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Study of the effect of chopping on the adhesion of codeposited mixed thin films

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Abstract. This paper reports adhesion of codeposited mixed films of Cu–Ag, Cu–Al, MgF_2–ZnS and MgF_2–cryolite and enhancement of adhesion by chopping technique. Results indicate that codeposited mixed films show higher adhesion than single films and chopping improves the adhesion further. The quenching of crystal growth seems to be more effective if two materials are codeposited and chopped. Chopping along with mixing increases the number of nucleation sites and decreases defects in the film. Decreasing defects and modifying the microstructure increases the adhesion of the films. Chopping also seems to increase oxygen affinity of both metallic and dielectric films for growth of interfacial bonding layer.

Keywords. Chopping; codeposition; mixed films; adhesion; non-chopped.

1. Introduction

Metallic and dielectric films are widely used in microelectronics and optical thin film systems. The designers of these systems have a very limited selection of materials. Mixed films offer a wider choice of materials. The most direct method is to prepare a mixture of the materials and deposit from a single source (Hiradayanath *et al* 1979; Vankar *et al* 1979; Ganner 1986). Simultaneous evaporation using several sources (Jacobson 1966) is a very flexible method. Our work (Vijaya *et al* 1980, 1987; Puri *et al* 1983) on codeposited mixed MgF_2–cryolite films has shown the superiority of chopped mixed films over single films.

In order to be functional the thin film has to be adherent to the substrate. Various methods like deposition on hot substrate (Pulkar 1982; Martin *et al* 1983; Pierce and Vaugham 1983), deposition in presence of plasma or glow discharge cleaning (Stoddart *et al* 1970; Fancey and Mathews 1990; Pulkar *et al* 1990), deposition of bonding layer (Mattox 1973), etc have been reported to increase the adhesion of the films.

This paper reports the adhesion of codeposited mixed films of Cu–Ag, Cu–Al, MgF_2–ZnS and MgF_2–cryolite and the effect of chopping (Vijaya *et al* 1980) on the adhesion of these films.

2. Experimental

Both metallic and dielectric films were deposited on glass by thermal evaporation (~ 7 Å/s) in a vacuum of better than 8×10^{-6} torr using two tungsten filaments for codeposited mixed films. Figure 1 gives the schematic diagram of the arrangement for deposition of mixed films and single films in the same vacuum cycle. The two materials were in the ratio 1:1 by weight. The two sources were kept opposite to each other at a distance of 10 cm. A metal partition was kept between the two sources to avoid

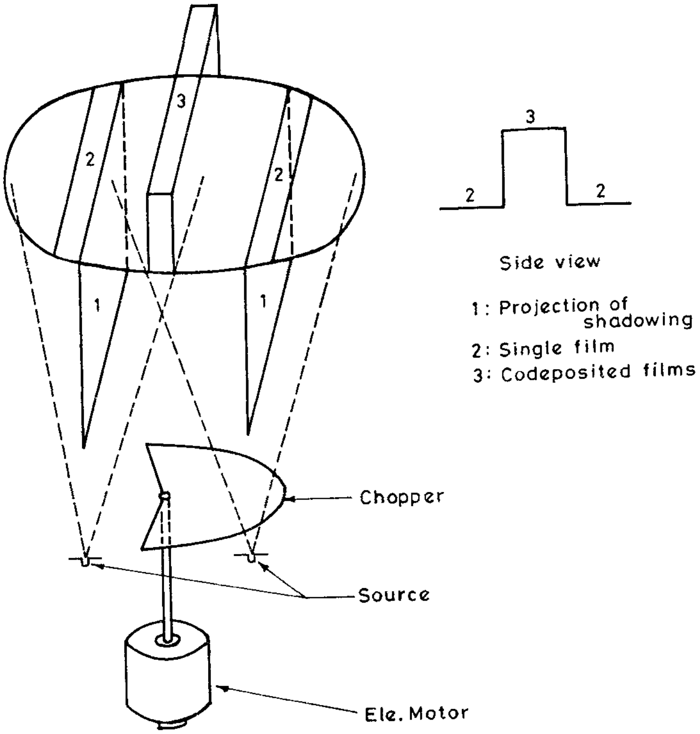

Figure 1. Schematic diagram of substrate holder along with chopper.

contamination of source. The height of the substrate holder was adjusted such that both codeposited mixed and single films had the same thickness. The substrates were glass slides maintained at room temperature during deposition. The thickness of the films was in the range 500–2500 Å. The chopper was a circular metallic vane with a V-shaped cutout kept above the evaporation source (figure 1) giving a chopping rate of 5–6 rot/sec. The adhesion was measured by direct pulloff (DPO) method by sticking aluminium studs of 0·5 cm diameter to the film surface and back of substrate and pulling by tensile testing machine until fracture occurred at the interface. The error in the measurement using the adhesion testing machine (Puri *et al* 1991) was $\pm 9{\cdot}81\,\mathrm{kgF/cm^2}$.

3. Results and discussion

The typical values of adhesion of codeposited mixed metallic and dielectric films as a function of thickness are shown in figures 2 and 3 respectively. Table 1 gives the average values of adhesion of codeposited mixed and single films. From the graphs and table it is seen that there is an appreciable increase in adhesion of the codeposited mixed films due to chopping. An interesting result is that non-chopped mixed films show higher adhesion than non-chopped single films, the exception being Al. Both metallic and dielectric films of thickness > 1000 Å show near-saturation effect as

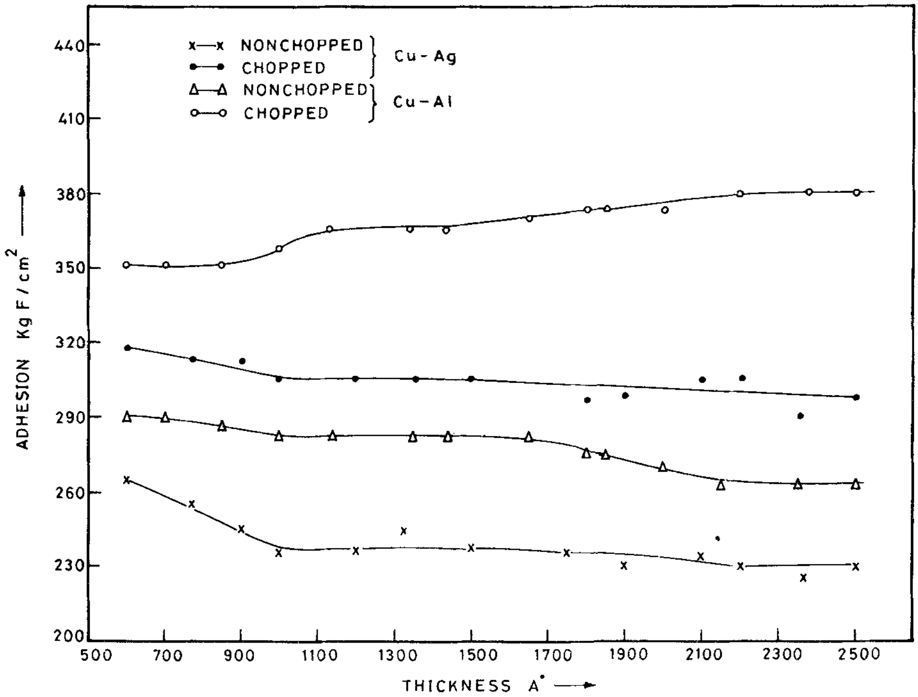

Figure 2. Typical values of adhesion vs thickness of codeposited mixed metallic films.

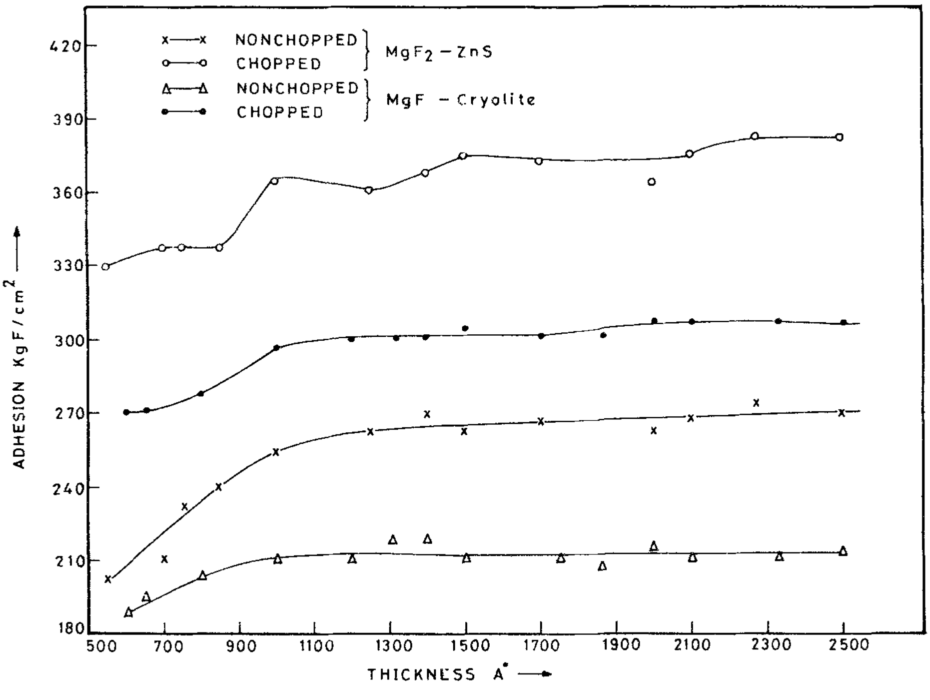

Figure 3. Typical values of adhesion vs thickness of codeposited mixed dielectric films.

Table 1. Average values of adhesion of codeposited mixed and single films.

		Adhesion (kgF/cm^2) Thickness range (Å)					
		500–1000		1000–1500		1500–2500	
Type of film		c	nc	c	nc	c	nc
	Cu–Ag	310	250	305	238	300	238
Codeposited mixed	Cu–Al	350	285	365	280	370	270
	MgF_2–ZnS	338	225	370	260	375	270
	MgF_2–cryolite	280	198	300	210	310	210
	Cu	280	145	210	105	208	105
	Ag	289	153	228	100	228	100
Single film	Al	353	314	280	275	280	275
	MgF_2	185	110	206	125	206	127
	ZnS	298	220	314	255	333	255
	Cryolite	162	98	193	110	193	110

C, Chopped; nc, non-chopped.

adhesion does not change appreciably with thickness. From figure 3 it is seen that thinner dielectric films have lesser adhesion than thicker films.

During codeposition a pseudodiffusion type of interface might be formed (Mattox and Rigney 1986) which might be the cause of enhanced adhesion of codeposited mixed films. It has been reported (Jacobson 1966) that evaporated mixtures have a finer structure than that of the individual materials. The films of Ag, Al, MgF_2, ZnS and cryolite have been observed to have columnar microstructures (Guenther and Jung 1976; Pulkar 1982).

During the process of chopping there is a continuous growth flux interruption. Due to this interruption there is a quenching of the columnar crystal growth. It has been reported (Ogura 1975) that pseudoboundaries are formed when there is a discontinuity in the rate of deposition. Formation of pseudoboundaries leading to pseudodiffusion becomes more prominent if two materials are codeposited and chopped giving more adherent films.

The chopping process also enhances surface mobility of adatoms whereby the defects are not frozen at the site of impingement. Due to the arrival of two adatoms of different species during codeposition the number of nucleation sites increases and constant flux interruptions prevent growth of large crystallites. Also due to the presence of different species there are chances of recrystallization thereby ordering the film microstructure. The net effect of these processes is to decrease the voids in the film due to lateral surface diffusion and change in the columnar morphology of the films. Since defects are not frozen and there is a decrease in grain size due to chopping, fractures do not propagate for large distances in the film. These contribute to the increase in adhesion of the film. Enhanced adhesion due to increased mobility, decreased void density and lateral surface diffusion has been reported (Martin *et al* 1983) for deposition on heated substrate. Our values of adhesion of chopped single films compare very well with those of deposition on heated substrate (Vijaya *et al* 1993).

Another process dominant while chopping is to assist the affinity of adatoms to oxygen. Our electron diffraction (Puri *et al* 1983) data for MgF_2 and cryolite show the presence of oxide in a complex form in the chopped films. The ESCA of MgF_2 and CeO_2 (unpublished work) also show excess oxygen in chopped films. Excess oxygen has been reported (Laugier 1981) to decrease stress in films. Decrease of stress increases the adhesion (Jacobs *et al* 1986). This oxygen incorporation mechanism is more dominant in chopped metallic films, except single aluminium films. Aluminium being an oxygen active metal, during deposition oxygen migration takes place in the Al–glass interface forming a bonding layer (Laugier 1981). The chopping process does not seem to enhance this effect in Al, since single chopped Al films do not show appreciable increase in adhesion compared to non-chopped films. Chopped Cu–Ag films show an appreciable increase in adhesion indicating the presence of interfacial bonding layer. The presence of excess oxide is also shown in DC resistivity data of these films (Vijaya *et al* 1993). The DC resistivity of chopped copper is 3·97 μohm cm and that of non-chopped copper 1·89 μohm cm, whereas for non-chopped and chopped aluminium the values are 5·2 and 5·6 μohm cm respectively. It has been reported (Sun 1973; Nagano 1980; Craig and Harding 1981) that there is a reduction in grain size and change in morphology due to increase in DC resistivity. These facts give validity to the argument that due to chopping there is change in morphology of the films, a decrease in grain size and enhancement of oxygen affinity. The refractive index and packing density of chopped films are higher than those of non-chopped films (Puri *et al* 1983) indicating a close packed structure formation due to chopping.

A combination of all the above processes seems to be operating in chopped films which is more effective when mixed films are codeposited, the net effect being increase in adhesion of the film.

4. Conclusions

Our studies indicate that mixing by codeposition gives films with better adhesion than single films, the process of chopping enhancing this. The chopping process seems to increase surface mobility, reduces defects and decreases grain size, whereby quenching of crystal growth takes place due to which a more compact film structure is obtained which increases the adhesion of the films. The presence of two different adatom species during codeposition enhances the above effect. Increase in oxygen affinity also seems to be a very dominant effect of chopping.

Chopping along with mixing by codeposition might give wide scope for tailoring of film properties both for microelectronics and optical purposes. The fact that chopped codeposited mixed films age the least (Vijaya *et al* 1987) and that chopped mixed films have higher adhesion indicate the use of chopping technique as a technologically useful process for functional coatings. Non-destructive analysis of the interface may give better insight into the adhesion process due to chopping.

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