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Coexistence of material science and sustainable technology in economic growth

Sisak, May 15th – 17th, 2019

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CORROSION BEHAVIOR OF CuAlMn AND CuAlMnNi ALLOY IN 0.9% NaCl SOLUTION

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Original scientific paper

Abstract

Corrosion behavior of CuAlMn and CuAlMnNi alloy ribbons, produced by melt spinning method, were investigated by electrochemical methods such as open circuit potential measurement, linear and potentiodynamic polarization method. Investigations were performed in deaerated 0.9% NaCl solution (T = 37 °C pH = 7.4). Results of electrochemical investigations have shown that CuAlMnNi alloy have higher values of polarization resistance and smaller values of corrosion current density, but in higher anodic potentials region anodic current density for CuAlMn is lower than for CuAlMnNi alloy which indicates higher dissolution of CuAlMnNi alloy. After polarization measurements CuAlMn and CuAlMnNi ribbon surfaces were investigated with light microscope and with SEM/EDS analysis and results have shown that CuAlMnNi alloy is prone to pitting corrosion, while the surface of CuAlMn alloy is partially covered with corrosion product without existence of pits.

Keywords: *corrosion, polarization, shape memory alloys, SEM/EDS analysis*

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INTRODUCTION

Cu-based shape memory alloys (Cu-SMA) have attracted considerable attention in order to be utilized in practical applications due to its favorable properties such as wide range of transformation temperature, relatively simple fabrication procedure, low production price and high thermal and electrical conductivity [1-4]. Cu-based SMAs are currently derived from three binary alloy systems i.e. Cu-Zn, Cu-Al and Cu-Sn [5]. Intensive investigations have been

performed on Cu-Zn based alloy containing alloying elements such as Al, Si, Sn, Ga or Mn as ternary alloy and Cu-Al based ternary alloys containing Ni, Be, Zn and Mn as alloying elements for their potential use [6-10].
Shortcoming of these alloys such as brittleness and low mechanical strength are closely related to the microstructural characteristics of Cu-based shape memory alloys such as coarse grain size, high elastic anisotropy and the segregation of secondary phases or impurities along the grain boundaries [11]. It has been found that CuAlMn alloy shows good strain recovery and better ductility than CuAlNi alloy which can be correlated to decreasing the degree of order of the β parent phase [4,12,13]. Also CuAlMn alloys have higher shape memory strain, larger recovery power, better ductility, and higher damping capacity [4,14].
Most of the commercial Cu-based shape memory alloys are produced by conventional casting methods. Generally there are four advantages of rapid solidification over the slow conventional solidification techniques. These are an ability to form metastable phases, increasing the solubility above the equilibrium solubility, decreasing the segregation of additions and refining the microstructure [15-17].
In this paper, results of corrosion behavior of CuAlMn and CuAlMnNi alloy ribbons, produced by melt spinning method, in 0.9% NaCl solution have been presented.

MATERIALS AND METHODS

Rapidly solidified ribbons of CuAlMn (Cu-8.3%Al-9.4%Mn in wt.%) and CuAlMnNi (Cu-13%Al-4%Ni-2.5Mn in wt.%) were manufactured with the single roll melt spinning apparatus. The cast precursors were inserted into the graphite crucible and inductively melted in Ar atmosphere and then ejected through the nozzle with the circular shape orifice on the outer surface of the cooled rotating copper wheel. The CuAlMn and CuAlMnNi alloy ribbon samples for the electrochemical measurements were prepared by cutting to the appropriate dimensions and then soldered on an insulated copper wire to gain proper electrical contact. Polirepar S protective mass was used to isolate soldered joint points to prevent the evaluation of galvanic corrosion in contact with the electrolyte. As ribbons have very small thickness its mechanical treatment by grinding and polishing could not be performed, so the surface of the electrodes was processed by ultrasonic degreasing in ethanol, washed with deionized water and immersed in the electrolyte.
Princeton Applied Research PAR M273A potentiostat/galvanostat connected with PC was used to perform electrochemical investigations. All measurements were taken in double wall glass cell which allowed maintenance of desired electrolyte temperature, equipped with saturated calomel electrode as reference electrode, Pt-sheet electrode as counter electrode and prepared working electrode. Investigations were performed in 0.9% NaCl solution pH = 7.4 and T = 37 °C. Electrolyte solution was purged with Ar for 20 minutes prior working electrode immersion in electrolyte, and purging were continued during the electrochemical measurement with very week intensity.

The evaluation of corrosion behavior of CuAlMn and CuAlMnNi alloy in 0.9% NaCl solution was performed by open circuit potential measurements (E_{OC}) in 60 minutes time period, linear polarization method in the potential region of ±20 mV around E_{OC}, with the scanning rate of 0.2 mV s^{-1} and potentiodynamic polarization method in the potential region of −0.250 V from open circuit potential to 0.7 V, with the scanning rate of 0.5 mV s^{-1}.
After the polarization measurements electrode surfaces were cleaned ultrasonically in deionized water, dry in desiccator and investigated with optical and SEM/EDS analysis.

RESULTS AND DISCUSSION

Table 1 shows the results of open circuit measurements for the CuAlMn and CuAlMnNi alloy in 0.9% NaCl solution. Measurements were started immediately after electrode immersion in electrolyte with collecting the values of electrode potential every 30 sec in 60 minutes period of time. Tabular values refer to the electrode potential values in ten minutes intervals, Table 1.

Table 1. Open circuit potential changes for CuAlMn and CuAlMnNi in 0.9% NaCl solution

Time/ Alloy	0 min	10 min	20 min	30 min	40 min	50 min	60 min
CuAlMn	-0.313	-0.321	-0.323	-0.324	-0.325	-0.325	-0.325
CuAlMnNi	-0.342	-0.334	-0.325	-0.321	-0.319	-0.318	-0.317

From the Table 1 it can be observed the opposite trend in changes of open circuit values for CuAlMn and CuAlNiMn alloys, i.e. for the CuAlMn alloy values of open circuit potential change slowly with time towards negative direction while for CuAlMnNi changes of E_{OC} were observed towards more positive values. It is also clear that stabilization of E_{OC} for both alloys occurs in period of 40 minutes and that final E_{OC} values do not differ significantly.
Linear polarization measurements were performed in order to determine the values of polarization resistance (R_p) which represents the resistance of metal to corrosion, and is defined by the slope of the polarisation curve near the corrosion potential, by the equation (1):

$$R_p = \frac{\Delta E}{\Delta i} \ (\Omega \text{ cm}^2) \tag{1}$$

Results of these investigations were presented on Figure 1 and in Table 2. It can be seen that CuAlMnNi alloy have higher curve slope then CuAlMn alloy i.e. higher values of R_p. As R_p value is reverse proportional to the corrosion current density (i_{corr}), higher value of R_p means lower value of i_{corr} i.e. lower corrosion.

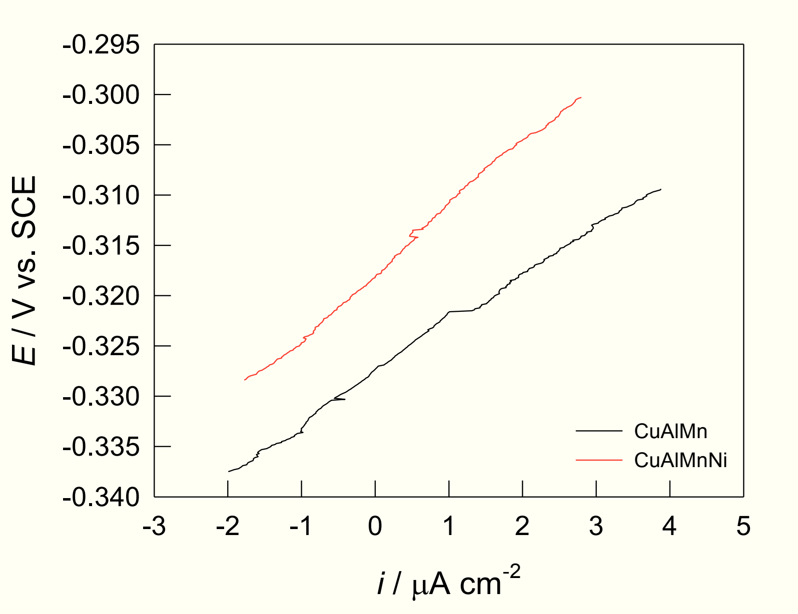


Figure 1. Linear parts of polarization curves for CuAlMn and CuAlMnNi alloys in 0.9% NaCl solution

The last applied electrochemical method was a potentiodynamic polarization method which was carried out in a wide range of potentials to gain insight into the anodic behaviour of CuAlMn and CuAlMnNi alloy in NaCl solution. Results of these investigations were presented at Figure 2 and Table 2. From the Figure 2 it can be seen different corrosion behaviour of alloys to some extent, which is manifested in anodic and cathodic parts of the curves, which is result of different alloy composition. CuAlMn alloy shows more negative corrosion potential and lower anodic current densities in higher anodic region (above 0.25 V), while in Tafel region its anodic current is slightly higher compared with CuAlMnNi alloy. After Tafel region anodic current continue to rise for both alloys and achieves its maximum after which it reduces to some extent due to formation of harder soluble corrosion compounds which act as a surface barrier and slow down the process of dissolving the alloy [18-21]. From the Figure 2 it can be seen that reduction of anodic current density for CuAlMn alloy begins earlier (on lower anodic potential) with higher reduction then CuAlMnNi alloy.

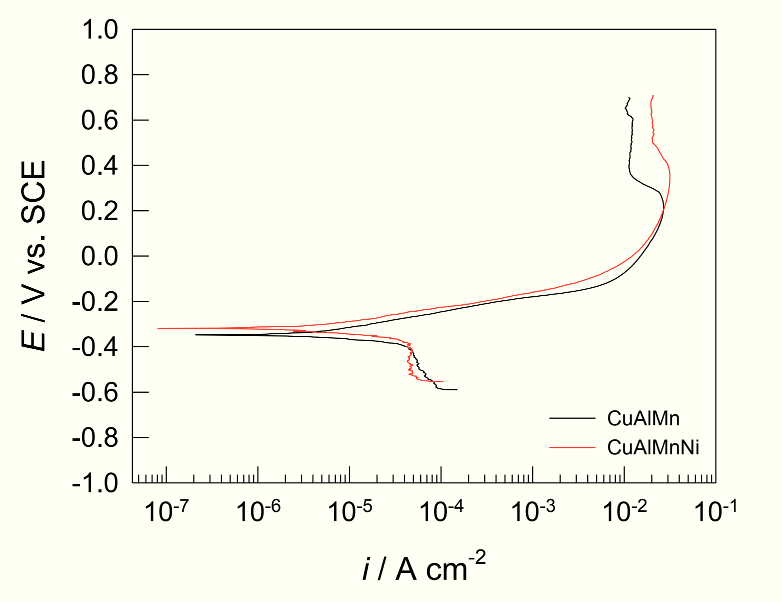


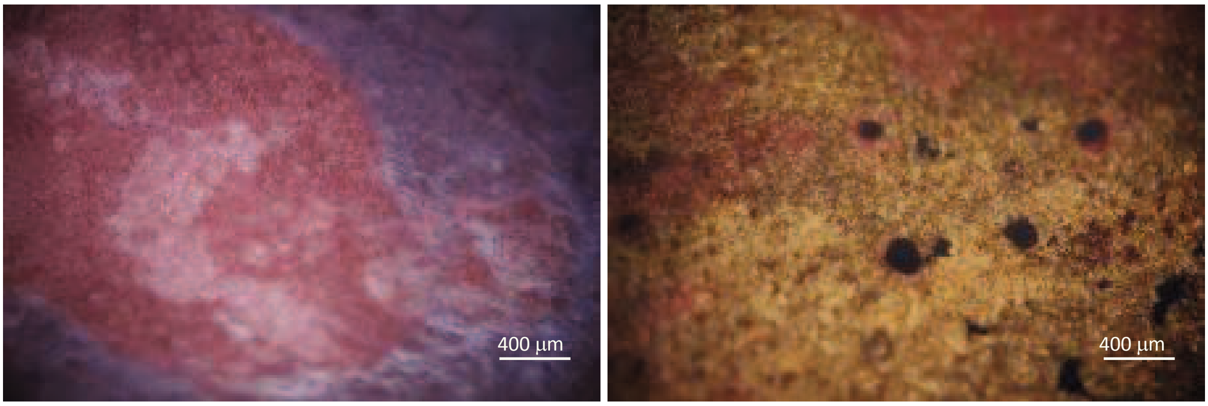
Figure 2. Potentiodynamic polarization curves for CuAlMn and CuAlMnNi alloys in 0.9% NaCl solution

Corrosion parameters for CuAlMn and CuAlMnNi alloy obtained by polarization measurements are given in Table 2.
From the Table 2 it can be seen that CuAlMnNi alloy shows lover values of corrosion current density and higher values of polarization resistance which suggest the better corrosion resistance.

Table 2. Corrosion parameters obtained from polarization measurements

Alloy	E_{corr} / V	i_{corr} / μA cm^{-2}	R_p / kΩ cm^2
CuAlMn	-0.346	3.5	4.907
CuAlMnNi	-0.322	2.28	6.445

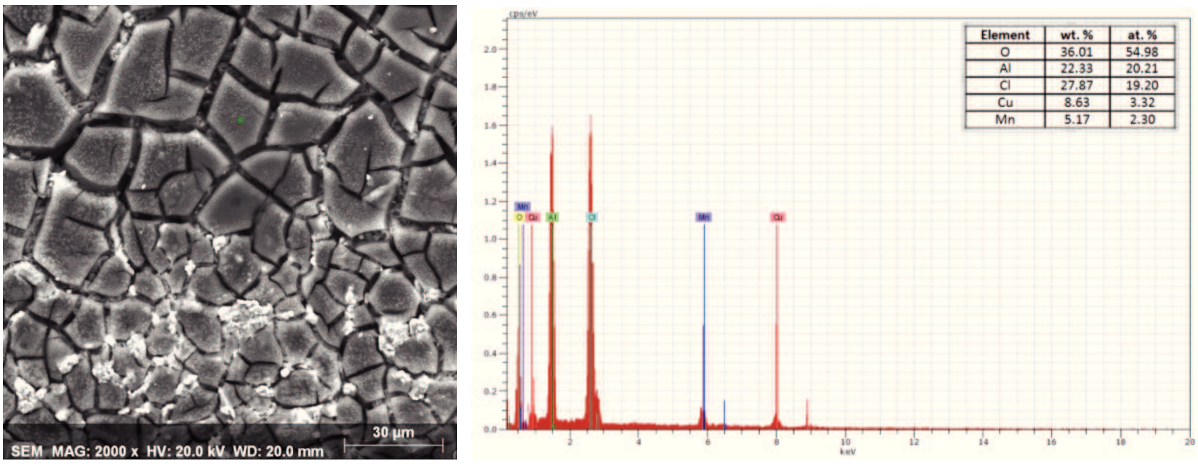
After the polarization measurements, electrode surfaces were cleaned ultrasonically in deionized water, dried in desiccator and then investigated with light microscope with magnification of 50 times. Results of these investigations are presented in Figure 3.



a) b)

Figure 3. Optical micrographs of the corroded CuAlMn alloy surface a) and CuAlMnNi alloy surface b) in 0.9% NaCl solution

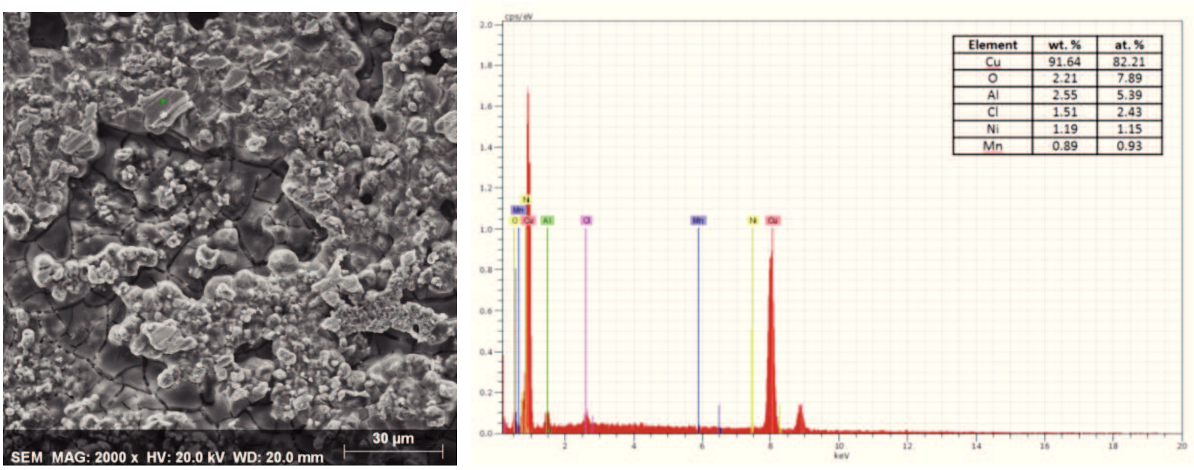
Optical images of corroded CuAlMn and CuAlMnNi surface reveal the differences in corrosion mechanism with addition of alloying element which are present in investigated alloys. On the CuAlMnNi alloy surface it can be clearly seen the existence of pitting corrosion, while the surface of CuAlMn alloy is partially covered with corrosion product without existence of pits. Earlier corrosion investigations on CuAlNi alloys have confirmed that dominant corrosion attack on the surface in chloride solution is pitting corrosion [21, 22]. Pitting corrosion is also observed on the corroded surface of the cast CuAlMnNi alloy in similar investigations in 0.9% NaCl solution but in a smaller extent compared with CuAlNi alloy [23]. It seems that the presence of Ni along with a smaller percentage of manganese and aluminium in CuAlMnNi alloy facilitates the appearance of pitting corrosion.
More detail information about surface condition of corroded alloys was obtained by SEM/EDS analysis (Figure 4 and Figure 5).



Element	wt. %	at. %
O	36.01	54.98
Al	22.33	20.21
Cl	27.87	19.20
Cu	8.63	3.32
Mn	5.17	2.30

a) b)

Figure 4. a) SEM images and b) EDS analysis of the CuAlMn alloy surface after polarization measurement in 0.9% NaCl solution



a) b)

Figure 5. a) SEM images and b) EDS analysis of the CuAlMnNi alloy surface after polarization measurement in 0.9% NaCl solution

SEM/EDS analysis has shown that the cracked surface of CuAlMn alloy consist mainly of aluminum and copper oxides and chlorides, with small percentage of copper and manganese (Figure 4). A different appearance of the surface shows the CuAlMnNi alloy on which many creases and channels are formed by the corrosion process. Copper is dominant element on the surface with the small amount of oxygen, aluminum, chlorine and manganese.

CONCLUSIONS

Measurements of open circuit potential reveal the opposite trend for CuAlMn and CuAlNiMn alloy, i.e. for the CuAlMn alloy values of open circuit potential change slowly with time towards negative values while for CuAlMnNi changes of E_{OC} were observed towards more positive values. Stabilization of E_{OC} for both alloys occurs in period of 40 minutes and final E_{OC} values do not differ significantly.

Polarization measurements of CuAlMn and CuAlMnNi alloy in 0.9% NaCl solution have shown that CuAlMnNi alloy have higher values of polarization resistance and smaller values of corrosion current density. In higher anodic potentials anodic current density for CuAlMn is lower than for CuAlMnNi alloy which indicates lover dissolution of CuAlMn alloy.

On the CuAlMnNi alloy surface it can be clearly seen the existence of pitting corrosion, while the surface of CuAlMn alloy is partially covered with corrosion product without existence of pits. It seems that the presence of Ni along with a smaller percentage of manganese and aluminium in CuAlMnNi alloy facilitates the appearance of pitting corrosion.

SEM/EDS analysis has shown that the cracked surface of CuAlMn alloy consist mainly of aluminum and copper oxides and chlorides, with small percentage of copper and manganese while surface of the CuAlMnNi alloy have many creases and channels which are formed by the corrosion process. Copper is dominant element on the CuAlMnNi surface with the small amount of oxygen, aluminum, chlorine and manganese.

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