Intense Pulsed Ion Beam Mixing of Al, Ti, Ni, Cu Film/Substrate Systems[1]

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Abstract – Intense pulsed ion beam (IPIB) benefits mixing more than traditional ion beam does because of its high power density and high energy deposition density. It may useful for forming surface with excellent serving properties. In this work, the IPIB mixing effect of different film/substrate combinations, Al/Ti, Ti/Al, Ni/Al, Ni/Ti and Ni/Cu were studied. It was found that the mixing effect and the film material depth distribution are independent of the solubility of two materials of film and substrate, but obviously inversed relate to the difference on some thermodynamic parameters of them. Mass loss of film material was serious during IPIB irradiation. However, the thickness of mixing layers of all combinations are over 1 μm, although in some case only a small amount of film materials were retained. The IPIB mixing mechanisms and the reason of mass loss of film materials are discussed.

1. Introduction

Intense pulsed ion beam (IPIB) mixing was the first study object of IPIB application some 20 years ago [1]. It was reported that some combinations, such as Ti/Al, Au/Cu, Co/Si, Ti/Si,, Ag/Si, Mo/Si, Pt/Si, Cu/Mo, Pb/Fe, Cr/Cu, Hf/Al alloy, Si/Al alloy, Pt/Ti alloy, etc, mixing by IPIB bombardment [1–6] were studied. Mixing layers were achieved for some of above-mentioned combinations, but not for all of them. The basic rules of forming mixing layers by IPIB bombardment are not clear yet.

Considering that the materials experienced IPIB irradiation all underwent intense thermodynamic process, we choose material combinations with various differences of thermodynamic parameters to study the relation between their thermodynamic parameters and their mixing effect.

2. Experimental

For simplifying the research model, 4 pure metal Al, Ti, Ni, and Cu were chosen as test materials. Some important thermodynamic parameters of them are list in Table I.

Table I. The melting point T_m, the thermal expansion coefficients α and the surface tensions at the melting point σ

Material	T_m (K)	α (ppm/K)	σ (N/m)
Al	933	23.03	0.84 [7]
Ti	1943	8.35	1.57 [7]
Ni	1726	13.30	1.73 [8]
Cu	1358	16.50	1.30 [7]

Table II. The film thicknesses, the peak current intensity (I_p) of IPIB, and the numbers of shooting for different samples

film/substrate	film thickness (nm)	I_p (A/cm^2)	sample's group no.		
			1 shot	2 shots	5 shots
Al/Ti	500**	60	Ti01	Ti02	Ti03
Ti/Al	300*	100	Al01	Al02	Al03
	400**		Al04	Al05	Al06
	500***			Al08	Al09
Ni/Al	150*	100	Al10	Al11	Al12
	300**		Al13	Al14	Al15
	450***		Al16	Al17	Al18
Ni/Ti	150*	100	Ti09	Ti10	Ti11
	300**		Ti13	Ti14	Ti15
Cu/Ni	150*	100	Ni01	Ni02	Ni03
	300**		Ni07	Ni08	Ni09
	450***		Ni13	Ni14	Ni15

* thinner than the ion range in film material
** approximated to the ion range in film material
*** thicker than the ion range in film material

Five kinds of combinations were combined as following (film/substrate):

a. Al/Ti: big differences between their thermo- dynamic parameters, and the melting point of the film is much lower than the one of the substrate;
b. Ti/Al: big differences between their thermo- dynamic parameters, and the melting point of the film is much higher;
c. Ni/Al: big differences between their thermo- dynamic parameters, and the melting point of the film is much higher;

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d. Ni/Ti: their thermodynamic parameters are closed;
e. Cu/Ni: small differences between their thermodynamic parameters, and the melting point of the film is lower.

The substrates of samples were mirror polished and cleaned by ultrasonic bath in ethanol and then in deionized water. Films deposited in different thickness (Table 2) by means of magnetron sputtering.

IPIB irradiation was carried out with TEMPII accelerator at High Voltage Institute, Tosmk Polytechnic University, Russia. The IPIB is C^+ with energy of 250 keV and pulse duration of 60ns. The peak value of current density were chosen to be sure that the surface layer would be melt but far from being evaporated (Table 2). The fluence in one pulse was about 10^{13} cm^{-2}.

3. Mixing effect

The mixing effects were analyzed with Rutherford Backscattering Spectroscopy (RBS), optical microscope, scanning electronic microscope (SEM), and energy dispersive X-ray spectroscopy (EDX).

According to the RBS analysis, we found that mixing layer is formed in every combination, and the thickness of the mixing layers are all thicker than 1 m, but the mixing effect is quite different.

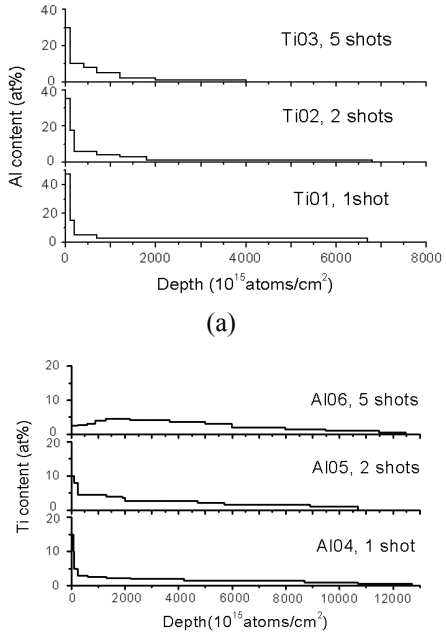

Fig. 1. (a) Depth distributions of Al concentration in respective Al/Ti samples; (b) depth distributions of Ti concentration in respective Ti/Al samples

Fig. 1 shows if the difference of thermodynamic parameters of film and substrate is big, the profile of depth concentration of film material in the mixing layer is L shape. The Ni/Al combination has similar mixing effect.

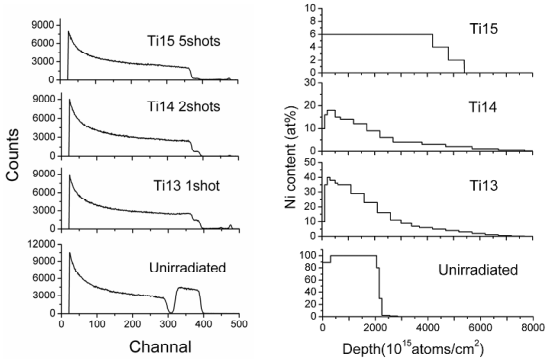

Fig. 2. (a) Backscattering spectra and (b) depth distributions of Ni concentration in respective Ni/Ti samples

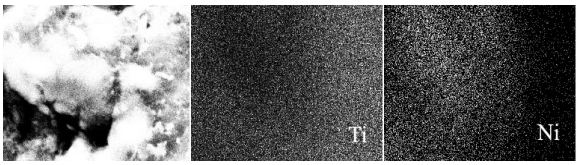

Fig. 3. EDX maps of Ti13 cross-section. The left side is surface. The graph scale is 3 m

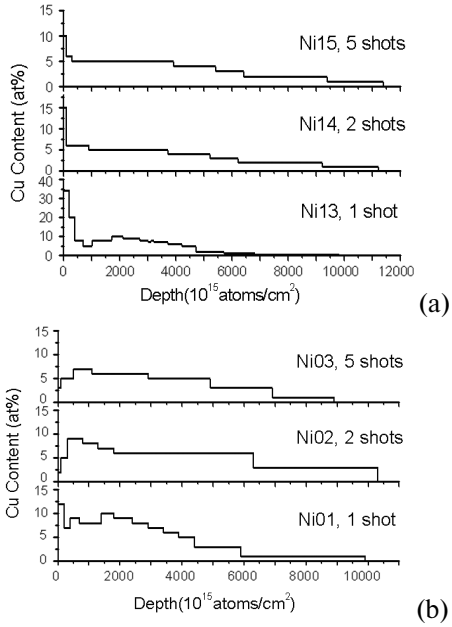

Fig. 4. Depth distributions of Cu concentration in respective Cu/Ni samples: (a) original Cu film thickness is approximated to the ion range in Cu; (b) original Cu film thickness is thicker than the ion range in Cu

In Fig. 2 it can be seen that the gradient depth distribution of film material concentration in the mixing layer is formed, if the thermodynamic parameters of film and substrate are closed. This gradient depth distribution can be also seen clearly in the EDX maps of sample Ti13 (Fig. 3). Popp et al reported a similar result for Cr/Cu combination in reference [4].

If the difference of thermodynamic parameters of film and substrate is small, their mixing effect will be somewhat between above two cases. Fig. 3, *a* looks more like Fig. 1, *a* and Fig. 3, *b* is more like Fig. 2, *b*.

It is obvious that the effect of one IPIB pulse mixing is the best, whatever the target condition is.

We define the IPIB mixing rate $\chi(J^{-1}\cdot cm^{-2})$:

$$\chi = \frac{N \cdot x}{\varepsilon}, \quad (1)$$

wherein N (cm^{-2}) is the total amount of film material that is mixed into the substrate in unit area, x (cm^{-2}) is the mass thickness of the mixing layer, and ε (J/cm^2) is the energy deposit in unit area.

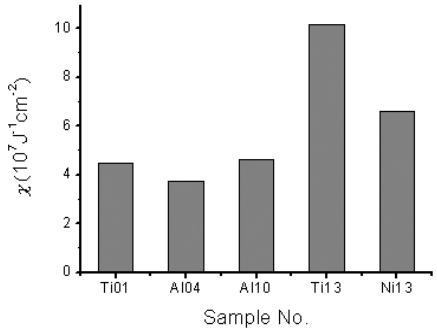

Fig. 5. The highest mixing rates of different combinations

The mixing rates of samples Ti01, Al04, Al10, Ti13 and Ni13 are highest in their respective combinations. Then we get Fig. 5. Here we found that the mixing rate has reversed relativity with the difference of thermodynamic parameters between film and substrates.

4. Discussion

According to the different mixing effect for different combination, we supposed there are different mixing mechanisms.

In the case of combination a, b and c, the film and the substrate can hardly be melted simultaneously during the first shot of IPIB [9], so the mixing process must happen in liquid-solid or solid-liquid states. Imitating the low current ion beam mixing to define a "equivalent diffusion coefficient" D [10]

$$Dt = \sigma^2/2, \quad (2)$$

where t is irradiation time and σ is the half width of the Gaussian fitted depth distribution of film material concentration.

We found that the "L shape" depth distribution can be consider as two Gaussian distributions that related to different equivalent diffusion processes, respectively: one is caused by collision regime, which including "ballistic regime" and "thermal spike effect" and in only several tens nm range; the other may be the enhanced diffusion driven by the non-equilibrium thermodynamic process induced by IPIB irradiation that can affect a thickness range of m. The "equivalent diffusion coefficients" for Gaussian fit 1 and Gaussian fit 2 in fig. 6 are D_1=9.0·10^{-5} cm^2/s and D_2=0.13 cm^2/s. Such a "diffusion coefficient" can only be a value for relative comparing, not make physical sense.

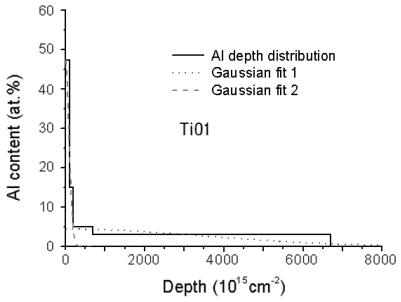

Fig. 6. The "L shape" depth distribution of film material concentration in mixing layer can be decomposed as two Gaussian distributions that related to two different equivalent diffusion processes, respectively

In the case of combination d, the film and the substrate can be melted simultaneously. The liquid state mixing process, such as diffusion and convection in liquid state, may take place. So the highest mixing rate is achieved.

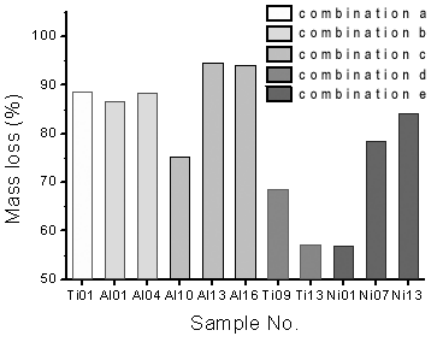

Fig. 7. Film material mass loss after one pulse IPIB irradiation

The mass loss of film material is serious to every combination (Fig. 7). In most cases, peeling off and ablation of film material are main reasons of mass loss, because their thermal expansion coefficients, melting points and the surface tensions at the melting point are quite different with their substrates ones. Normally the thicker the film is, the more mass loss is. In contrast, for the combination with closed thermodynamic parameters (Ti09 and Ti13), the mass loss goes down when the film is thicker. It implies that no peeling off took place; the only reason of mass loss is ablation.

5. Conclusion

IPIB bombardment can induce metallic film/substrate mixing effectively. The mixing rate, the depth distributions of film material in mixing layer, and the film material mass loss strongly rely on the differences between thermodynamic parameters of film and substrate.

The IPIB mixing mechanisms may be different to different film/substrate combinations. Diffusion and convection in liquid state is a reasonable explanation to the mixing of combinations with closed thermodynamic parameters. Collision plus enhanced diffusion driven by nonequilibrium thermodynamic process induced by IPIB irradiation may be more suitable mixing mechanism to those combinations that the thermodynamic parameters of their film and substrate are quite different.

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