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A REVIEW OF MICROSTRUCTURE AND CORROSION BEHAVIOR OF CU AL NI SHAPE MEMORY ALLOY

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ABSTRACT

The shape memory alloys (SMAs), that consist of Cu-Al-Ni, have been established and developed for the applications used at higher temperature values due to their capability to return to its original shape when heating close to its transformation temperature. Cu-Al-Ni alloy has high value of transformation temperature and show small hysteresis as compared to the other types of SMAs. This work present a general review about how SMAs have developed by adding several metals such as, Ti, Be, B, Mn, and Zr or by changing its content (Ni or Al) by either decreasing or increasing. That might show a significant impact on the phase transition and enhancing the corrosion behavior and mechanical properties of the presented alloys. However, the transformation of the martensite phase is the critical factor that might change the properties of Cu-Al-Ni SMAs. This phase is largely affected by addition of the alloying element mentioned above. A review about effect of addition some alloying elements for enhancing the corrosion characteristics of the alloy and phase transition is presented.

Keywords: Corrosion, Cu-Al-Ni, Shape Memory Alloys, Phase Transformation, SEM

البنية المجهرية و سلوك التاكل لسبيكة Cu Al Ni ذاكرة الشكل
(مراجعة)

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الخلاصة

تم إنشاء وتطوير سبائك ذاكرة الشكل (SMAs) ، التي تتكون من Cu-Al-Ni ، للتطبيقات المستخدمة في درجات الحرارة العالية نظرًا لقدرتها على العودة إلى شكلها الأصلي عند التسخين بالقرب من درجة حرارة التحول. سبيكة Cu-Al-Ni لها قيمة عالية لدرجة حرارة التحول وتظهر تباطؤًا صغيرًا مقارنة بأنواع SMAs الأخرى. يقدم هذا العمل مراجعة عامة حول كيفية تطور SMAs عن طريق إضافة العديد من المعادن مثل Ti و Be و B و Mn و Zr أو عن طريق تغيير محتواها (Ni أو Al) إما بالتناقص أو الزيادة. قد يظهر ذلك تأثيرًا كبيرًا على التحولات الطورية وتحسين السلوك التاكلي والخصائص الميكانيكية للسبائك المقدمة. ومع ذلك ، فإن تحول مرحلة المارتينسايت هو العامل الحاسم الذي قد يغير خصائص Cu-Al-Ni. تتأثر هذه المرحلة إلى حد كبير بإضافة عنصر السبيكة المذكور أعلاه. يتم تقديم مراجعة حول تأثير إضافة بعض عناصر السبائك لتحسين مقاومة التآكل للسبيكة والتحولات الطورية

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INTRODUCTION

Since the commencement of the current century ,Shape Memory Alloys (SMAs) were considered from the smart materials, their application vary from the medical uses to the military ones, and the Cu–based SMAs are extensively on request for rate effective with good mechanical and shape memory properties. Cu- based alloys are a proper substitute for the (NiTi) alloys in the industrial applications, like valves, actuators, MEMS, and damping uses, owing to their less cost, easy production as well as a high temperature range Lexcellent (2013). They show a good strain recovery (2- 4%) and a widespread range of the temperature required for the transformation (100- 170) °C Lagudas (2006) making them a good alternative for NiTi alloys, but big elastic anisotropy, big size of grain, their brittleness, and less strength impede their practical uses Kim et. al.(1990). Also, their brittleness makes a difficulty to machine them but this problem can be overcome by using the electric discharge machining (EDM) process or wire (WEDM)cutting process. Since , the quadric and the fifth element to any alloy can make changes in this alloy and properties Mhod et. al.(2015). However, alloying element addition to the Cu-Al-Ni alloys increases the resistance of corrosion and increases the mechanical properties. While the existence of the Al/Ni-X (where X is the alloying element) is crucial for Cu-Al-Ni alloys passivation

Phase diagram of Cu-Al-Ni SMAs

The cross section of the ternary alloys of Cu-Al-at 3 wt.% of Ni is presented in Figure(1). This alloy can demonstrate the properties of the SMA in the presence of materialized martensitic transformation. With the purpose of establish under-cooling, in which it vigorous to impose the martensitic transformation, with a long of completely reflection that the heat treatment can never be prohibited. That is actually come with temperature variety annealing of the stable phase (β phase) in order to ensure the water quenching and that leads to formation of this phase. The characteristic of the SMA of represented by Cu-Al-Ni are totally depends on the body center cubic β phase properties for the Cu-Al, that considered as binary alloys Chen (2009). The β phase undergoes to the eutectoid decomposition of $\beta \rightarrow \alpha + \gamma 2$ when this phase cooling down from 565°C. However, the higher rate of the cooling can prevent eutectic decomposition of this phase and allow the martensitic transformation. The body center cubic structure transforms to DO3-type super lattice as the Cu-Al-Ni SMA hold more than 11% of the Al content. This transformation accomplished by transferring the phase β to order β1 phase before the martensitic transformation. At this point, one can maintain that the martensite "inherits" the organized structure. Al content changes the formation of the phases. When the content of Al placed between 11 and 13 wt %, the martensite β′1, showed a monoclinic structure of 18R1 as the most component appeared in the structure. As the Al content increase above the 13 wt %, other phase just appeared and become the prevailed one, namely 2H-type $\gamma'1$ martensite. The presence of these formations of the phases is largely depends on the stress and temperature conditions. Furthermore, beside these phases, there are other types of the martensite phased that can be formed simultaneously and parallel to the first two phases, as shown in Figure(2). The temperature of characteristic of the Cu-Al-Ni alloys lie within the range of −200 and 200°C. Determination of this range depends on the Al and Cu contents. It is worth to mention that Al content has a critical impact by providing the required permittivity that makes them used for the applications requires high temperature resistance. The temperature of the transformation of the Ni–Ti alloys can be customized to be lie within the range of −200 and 120°C Mavroidis (2000). The A_f temperature specified of Fe-based SMAs can be controlled and increased even to 300°C. However, M_s stay at the level of the room temperature or may be lower than that. The last temperature can be calculated by using a specific empirical equation prepared for that, so that Scherngell (2000).

$$M_s(°C) = 2020 - 134 \times (wt.\ \%Al) - 45 \times (wt.\%Ni) \quad (1)$$

In order to decrease the temperature of the transformation, Al wt% is added to the Cu-based SMAs. For example, the transformation of the martensite is going to start at the room temperature at addition of Al at 14 wt%. However, other new phases may be presented with addition of Al, such as phase γ_2 (i.e., it denotes to the cubic intermetallic compound of Cu_9Al_4). The presence of this new phase is going to increase the alloy brittleness. In the other side, addition of Ni plays a crucial role for controlling the rate of diffusion of the Cu onto Al. This in turns, lead to hold single phase; either β or β1 until the transformation of the martensite phase has been achieved throughout the process of cooling.

On the other hand, the increasing in the Ni percentage in the Cu-Al-Ni SMAs ternary alloy will lead to increase the brittleness that accompanied with the eutectoid point shifting to higher values. Therefore, the chemical composition of both Ni and Al can be optimized within the range of 3.5-4 and 14wt%, respectively Otsuka, and Wayman (1999). On the other hand, several drawbacks have existed in these alloys, such as the low reversible transformation. This transformation involves 1.5% and 4% of the two-way and one-way SAMs effects, respectively. The disadvantages of these alloys were mainly represented by the presence of the cracks (intergranular) even at the low levels of the induced stresses. The main reason beyond crack failure at low stress level is the large size of the grains, the intense reliance if the strain transformation on the orientations of the crystal, high level of material anisotropy, and grain boundary segregation. The first three of the reasons are valid when the shear stress concentration is high at the boundaries of the grains. While the fourth is principally due to weakening in the boundaries of the grain Miyazaki (1996).

Phase transformation morphology

In general, the martensitic transformation can be developed by applying external stress or thermal stress. In different words, reducing the temperature in the presence of applied stress leads to drive the transformation of the austenite to the martensite. In fact, the relationship between the two forces is linear. This relation is basically derived from the conventional relationship of the phase transformation thermodynamics relationships, that called Clausius-Clapeyron relationship. In addition, the martensitic transformation characteristics are largely affected by the heat treatment Zhang and Shouwen (2000). The hysteresis and the temperature of transformation of the martensite are very sensitive to the precipitation process and β phase order degree Nó ML et. al.(2009) and Qiao (2011). Cu-based SMAs show the transformation of the martensitic from the β-phase to the packed structure when cooling down. In addition, the bcc structure of the Cu-Al-Ni and Cu-Zn-Al SMAs disordered because of the high value of the temperature of the β-phase Xu JW.(2008). There are two types of the martensite forms, β′1 and γ′1, that exist in the Cu-Al-Ni alloys. The presence of these forms depends on the heat treatment and composition of the alloys itself MLaRR(1999) , Agrawal and Dube (2018), Saud (2015), Gustmann (2017). β-phase stability decreases with the temperature. For example, this phase can stay at the metastable phase at a lower temperature and proper air cooling Gera (2018) Velmurugan and Senthilkumar (2018), Li D.Y (2016). The limit of the stability of the β-phase must be determined in order to prevent its ordination state expansion and/or the stable phases precipitation. However, the mechanical properties of Cu-Al-Ni SMA can be enhanced and controlled by controlling the grain size to produce a very fine grains Otsuka(2011). In addition, the alloys structure varies with the heating and cooling processes within the martensitic region. Furthermore, during the transformation processes from martensite into austenite forms, a usable force just induces due to the thermal cyclic load. This force might help for using these alloys for several important applications Bayram and Maraşlı(2018) , and Braga(2017). It is noted that the martensite transformation needs higher transformation energy than that corresponding in the reverse direction (reverse transformation). Wei ZG et. al. (1996).

The Cu-Al-Ni SMA's microstructure

The Cu-Al-Ni SMA's microstructure could be established in a needle and/or a plate-like martensites with morphology of self-accommodation Sampath(2005). During the addition of 4.3% Ni and 13.3% Al to Cu-Al-Ni SMAs, two various phases could be excited: (i) morphology of self-accommodating $\gamma'1$ and (ii) morphology of acicular $\beta'1$. Increasing the content of Al within the Cu-Al-Ni SMA would transform the martensite $\beta'1$ to $\gamma'1$ by means of a $\beta'1 + \gamma'1$ composition Recarte(1999) and Bhattacharya et. al.(2003). The β martensite would transform to $\beta'1$ martensite at high rate of cooling with very small quantities of $\gamma'1$ phase, but at low rate of cooling the $\beta'1$ would transform to $\gamma'1$ martensite.

Forming of $\gamma'1$ martensite was could not be avoided regardless the processing conditions in case of Al content was more than 14.2 wt.%. Small additions of Al to the Cu-Al-Ni SMAs could yield intermetallic compounds with Al. Nevertheless, if the Al matrix decreased the $\beta'1$ martensite would form. If the content of Al is approximately 11.9 wt.%, $\beta'1$ martensite formed as fine plates. $\beta'1 + \gamma'1$ mixtures were noticed in Cu-13.03 wt.% Al-4.09 wt.% Ni Chen and Liu)2003 and martensite would form primarily the M18R type having an orthorhombic structure Duerig (1990). On the other hand, Chentouf (2009) analyzed both of the microstructural and thermodynamic behaviour of hypoeutectoidal Cu-Al-Ni SMAs and found that Ni and Al contents have the dominant influence on the precipitated phase morphology as depicted in Figure (3). The formation of large precipitated (XL) particles in the SMAs of Cu-Al-Ni leading to transform 18R basal plane order at the precipitate-free and precipitate-matrix interfaces into 2H martensite. Ratchev et. al. (1993) referred to incidence of 18R changing in the sequence owing to the stress improvement around the precipitates. Karagoz and Canbay (2013) noticed that increasing the amount of Al made the β phase to dominant the $\gamma'1$ and $\beta'1$ phases total martensitic transformation throughout the process of homogenization and the grains were formed in various orientations with shape of V-type. Chang (2011) found that adding 13 wt.% of Al led to form zig zag groups of self-accommodating martensite at room temperature. Figure(4a) shows the typically formed $\beta'1$ martensite with 18R structure. On the other hand, figure(4b) showed that adding Al equals to 13.5 wt.% resulted in forming a few coarse variants of $\gamma'1$ (2H) structure that located at β'_1 (18R) matrix. Additional increasing of Al content to 13.7 and 14 wt.% made the microstructure to be more distinctive and showing a γ'_1 (2H) martensite or β'_1 (18R) in addition to the abundant of γ_2 phase precipitate as shown in Figure(4c) and d. Others Recarte (1999) , Recarte(2004), and Recarte(1996) reported the relationship between the composition of Cu-*x*Al-4Ni shape memory alloys and a set of transformed martensite. This relationship exhibited that γ'_1 (2H) martensite existed in Cu-14.0Al-4Ni SMAs, while both γ'_1 (2H) martensite and β'_1 (18R) should coexisted in Cu-13.7Al-4Ni SMA. On the other hand, adding Ti to Cu-Al-Ni SMA led to form a new phase called X-phase rich by Ti as found by Sugimoto et al.(1982). Additionally, the X-phase volume fraction showed linear increasing with the increase in the content of Ti. Another study achieved by Dutkiewicz et. al.(1996) showed that adding Ti to Cu-Al-Ni resulted in forming elongated and smaller grain size as it limited the growth of the grain as depicted in Figure 5(a–d).

Alloying elements effects on the corrosion behavior

Copper and copper alloys are widely used in the applications that require the mechanical strength associated with high thermal and electrical conductivity and good corrosion resistance Liberto et. al. (2007). Adding Al to the Cu-based alloys increased its resistance against corrosion due to the formation of the protective layer of alumina, which builds up quickly on the surface after the exposure to the corrosive environment. The existence of Ni was essential in Cu–Ni alloys passivation due to its inclusion in the Cu(I) oxide that was formed on the alloy's corroded surface that could reduce the cation vacancies number that existed in the Cu(I) oxide Schussler and Exner(1993), Schussler and Exner(1993), Badawy et. al. (2009), Nady et.al. (2012). Copper based shape memory alloys have been intensively studied from the viewpoints of metallography, fracture behaviour, mechanical properties and activation energy of the precipitate grow process, but relatively little attention has been devoted to their electrochemical behaviour Gojić et. al.(2011), Husain and Clapp (2011), Sathish et.al. (2014), Raju and Sampath (2011), Kneissl et. al. (2008), Čolić et. al.(2010), Živković et. al. (2014), Kumar et. al. (2016), Ivanić et.al. (2017). As the practical application of Cu-SMA is constantly increasing, these alloys maybe exposed to diverse corrosion media during long time and that could lead to the appearance of corrosion damage on their surface, jeopardize their structural integrity and cause immense consequences if the corrosion processes are not observed in time. Therefore, it is crucial to know the corrosion behaviour of Cu-based SMAs in different environments, and in turn improving their resistance against corrosion before their industrial applications. Adding alloying elements to Cu−Al−Ni alloys is important to increase their corrosion resistance and mechanical properties as well; however, the existence of Al/Ni−X precipitates (where X denotes to the alloying element) is necessary to passivate the Cu−Al−Ni alloys owing to its inclusion in the Cu(I) oxide that can be formed on the alloy's corroded surface. Kuo et. al. (2006) investigated the Cu−Al corrosion behaviour in H_2SO_4 media where adding Al could slightly decrease the density of pseudo-passive current at the potential of anode. The main reason of improving the Cu−Al alloy resistance against corrosion was the quick building of an obstacle film of Al_2O_3 Badawy et. al. (2014). Other researchers ascribed this improvement to the formation of a double layer of $Cu_2O•Al_2O_3•XH_2O$. Badawy et. al. (2014). The presence of nickel in the binary Cu−Ni alloy led to enhance the alloy's resistance against corrosion owing to the nickel inclusion into the shielding film of Cu_2O during the solid-state reaction Badawy et. al. (2010). On the other hand, the effect of using different concentrations of nickel on the Cu−Al−Ni alloys corrosion behaviour in aqueous solutions was studied by Badawy et. al. (2010). The study showed that higher concentration of nickel made the alloy surface enriched with nickel and, therefore, it decreased the alloy's corrosion resistance. Many studies have focused on studying the mechanical properties of Cu-based SMAs, whereas the electrochemical properties of Cu−Al−Ni−Mn and Cu−Al−Ni−Ti SMAs were not mentioned somewhere else. Saud et. al.(2015) studied the corrosion behaviour of Cu−Al−Ni−Mn and Cu−Al−Ni−Ti SMAs using different concentrations of Mn or Ti (0.4% to 1.0%) in a 3.5% sodium chloride solution. Moreover, the influence of adding different concentrations of Mn or Ti on the corrosion characteristics and microstructure of Cu−Al−Ni alloys was studied as well. A new Cu-based SMAs was produced by Saud et al. (2015) which used silver nanoparticles (0.25 wt%) with Cu–Al–Ni SMAs to enhance the corrosion resistance and structural properties of the alloy. The corrosion potential was shifted from -277.1 to -307.4 mV. by adding silver nanoparticles to Cu–Al–Ni SMAs and the current density was reduced to 5.61 $\mu A/cm^2$ that means the corrosion protection has been considerably improved. SEM images and EDX analysis before the polarization test indicated the presence of corrosion products formation such as AgCl, Cu_2O and CuCl in addition to Ag_2O and $Al_2O_3/Al(OH)_3$. In the same trend, adding Al or Al–Ag into Cu led to enhance the corrosion behaviour of Cu–Al–Ag, Cu–Al, and pure Cu. Other studies investigated the influence of adding Zn, Fe, Zr, B, Mn, Ni, Ti and CNTs as micro-alloying on the mechanical and structural properties of Cu–Al. The influence of adding chromium on structure of the martensite, temperature of transformation, corrosion behavior and mechanical properties of Cu–Al–Ni SMAs was firstly considered by Zare and Ketabchi(2016). They used thermo–mechanical treatment that included consecutive hot rolling, annealing, and passes of hydraulic pressing. Samples that were treated under thermo–mechanical conditions displayed perfect reversibility and shape-memory behaviour. Further it was observed by Al-Hassani et. al. (2017) that the austenite rate of corrosion was greater than martensite in CuAlNi SMAs. Moreover, the specimen in the austenitic structure have current density higher than for martensite structure which proved the higher corrosion resistance of SMAs than other traditional alloys because the hyperactive elastic behavior of polycrystalline structure. KUO et. al. (2006) studied the corrosion behavior of Cu–Al-Ni shape-memory alloys in 0.5 ml H2SO4 solution at 25°C which studied by means of anodic polarization and cyclic voltammeter and the results showed that anodic dissolution rates of alloys decreased slightly with increasing the concentrations of aluminum, however the formation of barrier film of Al2O3 on the alloy surface was the main cause behind the improvement of Cu−Al-Ni alloy's corrosion resistance Badawy et.al. (2014). Abid Ali and Al-Tai (2010) studied the effect of iron addition on the corrosion behavior of Cu-Al-Ni SMA in 5%NaOH and different amount of Fe (0.4,0.8,1.2weight%). The results showed that the Cu-Al-Ni shape memory alloy in austenitic structure has better corrosion resistance than in the martensitic structure and the higher corrosion resistance achieved when the Fe addition was 0.4%. Gojic et al. (2011) studied the electrochemical and microstructural of Cu–Al–Ni SMA the corrosion behavior of Cu–Al–Ni SMA in 0.5ml NaCl solution by linear and open circuit potential measurements The results showed that the chloride ions adsorption on the surface of electrode led to reduce the EOCP values and the collapse of the oxide protective film. Saud et. al. (2017) studied the effect of adding Ta on the microstructure, damping, corrosion resistance and shape memory behavior of pre-alloyed Cu-Al-Ni SMAs. Polarization tests in 3.5%NaCl solution showed that the corrosion resistance of Cu-Al-Ni-Ta SMA improved with escalating Ta concentration, higher corrosion potential, and formation of stable passive film. Hallem and. Hashim (2018) investigated the corrosion resistance and microstructure of Cu−Al−Ni SMA with different contents of Ge additions (1,2 and 3wt %) in a 3.5% NaCl solution. They Studied the corrosion resistance in 3.5%NaCl for Cu-Al-Ni SMA prepared by powder metallurgy with addition of (1 ,2 and 3wt%) of germanium. The electrochemical test showed that the corrosion rate decreased from 34.99 (mpy) to 4.72 (mpy) when the addition of germanium was 3%. They concluded that increasing the Germanium addition led to enhance the Cu-Al-Ni SMA corrosion resistance.

Summary

This paper reviewed the previous researches concerning Cu-Al-Ni SMAs characteristics in terms of microstructure, phase transformation morphology, and corrosion. The highest benefits of Cu-Al-Ni alloys could be obtained after the modification processes. The SMAs selection requires special consideration regarding with the cost of manufacturing and performance. Therefore, the Cu-Al-Ni SMAs exhibited considerable attentions owing to their lower cost compared with Ti-based SMAs. It is well known that Cu-based SMAs have some drawbacks especially the resistance against corrosion and they need special improvements to overcome this weakness. Accordingly, the phase characteristics and the microstructure have been modified by adding the alloying elements as a more considerable method. Adding fourth element to the ternary Cu-Al-Ni SMA could modify the martensitic phase structure and/or morphology and, therefore, improving the mechanical properties.

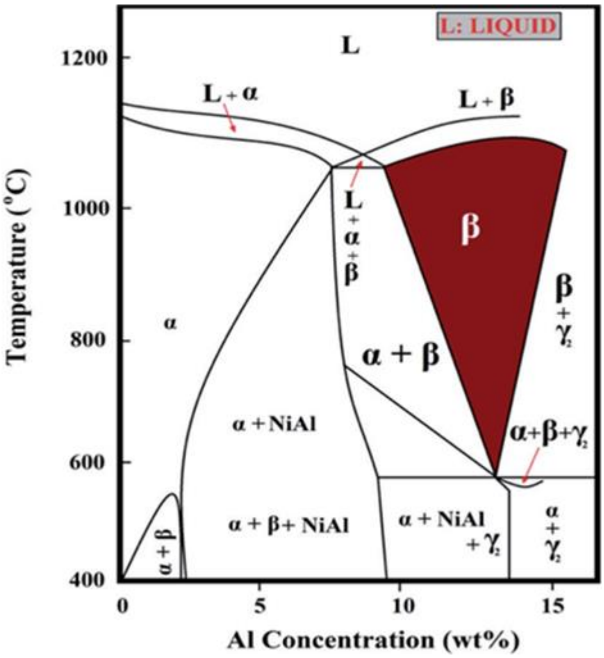

Fig.(1). Diagram of the cross-section of the Cu-Al-3 wt.% Ni ternary alloy(Dunne (1982))

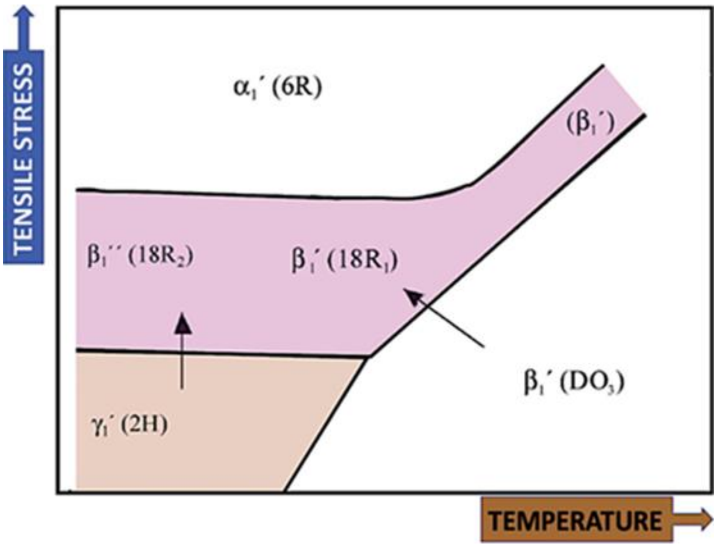

Fig . (2**):** Schematic phase diagram of Cu-Al-Ni alloy in temperature-stress coordinates (Otsuka and Wayman (1999)) (Delaey(1990))

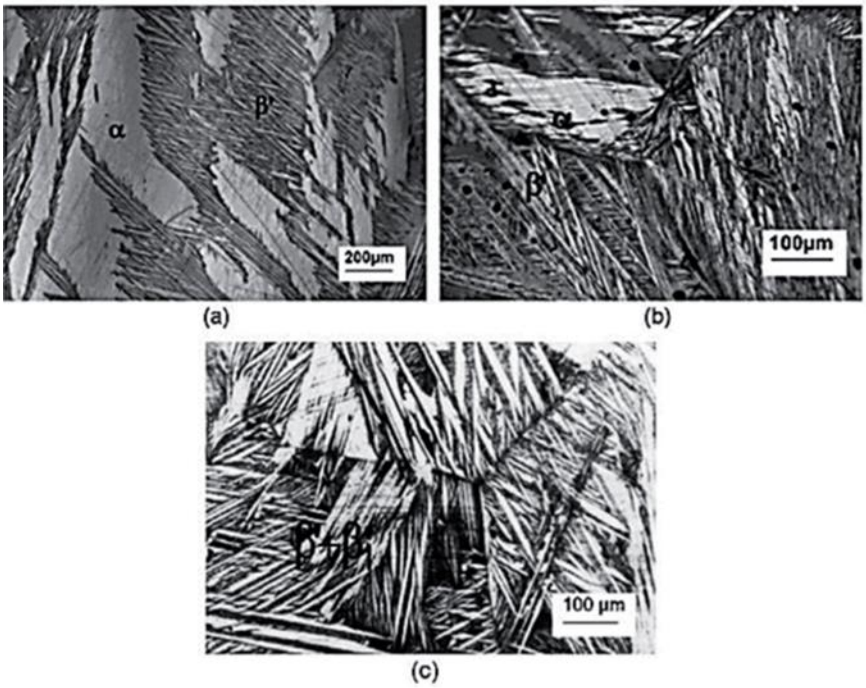

Fig. (3). Microscopic images for alloys: (a) Cu-9.9 wt.% Al-4.43 wt.% Ni, (b) Cu-11.25 wt.% Al-4.07 wt.% Ni and (c) Cu-11.79 Al - 4.37 wt.% Ni (Chentouf (2009**)).**

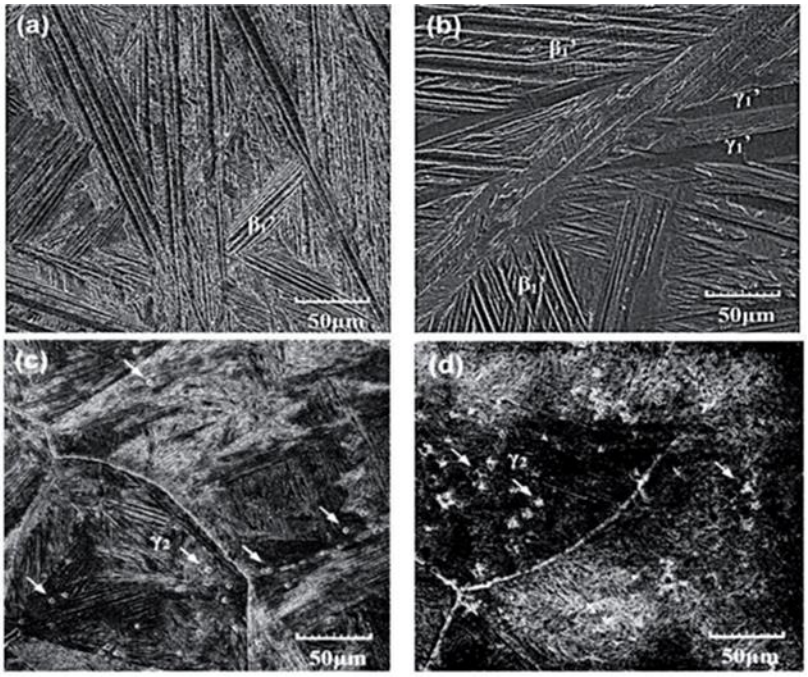

Fig.(4) SEM images of (a) Cu-13 wt.% Al- 4 wt.% Ni, (b) Cu-13.5 wt.% Al - 4 wt.%Ni, (c) Cu-13.7 wt.% Al - 4 wt.%Ni, and (d) Cu-14 wt.% Al - 4 wt.% Ni SMAs (Chang (2011)).

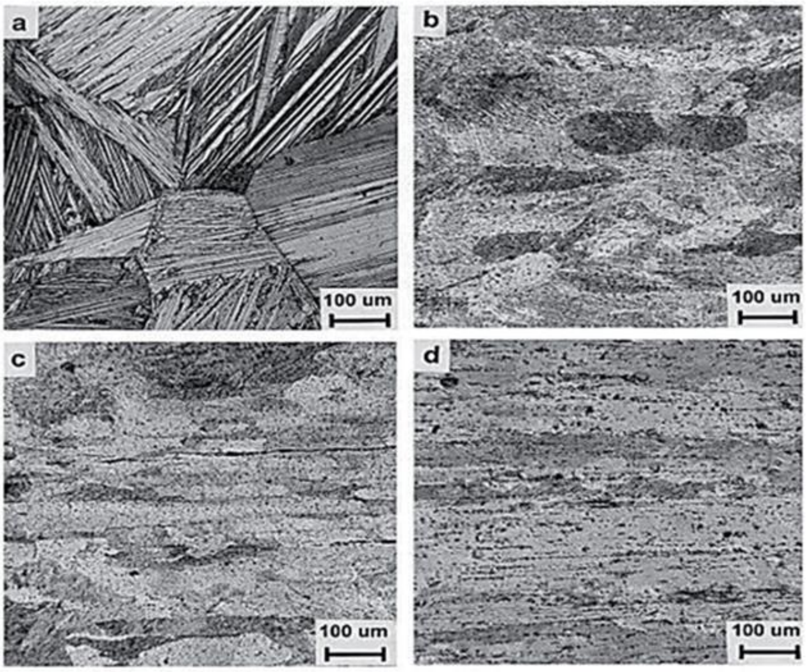

Fig.(5). Microscopic images of (a) Cu-11.85 wt.% Al-3.2 wt.% Ni-3 wt.% Mn, (b) Cu- 11.9 wt.% Al - 5 wt.% Ni - 2 wt.% Mn - 1 wt.% Ti , (c) Cu -11.4 wt.% Al-2.5 wt.% Ni-5 wt.% Mn-0.4 wt.% Ti, and (d) Cu-11.8 wt.% Al-5 wt.% Ni-2 wt.% Mn-1 wt.% Ti (Dutkiewicz (1999)).

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