STRENGTHENING OF ALUMINIUM-COPPER ALLOYS THIN FILMS

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Abstract : We have modelled, from characteristics of aluminium and its alloying elements, the microhardness evolution of sputtered on glass slides aluminium-copper alloys deposits. This one regularly increases with the copper concentration from 1300 MPa for sputtered pure aluminium, reaches a maximum of about 8500 MPa corresponding for 49 at.%Cu and then decreases till sputtered pure copper estimated about 2300 MPa. Such films, with a hardness higher than for corresponding conventional alloys, can be used as hard coatings for these alloys This phenomenon of significant mechanical hardening of aluminium by the means of transition elements is attributed essentially to a simultaneous effect of solid solution and grain size refinement due to the difference in the size of the atoms of the solvent and the solute.

Keywords: Sputtering; Hardness; Aluminium alloys.

1. Introduction

The addition of an alloying element to a metal reinforces the mechanical hardening of this one. The hardening observed in an alloy depends primarily on the nature of the base metal, on the additional alloying element and the mechanical alloying techniques used to enhance hardness, it is well known that the relation of Hall-Petch is the most general form, which describes the relation between mechanical hardness and the grain size in conventional alloys. This form of relation predicts that hardness evolution would increase by decreasing the grain size that is to refine microstructure by refining the grain size or including a distribution of particles. Sputtered binary aluminium- based alloys deposits as Al-Ti [1], Al-Cr [1,2] and Al-Fe [3] exhibit a microhardness much higher than corresponding conventional alloys, where hardening is essentially due to the solid solution effects on grain size refinement [4].

We have studied the microhardness behaviour of sputtered deposited binary aluminium-based alloys coatings. It is found that the microhardness varies almost in a general polynomial form. It increases from microhardness of sputtered pure aluminium, reaches a maximum and then decreases to pure alloying element. In this work we have predicted the microhardness evolution of sputtered aluminium-copper alloys deposits.

2. Modeling and discussion

Binary based-aluminium alloys deposits studied here were prepared by r.f. magnetron sputtering (0.7 Pa, 400 °K) by using composite targets in the form of disc of diameter ≤70 mm. These targets Al-Ti, Al-Cr and Al-Fe consist of a bulk or powder aluminium crown in which is inserted a disc of diameter d (d<70 mm) of compacted powder or bulk alloying element [1-3].

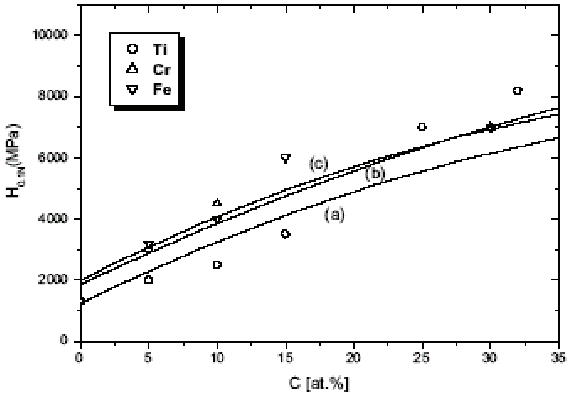

Fig. 1 : Variation of microhardness of sputtered deposits with concentration of Ti (a) [1], Cr (b) [2] and Fe (c) [3].

These sputtered deposits exhibit a microhardness (using a Vickers indenter with a load of 10 g) much higher than those observed for corresponding traditional alloys. Pure sputtered aluminium deposits have a microhardness of 1300 MPa, exceeding with 800 MPa that of bulk aluminium [1]. The microhardness of sputtered deposits increase regularly with the concentration of the additive alloying element. This microhardness reaches a maximum and then decreases to pure alloying element (**Figure 1**).

The observed evolution of the microhardness Vickers for sputtered binary aluminium-based alloys deposits with presence of titanium or chromium or copper [1-3] is almost a polynomial form :

$$H=H_0+B_1\times C+B_2\times C^2 \qquad (1)$$

The microhardness H_0 of pure aluminium is 1300 MPa and corresponds to a concentration equal to zero, this simplifies the equation 1 to :

$$H=1300+B_1\times C+B_2\times C^2 \qquad (2)$$

As the microhardness passes by a maximum, the derivative of the equation 1 in this point is zero, we can thus establish a relation between the coefficients B_1, B_2 and the concentration C_{max} corresponding to the maximum microhardness. To calculate the coefficients B_1 and B_2 of the equation 1 we use the characteristics of aluminium and its alloy elements (**Table 1**) and we plot the curve of concentration of alloying element as magnesium, titanium, chromium, iron and copper against the insert diameter d (<70 mm) in the targets (**Figure 2**).

Tab. 1. Extension of the solid solution in relation with characteristics of pure elements [3].

Element X	Al	Mg	Ti	Cr	Fe	Cu
Density ρ	2.70	1.74	4.51	7.19	7.86	8.96
Atomic weight A	26.98	24.31	47.90	51.99	55.85	63.54
Atomic diameter D (Å)	2.86	3.20	2.94	2.54	2.52	2.56
D_{Al}-D	0	-0.34	-0.08	0.32	0.34	0.30
Structure	fcc	hcp	hcp	bcc	bcc	fcc
Extension (300°K)		20	27	5	5	

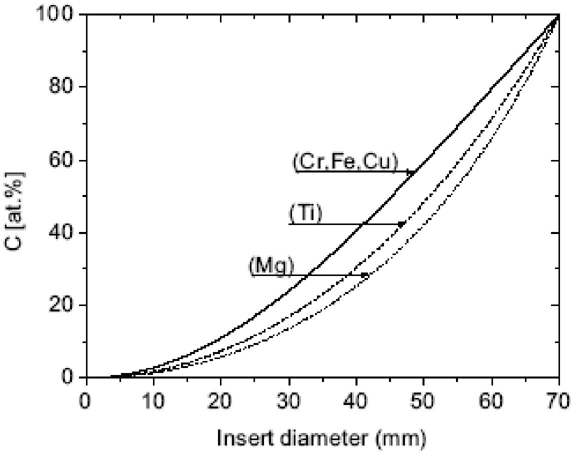

Fig. 2 : Evolution functions of alloying element concentration in the deposits with the insert diameter.

The targets are made with a bulk aluminium crown in which is inserted a bulk disc of alloying element. With this target configuration, the composition of the additional metal in the deposits C [at.%] is easily controlled. It increases in a hyperbolic curve with the insert diameter [4].

C[at.%]=(N×100)□(N+N□□) (3)

Where N and N□□□are the number of atoms of alloying element and aluminium in the target respectively.

$$C[at.\%]=R\times d^2\times 100/[R\times d^2+(490-0,1\times d^2] \qquad (4)$$

Where R is the ratio between the density and the atomic weight for each alloying element and d the insert diameter.

We observe that Al-Cu, Al-Cr and Al-Fe have almost the same curve. The limit of extension of the solid phase αAl in Al-Cu deposits must be close to that for Al-Cr and Al-Fe deposits which is about 5 at.%.

The role of relative atomic size of solvent and solute is more clearly shown when the logarithm of the slope of Hv-composition curve dH/dX (X=C) is plotted against the logarithm of the difference in atomic diameters ($\Delta D=|D_{Al}-D|$) between solvent and solute [5]. With an extension of the αAl supersaturated solid solution of about 5 at.% with the presence of Cr or Fe and 27 at.% for Ti, a linear plot is obtained (**Figure 3**) [4]. he microhardness corresponding to the αAl supersaturated solid solution about 5 at.% with Cu is then about 2700 MPa. The reduced effect of titanium appears in low limit extent range, and this is due to its diameter size which is larger than that of aluminium.

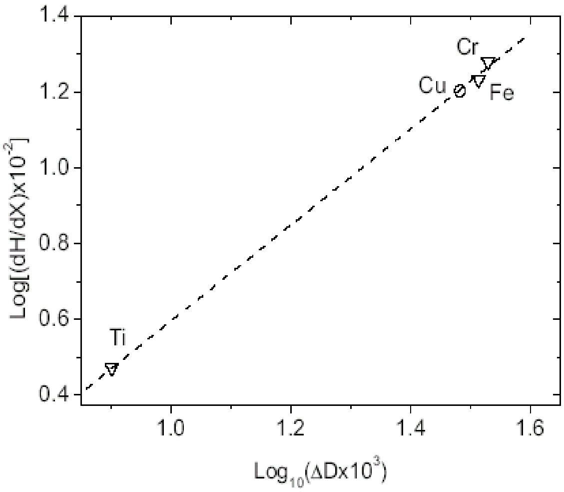

Fig. 3 : Relation between solid solution hardness and atomic diameters difference of solvent and solute in aluminium-base solid solutions.

To determine the microhardness of pulverized pure copper, we will use the mechanical properties of the aluminium and its alloying elements where sputtered pure aluminium was harder of about 800 MPa than bulk aluminium. As the microhardness varies linearly for diluted alloys and considering the properties of aluminium cited before we can then estimate the microhardness for pure copper of about 2300 MPa when the microhardness of bulk copper is taken 1500 MPa [6]. With 1300 MPa for sputtered pure aluminium, extrapolated 2300 MPa for respectively sputtered pure copper and 2700 MPa corresponding to approximately 5 at.%Cu for the limit extension of copper in aluminium, the deduced coefficients B_1 and B_2 in equation 1 are respectively 294 and –3 and the concentration C_{max} is 49 at.%Cu and the corresponding maximum of microhardness is 8500 MPa. So, the polynomial plot which governs the microhardness evolution of sputtered Al-Cu deposits with composition of Cu will be as :

$H= 1300+294\times C-3\times C^2$ (5)

The figure 4 shows the evolution of microhardness, for sputtered aluminium-copper deposits, shows that this one increases with the copper concentration, from 1300 MPa for sputtered pure aluminium, reaches a maximum of about 8500 MPa for a corresponding content of copper about 49 at.%Cu and then decreases until about 2300 MPa for sputtered pure copper.

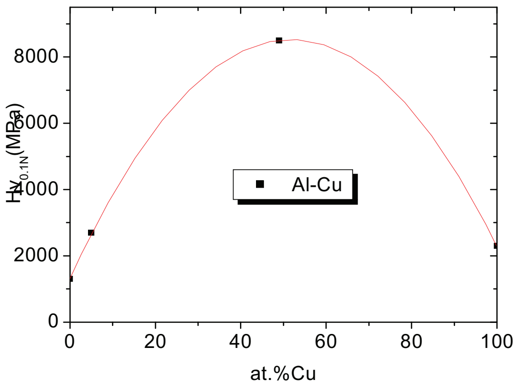

Fig. 4 : Predicted microhardness evolution with copper concentration in sputtered Al-Cu alloys deposits.

The Al-Cu deposits, with an expected microhardness up to 8500 MPa, are much harder than corresponding conventional alloys. It has been found that nitrogen incorporation into Al-Cr and Al-Ti coatings reinforces their microhardness [1]. But Al-Cu is an age hardening alloy [7,8], intermetallic precipitates as Al_2Cu will form and all changes in structure occurring under mechanical stress [9] cause discontinuities and this will not favours compactness of deposits and could decrease microhardness.

3. Conclusion

The sputtered deposits of binary aluminium-based alloys, where the alloying element is a transition element, have a microhardness, which almost evolves in a general polynomial form. The microhardness increases regularly with alloying element concentration, starting from sputtered pure aluminium, reaches a maximum and then decreases finally towards the pure alloying element. Compared with corresponding conventional alloys, sputtered deposits are harder and can thus be used as hard coatings for these latter. This phenomenon of hardening of as-sputtered deposits is essentially attributed to the difference in the size of the atoms of the solvent and the solute.

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