (4), It must be shown that, in contrast to the one-way memory effect, there is no need to apply mechanical loads to change the shape of the sample at low temperatures.

Due to that special property, shape memory alloy has many applications in military and medical techniques (Manufacturing stents, manufacturing bone braces, spinal support, heart valves, orthodontic braces, ... Although there are many alloys known for shape memory effect, only a few alloys can recover the shape memory effect to be applicable in commerce. Up to now, only a few alloys have been applied such as Ni-Al alloy; alloys based on copper such as Cu-Zn-Al and Cu-Al-Ni [15,17,18].

Recent studies have shown that shape memory alloys can operate at high temperatures (above 200°C) while some shape memory alloys with high memory temperatures have been developed. Among these alloys, there are several shape memory alloys that are of interest: NiTiZr, NiTiHf, NiTiPd, NiAl, and NiMnGa. But among those memory alloys, NiTiZr and NiAl have high brittleness, NiTiPd is expensive. Due to these reasons, scientists have focused on studying new shape memory alloys with cheaper cost that have good properties and high shape memory temperatures [15,17,19,20].

The shape memory alloys based on copper are of particular interest because of the low production cost and good shape memory effect. Cu-ZN-Al, Cu-Al-Ni, and Cu-Al-Mn alloys are some of the common shape memory alloys based on copper that have wide applications due to the wide range of memory temperatures. Among these above alloys, only the Cu-Al-Ni shape memory alloy can be used at temperatures near 200°C [21,22].

This study needs to demonstrate the two-dimensional memory effect of Cu-Al-Fe alloy through phase transformation, how the crystal structure changes in process rate, and how much memory is formed [23-25]. In the article, it was shown that the phase trip, the microorganism organization, the crystal structure formed, the percentage of the image is remembered

2. EXPERIMENTAL PROCEDURE

2.1 Sample preparation

Cu-Al-Fe alloy was selected for investigating. The required amount of sample was 300g with the chemical composition containing 9% Al (27g), 4% Fe (12g), the remaining part is pure copper about 261g. The sample was melted in the induction furnace with protective Argon gas. Liquid metal was poured into a metal mold with a size of 150mm × 150mm × 3mm) The bars of size 100mm × 20mm × 3mm were cut from the original ingot [26]. They were heat treated at a temperature of 1123K focused on β phase area in 6 hours with argon protective gas and water quenched. Also, thin sheets of 1mm thickness were obtained by hot-rolling ingot after casting at temperature 1073K. These rolling samples were heat treated at 1123k in 15min then water quenched, then tempered at 623K, finally put in water to room temperature.)

2.2 Shape memory experiment

Hot-rolled samples of 1mm thickness were bent around the U-shaped mandrel with a 5% deformation degree to determine original shape recovery degree according to SME formula = recovery angle when unloading / (180 - recovery angle when heated)

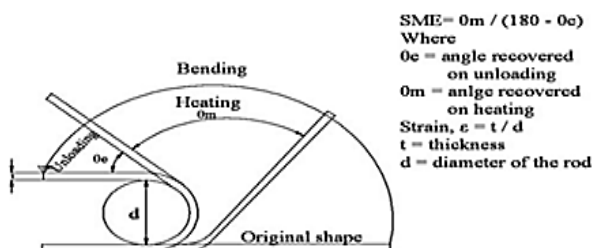


Figure 2: Schematic diagram of the bend test to determine strain recovery by SMA

2.3 Properties analysis

Using optical microscopy to observed microstructure and TEM method to determine the structure of the alloy [27]. Using scanning electron microscopy (SEM) and scanning EDX at multi-points mode to determine elemental composition by analytical region, distribution of alloy elements and impurities inside grain and at the grain boundary. Phase analysis by X-ray diffraction method by Cu $K\alpha$ electron radiation).

3. RESULT AND DISCUSSION

3.1 Microstructure

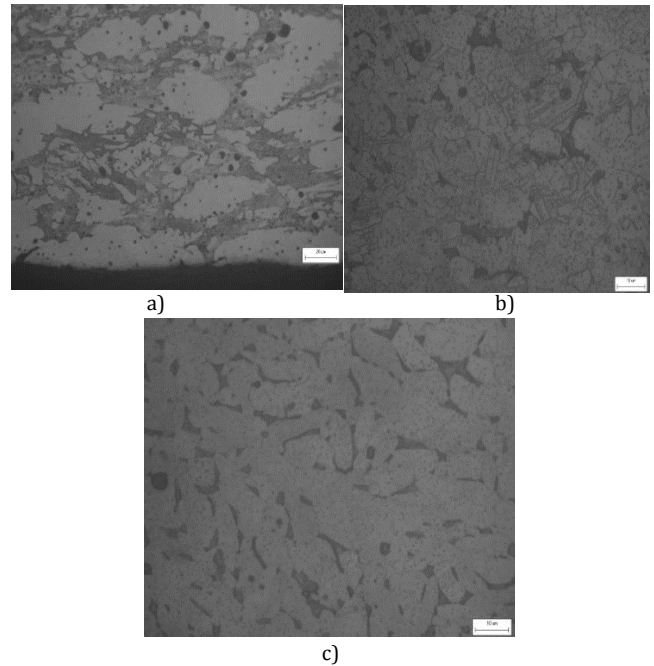


Figure 3: Microstructure of CuAl9Fe4 alloy: after rolling (a), before SMA (b) and after SMA (c)

After treating solid solution at 850°C in the $\alpha + \beta$ area that aimed to make solid solution homogenous, also reduced disaster area after casting. Next, water cooled to transform $\beta \rightarrow \beta'$ (solid solution based on Cu_3Al as β' but different about crystal lattice parameter), maybe existing α phase. On Figure 3a, microstructure after casting shows disappearance of disordered β' phase, can not see parallel interlace martensite plates in the microstructure, almost only exist dark β martensite (Cu_3Al) phase, light α phase and make the grain arrangement of the phase γ_2 so that the size is more suitable for the cast.

After rolling, the alloy was quenched at 850 ° C, then taken to bend test, the results in Figure 3b show a significant change in the dark β martensite phase (Cu_3Al), in detail, it occurred the formation $\beta \rightarrow \beta'$ again which appearing at light α phases boundary, created needle β' martensite plates along that boundary. The purpose of this period is to create stresses at the top of the martensite plate, to store energy to perform the heating again to check the amount of shape memory.

After finishing the shape memory test, the figure is shown in Figure 3c, which shows that the needle β martensite plates along the boundary between the light α phases disappeared due to the stress breakdown during heating. At this time, it occurred a transformation $\beta' \rightarrow (\alpha + \gamma_2)$, there was also an iron-rich $Fe(\delta)$ phase, similar to the original as-cast alloy, but γ_2 was precipitated which is finely dispersed homogenous in the microstructure.

3.2 XRD

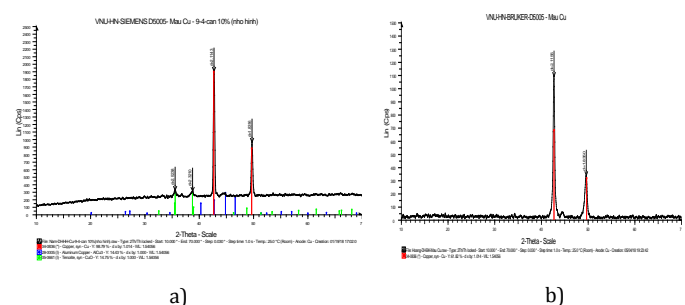


Figure 4: XRD of sample: before (a) and after (b) SMA

Compared to the above predictions, it proves that the above predictions are proper, as shown on the results of X-ray image 4a. After quenching at 850°C and then bending test after that, in the microstructure, only light phase α and disorder phase β' (orthorhombic crystal system). Finally, after the shape memory test, it seems that only β' martensite changed, in detail, only α and γ_2 (Cu₃Al) phases are present; in addition, also still have iron-rich Fe(δ) but the size is too small to observe on the result. Specifically, the standard peak tends to shift to the left because during the heat treatment process there are a change \rightarrow peaks changes, d changes \rightarrow the structure changes is as expected on the microstructure results.

3.3 TEM analysis result

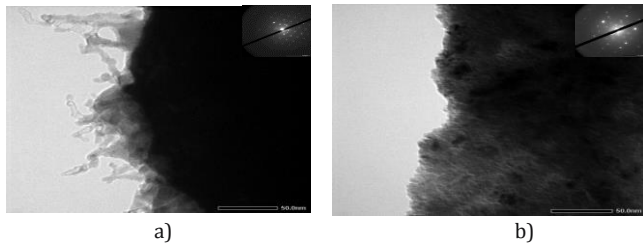


Figure 5: Result from about β martensite phase transformation of the sample before (a) and after (b) shape memory_ CuAl9Fe4

The result of TEM on Figure 5b of CuAl9Fe4 alloy shows more clearly the amount of γ_2 which is finally precipitated from martensite transformation. In addition, there may be α , similar to the origin after casting, but there is a change in the morphology, size and amount of precipitation of phase γ_2 (a solid solution based on Cu₃₃Al₁₉). While in Figure 5a, it seems that only β' martensite plate is observed as what is shown at before shape memory period. But specifically, it is the change of crystal lattice structure, size and precipitation amount of phase γ_2 (is a solid solution based on Cu₃₃Al₁₉) that tend to be better for shape memory effect.

3.4 Shape memory result

Table 1: SMA of sample

Before heat-treatment (°)	After heat-treatment (°)	Deformation (%)	SME (%)
90	96	5	7
150	164	5	47
150	174	10	80

Based on Table 1, it is shown that the obtained shape memory rate is relatively large (from 7 to 80%) expressed through the angle change before deformation and after the deformation is calculated according to SME formula = recovery angle/angle deformation. Through analysis with phase diagram, it shows that substance of the memory effect is the formation and transformation of martensite phase, from the period under the effect of phase β' until the effect of heat treatment to recover initial state ($\alpha + \gamma_2$), but this state has the properties, structure and grain size that change in order to be better for the shape memory effect.

4. CONCLUSIONS

This study analyzed and explained the mechanism of SME of the alloy system based on the transformation of the martensite phase of this alloy system. When heated and quenching, this alloy was created a martensite microstructure, but when it was heated again, the martensite structure decomposed into the forms: β' ; vitmantet and finally return to the initial state with α and γ_2 phases. The initial restoration of the structure will show the ability to remember the shape of this alloy system.

REFERENCES

[1] Hoang, A.T., Pham, X.D. 2018. An Investigation of Remediation and Recovery of Oil Spill and Toxic Heavy Metal from Maritime Pollution by A New Absorbent Material. *Journal of Marine Engineering Technology*, Pp. 1-11.

[2] Hoang, A.T. 2018. Waste Heat Recovery from Diesel Engines Based on Organic Rankine Cycle. *Applied Energy*, 231, Pp. 138-166.

[3] Thang, S.M., Nguyen, D.N. 2019. Study on Microstructure and Properties of Cu-9ni-6sn Alloy Applied for Electric Measurement. In *Applied Mechanics and Materials*. Trans Tech Publ.

[4] Pham, X.D., Hoag, A.T., Nguyen, D.N. 2018. A Study on The Effect of The Change of Tempering Temperature on The Microstructure Transformation Of Cu-Ni-Sn Alloy. *International Journal of Mechanical and Mechatronics Engineering*, 18 (4), Pp. 27-34.

[5] Nguyen, D.N., Hoang, A.T., Pham, X.D., Sai, M.T., Chau, M.Q., Pham, V.V. 2018. Effect of Sn Component on Properties and Microstructure Cu-Ni-Sn Alloys. *Jurnal Teknologi*, 80(6).

[6] Chieu, L.T., Thang, S.M., Nam, N.D., Khanh, P.M. 2016. The Effect of Deformation on Microstructure of Cu-Ni-Sn Aging Alloys. *Key Engineering Materials*, 682, pp. 113-118.

[7] Akselsen, O. 2010. *Joining of Shape Memory Alloys*, Intech.

[8] Borges, F.C.N. 2013. Iron Based Shape Memory Alloys: Mechanical and Structural Properties, In *Shape Memory Alloys-Processing, Characterization and Applications*. Intech.

[9] Sathishkumar, S., Kannan, M. 2018. Design and Fatigue Analysis of Multi Cylinder Engine and Its Structural Components. *Acta Mechanica Malaysia*, 2 (2), 10-14.

[10] Kihara, T., Xu, X., Ito, W., Kainuma, R., Adachi, Y., Kanomata, T., Tokunaga, M. 2017. Magnetocaloric Effects in Metamagnetic Shape Memory Alloys. In *Shape Memory Alloys-Fundamentals and Applications*, Intech.

[11] Nizamani, A.M., Daudpoto, J., Nizamani, M.A. 2017. Development of Faster Sma Actuators, In *Shape Memory Alloys-Fundamentals and Applications*, Intech.

[12] Stanciu, S., Bujoreanu, L. 2008. Formation Of B' 1 Stress-Induced Martensite in The Presence Of Γ -Phase, In *A Cu-Al-Ni-Mn-Fe Shape Memory Alloy*. *Materials Science and Engineering: A*, 481, Pp. 494-499.

[13] Stipcich, M., Romero, R. 1999. The Effect of Post-Quench Aging on Stabilization of Martensite in Cu-Zn-Al And Cu-Zn-Al-Ti-B Shape Memory Alloys. *Materials Science and Engineering: A*, 273, Pp. 581-585.

[14] Zhang, Z., Li, W., Ashraf, M.A. 2018. Allelopathic Effects of Various Aquatic Plants in Eutrophic Water Areas. *Journal of Coastal Research: Special Issue 82. Coastal Ecosystem Responses to Human and Climatic Changes throughout Asia*: pp. 137 - 142.

[15] Okabe, S., Suzuki, T., Yoshikawa, S. 2017. Shape Memory Wires in R3, In *Shape Memory Alloys-Fundamentals and Applications*, Intech.

[16] Martínez-Fuentes, R.J., Sánchez-Arévalo, F.M., García-Castillo, F.N., Lara-Rodríguez, J.A., Cortés-Pérez, J., Reyes-Solis, A. 2013. Micromechanical Behavior of Cualbe Shape Memory Alloy Undergoing 3-Point Bending Analyzed by Digital Image Correlation, In *Shape Memory Alloys-Processing, Characterization and Applications*, Intech.

[17] Lojen, G., Gojić, M., Anžel, I. 2013. Continuously Cast Cu-Al-Ni Shape Memory Alloy-Properties in As-Cast Condition. *Journal of Alloys and Compounds*, 580, Pp. 497-505.

[18] Malard, B., Sittner, P., Berveiller, S., Patour, E. 2012. Advances in Martensitic Transformations in Cu-Based Shape Memory Alloys Achieved by In Situ Neutron and Synchrotron X-Ray Diffraction Methods. *Comptes Rendus Physique*, 13 (3), Pp. 280-292.

[19] Jain, N., Rastogi, S. 2019. Speech Recognition Systems - A Comprehensive Study of Concepts and Mechanism. *Acta Informatica Malaysia*, 3 (1), 01-03.

[20] Pisarek, B. 2013. Model of Cu-Al-Fe-Ni Bronze Crystallization. *Archives of Foundry Engineering*, 13 (3), Pp. 72-79.

[21] Sutou, Y., Koeda, N., Omori, T., Kainuma, R., Ishida, K. 2009. Effects of Ageing on Bainitic And Thermally Induced Martensitic Transformations in Ductile Cu-Al-Mn-Based Shape Memory Alloys. *Acta Materialia*, 57 (19), Pp. 5748-5758.

[22] Abugalia, A. 2019. Effect of Corona on The Wave Propagation Along Overhead Transmission Lines. *Acta Electronica Malaysia*, 3 (1), 06-09.

[23] Hasan, F., Lorimer, G., Ridley, N. 1982. Crystallography of Martensite in A Cu-10al-5ni-5fe Alloy. *Le Journal De Physique Colloques*, 43 (C4), Pp. C4-653-C4-658.

[24] Gupta, R.D., Chakravarty, D.P., Chakravarty, R., Gupte, P.K. 1969. Study of Microstructure of Chill Cast Aluminium Bronzes (Cu-Ai-Fe-Mn). Pp. 123-132.

[25] Sutou, Y., Omori, T., Kainuma, R., Ishida, K. 2004. Characteristics of Cu-Al-Mn-Based Shape Memory Alloys and Their Applications. *Materials Science and Engineering: A*, 378 (1-2), Pp. 278-282.

[26] Van Humbeeck, J. 2012. Shape Memory Alloys with High Transformation Temperatures. *Materials Research Bulletin*, 47 (10), Pp. 2966-2968.

[27] Tian, X., Li, Q., He, C., Cai, Y., Zhang, Y., Yang, Z. 2018. Design and experiment of reciprocating double Track Straight Line Conveyor. *Acta Mechanica Malaysia*, 2 (2), 01-04.

