May 2003 • NREL/CP-520-33596

Conference Paper

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National Renewable Energy Laboratory

Presented at the National Center for Photovoltaics and Solar Program Review Meeting
Denver, Colorado
March 24-26, 2003

National Renewable Energy Laboratory

1617 Cole Boulevard
Golden, Colorado 80401-3393

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Contract No. DE-AC36-99-GO10337

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ABSTRACT

We report on the pulsed laser deposition (PLD) of Mo-doped indium oxide (IMO) films with mobilities of up to 125 cm^2/Vsec. Films have been grown from targets with 1-4 wt. % molybdenum. The optimum electrical and optical properties were obtained with the 2% target and yielded a maximum conductivity of 3717 S/cm with mobilities of 99 cm^2/V-sec on (100) yttria stabilized zirconia (YSZ) single crystal substrates. Films also exhibit greater than 90% transparency in the visible range. Compared to commercial indium tin oxide (ITO) films, these PLD-grown IMO films have similar conductivity but since they have substantially higher mobility they have a correspondingly lower carrier concentration. The lower carrier concentration should extend the infrared window of the transparency for films of the same conductivity. This may lead to improved performance in a number of applications requiring improved performance TCOs.

1. Introduction

Transparent conducting oxides (TCOs) are an important component in a number of technologies. These include thin-film photovoltaics, electrochromic windows, and a flat-panel displays [1]. Tin-doped indium oxide (ITO) is one of the most important TCOs in industry [2]. However, the search for improved performance through new dopants is of increasing interest [3].

Improved conductivity can be obtained by increasing the mobility or the carrier concentration. However, an increase in carrier concentration will decrease the transparency due to the free carrier adsorption. On the contrary, increasing the mobility can improve electrical conductivity without sacrificing optical transparency. Thus, high mobility materials would enhance overall TCO properties [3].

Recently, it was reported that molybdenum-doped indium oxide (IMO) thin films, as prepared by thermal reactive evaporation, exhibited exceptional electrical and optical properties [4,5]. A low resistivity of 1.7 x 10^{-4} ohm-cm, high mobility over 100 cm^2 V^{-1} s^{-1} and greater than 80% transparency were obtained [4,5].

In this work, we present a study employing the pulsed laser deposition of IMO films with different Mo target concentrations. The work replicated and improved on the initial results with targets that were 2 wt % Mo [4,5].

2. Experiment

PLD films were prepared from 1-inch pure indium oxide and molybdenum doped indium oxide (1, 2 and 4wt.% Mo) targets obtained from Cerac, Inc. A KrF eximer laser (Lambda Physik EMG 103, λ = 248 nm) was used for target ablation.. A laser power of 300 mJ per pulse and a pulse rate of 10 Hz was used during deposition. Films were deposited on glass and single crystal YSZ (100) substrates. The chamber base pressure was 10^{-7} Torr prior to deposition. The chamber was backfilled with pure oxygen to the total pressure of 2 millitorr prior to deposition. Films were grown at a substrate temperature of 500 °C. Annealing experiments were performed in a tube furnace at temperature of 400 °C for 12 hours in reducing atmosphere (4% hydrogen in argon).

The crystal structure of the films was analyzed with x-ray diffraction (XRD) using Cu $K\alpha$ radiation (Scintag DMS2000). The film resistivity, mobility, and carrier concentration were obtained from Hall measurements at room temperature by using the Van der Pauw method. The Hall measurements were performed on the BioRad HL5500 System. The film thickness was measured by using stylus profilometry (Veeco Dektek 3). The optical measurements were performed on the Nicolet FTIR spectrometer. The transmission and reflection are measured at normal and 30° incidence respectively. Atomic force microscopy (AFM) was used to determine grain sizes.

3. Results and discussion

The $\theta/2\theta$ XRD spectra of pure IO film on glass and 2% Mo-doped IO films on both glass and YSZ single crystal substrates are shown in Figure 1. All the XRD peaks are indexed to In_2O_3. No other compounds were observed when Mo was added as a dopant. From Figure 1(a), the XRD data reveal that the pure IO (i) and 2% Mo-doped IO (ii) on glass substrates are randomly oriented crystalline films. The 2%

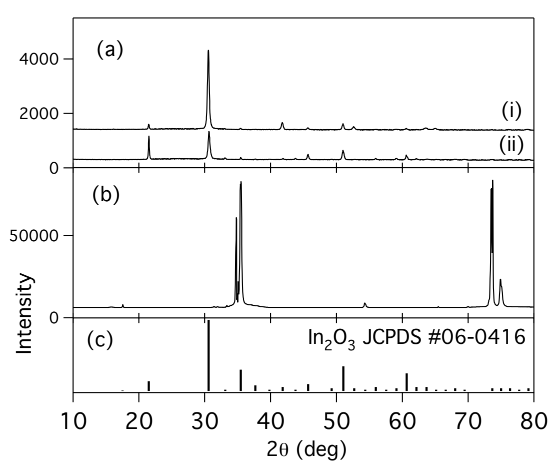

Figure 1. XRD spectra of pure IO and 2% Mo-doped IO films on glass and single crystal YSZ.

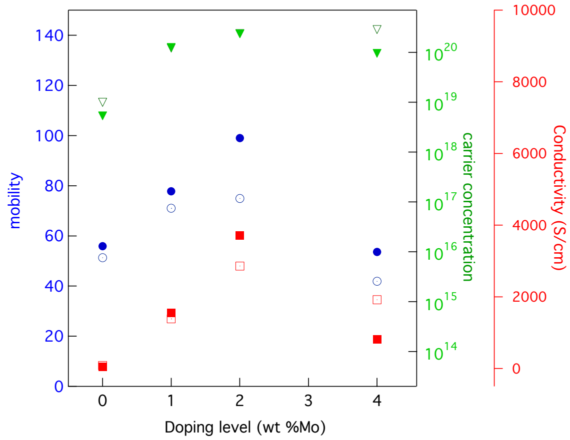

Figure 2. Electrical properties of pure IO and Mo-doped IO films.

Mo-doped film on YSZ has (200) preferred orientation, as shown in panel (b).

Hall measurement results performed on the IO and Mo-doped IO films with various doping levels are shown in Figure 2. Closed and open symbols represent films on YSZ and films on glass, respectively. The typical film thickness is in the range of 5000-7000Å. The results show that both pure and Mo-doped indium oxide thin films are n-type. Mo-doped IO films exhibit higher conductivity, carrier concentration, and Hall mobility than pure IO films. The pure IO film on YSZ has a carrier concentration of 5.3 x 10^{18} cm^{-3}. By adding 1% Mo dopant, carrier concentration has increased substantially to 1.3 x 10^{20} cm^{-3}. This suggests that Mo atoms substitute In atoms and donate extra electrons to the charge carriers. The carrier concentrations change slightly when more Mo dopants were added. The electrical properties (conductivity, carrier concentration, and mobility) are optimized when 2% Mo was added. The maximum conductivity of 3717 S/cm, carrier concentration of 2.35 x 10^{20} cm^{-3} and mobilities of 99 cm^2/Vsec were obtained from film on YSZ. Note that this film achieved a very high mobility of 125 cm^2/ V-sec on selected area. When doping level increases from 2 to 4%, mobility decreases

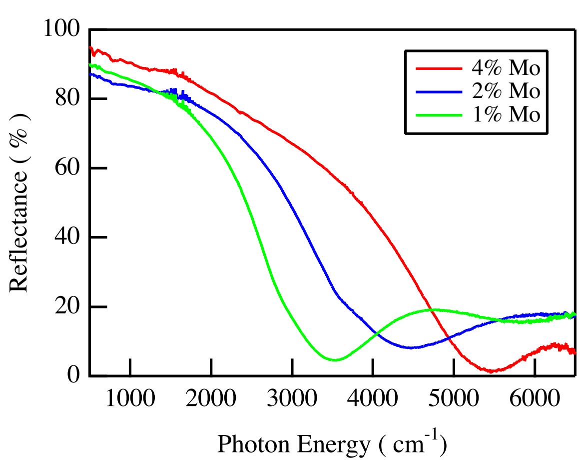

Figure 3. Reflectivity curves of Mo-doped IO films on YSZ substrates.

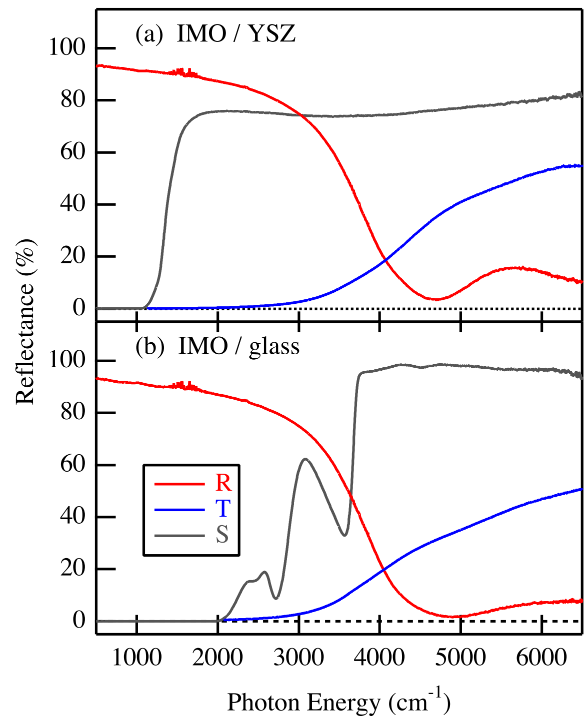

Figure 4. Optical properties of 2 % Mo-doped IO films on YSZ (a) and glass (b) substrates after annealing. R,:reflecton, T: transmission, S: substrate transmission.

substantially while the change in carrier concentration is insignificant. This suggests that Mo dopants may not be readily ionized and some may act as neutral impurities in the films.

Atomic force microscopy (AFM) studies show that the grain size of 2% Mo doped films is in the range of 70 nm. Even though, films on YSZ single crystal substrates have slightly higher mobility than films on glass, the role of grain boundaries is still inconclusive from our data.

Figure 3 shows reflection curves of Mo-doped IO films on YSZ. The reflection-transmission crossovers shift to higher wavenumber when doping level increases. This suggests that the carrier concentration increases monotonically as the Mo content increases. Note, however, that the results from the Hall measurements indicate that carrier concentration decreases upon increasing the Mo content from 2 to 4 wt.%. Figure 4 shows the reflection and transmission curves (normalized to the substrates) of the 2% Mo-doped IO films after annealing on YSZ (a) and on glass (b). Both films show similar reflection and transmission curves. This result is also consistent with the data from Hall measurements which show that both films have similar carrier concentrations.

4. Summary

We have grown IMO films by PLD from Mo containing targets from 0-4 wt% Mo. Best results were obtained for the 2 wt% target and replicate or exceed previously published work on this system. Mobilities to 125 cm^2/V-sec were observed. This system offers the potential for improved performance over current TCOs in transparency and conductivity.

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REPORT DOCUMENTATION PAGE

Form Approved
OMB NO. 0704-0188

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	May 2003	Conference Paper

4. TITLE AND SUBTITLE
Highly Conductive Textured Molybdenum Doped Indium Oxide Thin Films

5. FUNDING NUMBERS
PVP3.4701

6. AUTHOR(S)
C. Warmsingh[1], Y. Yoshida[1], D. Readey[1], J. Perkins, P. Parilla, C. Teplin, T. Kaydanova, J. Alleman, L. Gedvilas, B. Keyes, T. Gessert, T. Coutts, and D. Ginley

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National Renewable Energy Laboratory, 1617 Cole Blvd., Golden, CO 80401-3393

8. PERFORMING ORGANIZATION REPORT NUMBER
NREL/CP-520-33596

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
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U.S. Department of Commerce
5285 Port Royal Road
Springfield, VA 22161

12b. DISTRIBUTION CODE

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14. SUBJECT TERMS
IMO; pulsed laser deposition; solar cells; TCO; thin film

15. NUMBER OF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	UL