Computer modeling of ZnO-based optical multilayer coatings

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Abstract – A first principles computer modeling study is described which investigates the atomistic properties Al:ZnO and its adhesion to silver in an optical multilayer coating. It is shown that hydrogen, which is inevitably present during fabrication, weakens the interface between pure ZnO and Ag causing de-lamination under stress. However, by doping the ZnO with a small quantity of Al, the amount of adsorbed hydrogen can be reduced and the residual stress in the layer can be relieved. The calculations provide an atomic mechanism for the observed reduction in coating defects when the ZnO layer is replaced by Al:ZnO.

Optical multilayer coatings used for solar control often contain thin films of ZnO bonded to a low-emissivity metal such as silver. The interface between the oxide and the metal is relatively weak and due to the presence of residual compressive stresses is subject to de-lamination and corrosion [1]. The present study is motivated by the observation that replacing ZnO with Al:ZnO can inhibit interfacial de-bonding and hence strengthen the coating and improve its optical properties. However, the strengthening mechanism at the atomic level is unclear and is complicated by the gaseous environment during fabrication (magnetron sputtering) or the adsorption of gases afterwards while the coating is in service [2]. In this paper a first principles computer modeling study is described which investigates the chemical and mechanical effects of joining a (0001) layer of Al:ZnO to a (111) layer of Ag [3].

First, the defect chemistry and structure of bulk Al:ZnO (8.3 at.% Al) is studied where it is found that Al prefers to substitute on Zn sites in both oxygen rich and poor conditions. It is also found that this substitution partially relieves the stress in ZnO when it is subjected to compression in agreement with atomic size considerations. Then the surface segregation properties of an Al substituent is investigated where it is found that there is a strong driving force for segregation to an O-terminated ZnO (0001) surface but not to a Zn-terminated surface. Thus if Al is going to affect the (0001) surface properties of ZnO it will only be for the case of O-termination. However, the conditions under which the coating is fabricated are oxygen-rich and therefore O-termination is likely to be prevalent.

Following this the adsorption of hydrogen to the O-terminated Al:ZnO (0001) surface is studied where it is found that the presence of Al in the sub-surface layer reduces the adsorption energy by nearly 80%. Thus the coverage of hydrogen on this surface is expected to be significantly less than on the pure ZnO (0001) surface. Accordingly, there should be less H at the Ag/Al:ZnO interface following deposition of the Ag.

Finally, the strength of the Ag/Al:ZnO interface is studied through work of separation and binding energy calculations and these show that the presence of Al at the O-terminated interface significantly weakens it. This is supported by a Mulliken population analysis, the density of states and electron density difference maps. Only the strength of the Zn-terminated interface is enhanced by the presence of Al but the surface calculations predict that very little Al will accumulate at this interface. The Zn-terminated interface is unlikely to be present anyway because of the deposition conditions. Focusing then of the O-terminated Ag/Al:ZnO interface it is seen that although the incorporation of Al weakens it (the work of separation reduces from 1.54 J/m^2 to 0.66 J/m^2) the resulting value is still significantly higher than that found for the Ag/ZnO interface with adsorbed hydrogen (0.19 J/m^2). Thus one of the roles of Al is to eliminate hydrogen from the ZnO surface thereby reducing the number of de-lamination defects, increasing the strength of the Ag/Al:ZnO interface and improving the visual appearance of the coating. Therefore in conclusion, the effect of aluminium incorporation into the ZnO layer is two-fold: it relaxes the residual stresses in the coating but also improves the chemical bonding at the metal/oxide interfaces by removing the weakening effects of gaseous adsorption.

References

[1] Z. Lin and P. D. Bristowe, Phys. Rev. B 75, 205423 (2007)
[2] Z. Lin and P. D. Bristowe, J. Appl. Phys. 102, 103513 (2007)
[3] Z. Lin and P. D. Bristowe. J. Appl. Phys. to be published