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Electrical and Raman Studies of Bilayer Mg/Co and Mg/Mn Thin Film Metal Hydrides

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Abstract- Bilayer Mg/Co and Mg/Mn (700nm) thin films were prepared using thermal evaporation method at pressure10-5 torr at room temperature. The films were rapid thermal annealed (RTA) using halogen lamp to get a homogeneous structure of thin films. The hydrogen gas was introduced in hydrogen chamber, where samples were kept at different pressure from 10 to 40 psi of H2 for thirty minutes. The conductivity has been found to be decreased with increasing pressure of hydrogenation and also intensity of Raman peaks is decreased. I-V characteristics and Raman spectroscopy of annealed hydrogenated thin films have been studied to find out the effect of hydrogenation.

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ELECTRICALANDRAMANSTUDIESOFBILAYERMGCOANDMGMNTHINFILMMETALHYDRIDES

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Electrical and Raman Studies of Bilayer Mg/Co and Mg/Mn Thin Film Metal Hydrides

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Abstract- Bilayer Mg/Co and Mg/Mn (700nm) thin films were prepared using thermal evaporation method at pressure10^{-5} torr at room temperature. The films were rapid thermal annealed (RTA) using halogen lamp to get a homogeneous structure of thin films. The hydrogen gas was introduced in hydrogen chamber, where samples were kept at different pressure from 10 to 40 psi of H_2 for thirty minutes. The conductivity has been found to be decreased with increasing pressure of hydrogenation and also intensity of Raman peaks is decreased. I-V characteristics and Raman spectroscopy of annealed hydrogenated thin films have been studied to find out the effect of hydrogenation.

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I. Introduction

Metal hydride technologies have reached more practical and applied stages in recent years. The hydriding/dehydriding kinetics of metal hydrides are relevant to areas of design and applications of various metal hydride devices, especially in energy conversion devices such as heat pumps, refrigerators, automobiles, power generators, batteries and thermal energy storage units [1]. Several methods have been employed to overcome the kinetic limitation and/or thermodynamic stability of Mg-based hydrogen storage materials, including surface modifications addition of catalysts, and formation of metastable structures or multi-phase materials. Transition metals have shown good catalytic effects on hydrogen desorption of MgH_2 after being mechanically milled with the hydride [2]. Electrical measurements such as current/voltage provide detailed information about the electronic effects of hydrogen. A "universal alignment" model successfully describes the electronic behavior of hydrogen in a wide range of materials and allows for prediction for materials in which the role of hydrogen is yet to be explored [3, 4]. The interaction of hydrogen in a metal hydride can be understood by using different models, i.e. the anionic model of Wallace [5, 6], the protonic model of Westlake [7] and the band covalent model as described by Switendick [8]. In the present work the current-voltage measurements and Raman spectroscopy with hydrogen pressure have been presented. Current-voltage and Raman characteristics have been studied to study role of hydrogenation in thin films.

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II. Experimental

The samples were prepared by thermal evaporation method using vacuum coating unit. The HIND HIGH VACUUM unit was used for this purpose and vacuum chamber contains pressure of the order of 10^{-5}. Mg granules (99.999%), Mn powder (99.98%) & Co powder (99.998%) pure were purchased from Alfa Aesar, Jonson Matthay Company, U.S.A. used for the present study, is placed into different boats in the vacuum chamber. The source to substrate distance was kept 15 cm. The bilayer thin films of Mg/Co and Mg/Mn (700nm) have been performed by stacked layer method in situ evaporation. The thickness of film measured by Quartz crystal thickness monitor. The thin films of Mg/Co and Mg/Mn were rapid thermal annealed (RTA) by Halogen light lamp (1000 W) to get homogeneous mixture. For this process thin films were kept in quartz tube and then annealed by halogen light lamp at 400ºC for 30 minutes in atmospheric conditions for mixing to get homogeneous structure and interdiffusion of thin films of Mg/Co and Mg/Mn. Hydrogenation of Mg/Co and Mg/Mn thin films have been performed by keeping these in hydrogenation cell, where hydrogen gas was introduced at different pressures (10-40 psi). Transverse I-V characteristics of as grown hydrogenated and annealed hydrogenated samples have been recorded using Keithley-238 high current source measuring unit. The applied voltage was with in the range of -2.0 to +2.0 volts with increasing step of 0.1 volt. For I-V characteristics, electrode contacts have been made using silver (Ag) paste on the thin films. I-V characteristics of thin films have been monitored with the help of SMU Sweep computer software. Raman spectra of annealed and hydrogenated Mg/Co and Mg/Mn samples are taken by a continuous wave-Green laser with a constant wavelength 532 nm by help of R-3000 Raman system. All the measurements have been performed at room temperature.

III. Results and Discussion

Fig.1. shows that the I-V characteristics of pristine Mg/Co and Mg/Mn bilayer thin films have found to be ohmic. But the conductivity has been found to slightly smaller in case of Mg/Mn thin films. Fig.2&3 shows I-V characteristics curves for annealed Mg/Co

and Mg/Mn thin films, which shows the effect of annealing on the thin film structure indicating the possibility of mixing of structure at the interface showing the partially semiconducting behavior in I-V characteristics and The conductivity has been found to be decreased from 9.4308×10^{-5} to 4.014×10^{-5} Ω^{-1}-m^{-1} for Mg/Co and from 8.310×10^{-5} to 1.986×10^{-5} Ω^{-1}-m^{-1} for Mg/Mn thin films with increasing pressure (10-40 psi) of hydrogenation. It means hydrogen passivated defects at interface or it takes electrons from the conduction bands of each of the samples during the hydrogen absorption process and blocks the flow of charge carriers across the interface and current decreases in forward as well as reverse direction. The electronic passivation of host impurities induced by atomic hydrogen in semiconductor well agrees as reported by Pankove et al [9]. Hydrogen interacts with metals and semiconductors and takes electron from conduction band of metal as anionic model. This similar to our earlier work [10] in witch we found that the electrical conductivity of CdTe/Mn bilayer thin film decreased from 1.88×10^{-5} to 4.80×10^{-6} $\Omega^{-1}m^{-1}$ in the case of annealed samples. And also Rusu et al [11] show that the electrical conductivity of CdTe semiconducting thin film are of ranged from 10^{-6} to 10^{-4} Ω^{-1}-m^{-1}. The conductivity decreases with hydrogen pressure in both cases of Mg/Co and Mg/Mn thin films. But the current are slightly smaller in case of Mg/Mn thin film. This indicates that the Mn accelerates the hydrogen absorption rate and absorption capability of Mg/Mn thin films and Mn promotes the hydrogen absorption activation. This is similar to an earlier work by Singh et al. [12]. This found that the activation of FeTi by the mixing of Mn was found to promote the absorption rate and remove the slow rate period of hydrogen absorption in FeTi thin films.

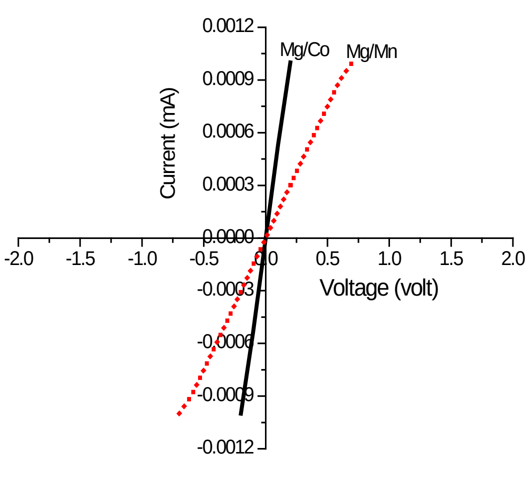

Figure 1 : I-V characteristics of as grown bilayer Mg/Co and Mg/Mn thin films

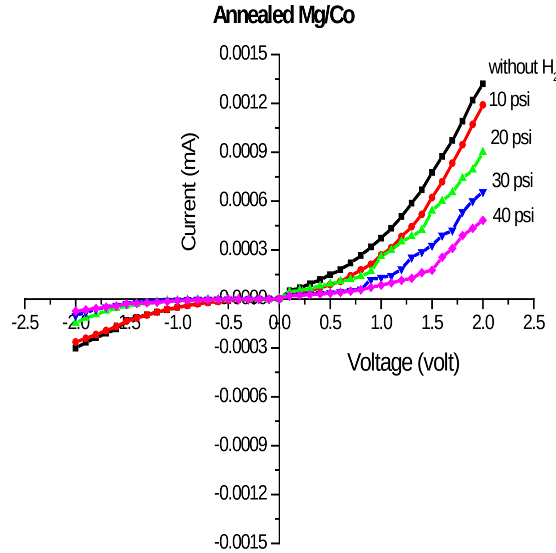

Figure 2 : I-V characteristics of annealed bilayer Mg/Co thin films

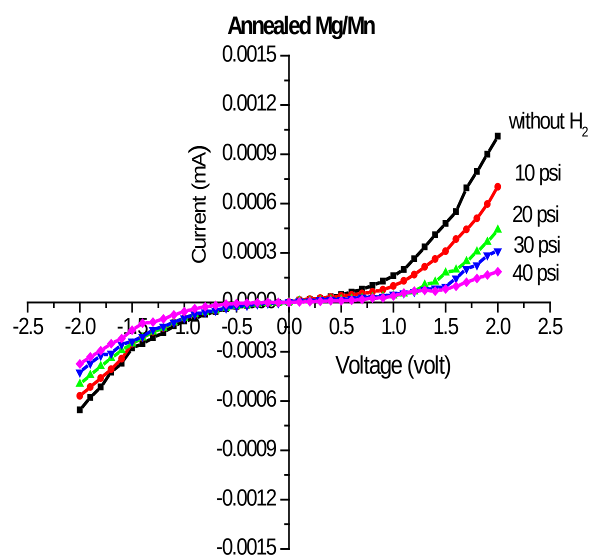

Figure 3 : I-V characteristics of annealed bilayer Mg/Mn thin films

Fig. 4 & 5 shows the variation in intensity versus wave number of Raman spectroscopy. In these spectra intensity of Raman peaks is decreased for both Mg/Co and Mg/Mn thin films with hydrogen pressure and one predominant peak is observed at 47.70 cm^{-1} for Mg/Co & at 43.15 cm^{-1} for Mg/Mn that shows the clear evidence of presence of hydrogen in sampled at room temperature and also noted decrease in broadening of peaks. It suggests that hydrogenation may change the phase or make the bonding with metal interstitial as well as surface locations. In case of Si crystalline structure, hydrogen absorption peak were observed at 590 cm^{-1} by fukata et al [13]. Raman studies of rare earth hydrides under high pressure carried out by Kume et al

[14] according them optical band gap disappear at higher pressure in case of YH_3 and ScH_3 and suggested that there was a common mechanism for the structural transformation from hexagonal to the intermediate phase. In our case we also relate the decrease in intensity of Raman peak with phase transformation and confirmation of presence of hydrogen in thin films.

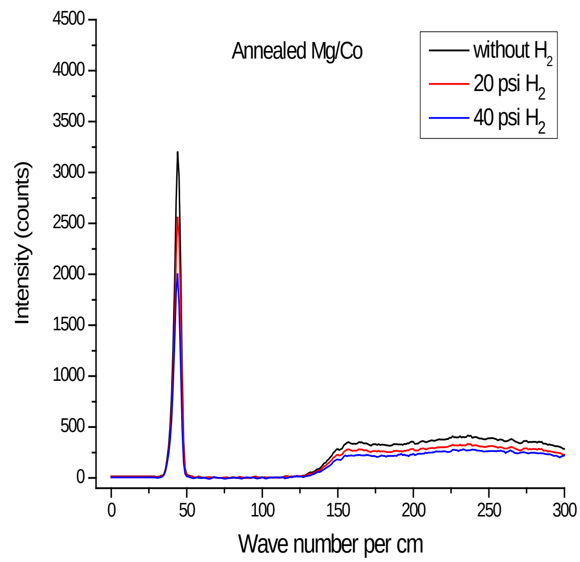

Figure 4 : Raman spectra of bilayer Mg/Co thin films

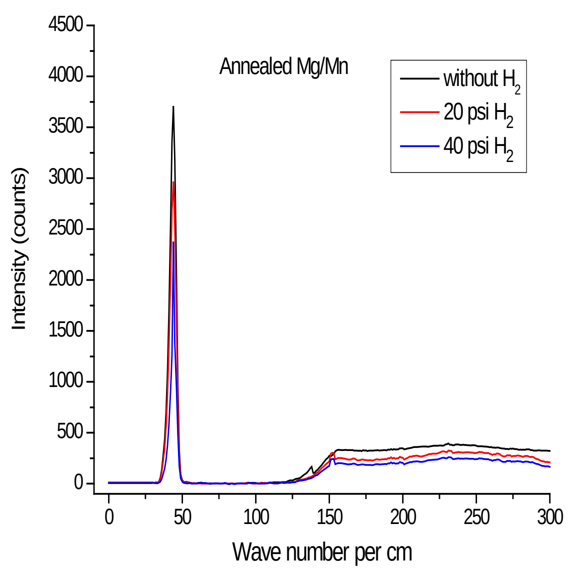

Figure 5 : Raman spectra of bilayer Mg/Mn thin films

IV. Conclusions

It is concluded from the above study that I-V characteristics of as grown Mg/Co and Mg/Mn thin films have been found to be ohmic and the I-V characteristics of annealed samples shows semiconductor behavior and conductivity has been found to be decreased with increasing pressure of hydrogenation. Decrease in intensity of Raman peak may be due to phase transformation.

V. Acknowledgements

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