# Development of High Performance Indium Tin Oxide Films at Room Temperature by Plasma-Damage Free Neutral Beam Sputtering System

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#### Abstract

New ITO thin film of good performance has been developed by brand-new, plasma-damage-free sputtering process at the room temperature. The room temperature-processed ITO films with optimized conditions as neutral beam acceleration bias of -30V and In & Sn composition ratio of 99:01 gives lower resistivity as  $4.22 \times 10^{-4} \Omega$ -cm and higher transmittance over 90% a wavelength of 550 nm. The transmission electron microscope (TEM) images of the films show a nano-crystalline structure.

#### 1. Introduction

There have been many technological advances and issues with low temperature thin film transistor process for plastic based flexible displays. Until now many application done by plasma and heat treatment processes, but these might be harmful and limited to the organic material based device such as OLED, OTFT, and various flexible display, in terms of charge and thermal damages [1-4].

Recently, organic TFT array and its LCD module are to be combined with several technical breakthroughs including new organic gate dielectric materials, improvement of ohmic contacts, and well-optimized process-architectures. Base on the newly developed organic gate dielectric with pentacene as an organic semiconductor, the world-record field effect TFT mobility in excess of  $7 \, \mathrm{cm}^2/\mathrm{Vs}$  and excellent on/off ratios as  $\sim 10^6$  have been presented by Samsung Electronics [5]. For jumping up the OTFT research activities from laboratory scale to real production line,

there need key technologies. One of them is new ohmic contact technology with very common materials in the current LCD manufacturing such as ITO; requirement of the bottom contact structured OTFT with ITO source-drain electrode is much the same performances as that with gold source-drain electrode without thermal and plasma damages during the TFT fabrication processes.

Organic light emitting diode (OLED) had experienced much development in display industry. Recently, mass production of OLED requires simpler, cheaper, and larger area applicable processes. The deposition process for cathode electrode is one of the problematic processes in mass production. Most of production is conducted by thermal evaporation or ebeam evaporation. But these processes have some trouble in large area applications. In LCD applications, the metal is deposited by magnetron sputtering which is well developed for several decades. But sputtering process could not be applied to OLED, because sputtering causes some critical damage to underlying organic layers. The origin of damage is supposed to be caused by high energy ions in plasma process.

In this study, we have developed new ITO films with good optoelectronic properties at room temperature by using neutral beam based sputtering technique for plasma-damage-free deposition process, named as Hyper-thermal Neutral Beam (HNB) Technology [6]. During the sputtering deposition, instead of heating the substrate to supply reaction energies to the reactive atoms on the substrate, we produce reactive atomic beams which are already

accelerated enough for the reaction energy in the neutral beam source earlier before they reach the substrate.

#### 2. Experimental Procedure

ITO films were deposited on glass at the room temperature by the Hyper thermal Neutral Beam (HNB) based sputter. Our developed neutral beam source consists of an inductively coupled plasma (ICP) source, a magnetron sputter source, reflector, and limiter as shown in Fig 1 as described in our other paper [6]. The magnetron sputter source supplies solid elements such as indium and tin atoms into the plasma in which the solid elements are ionized. The ions are accelerated in the plasma sheath between the plasma and reflector, and then neutralized mainly through the Auger neutralization [7]. The neutralization efficiency depends on the impinging angle, reflector material, surface roughness, etc [8]. We use a polished stainless steel (SUS316L) plate for the reflector. The neutral beam energies are a half of the impinging ion energies [9]. The accelerating potential is the sum of the plasma potential and biased voltage. For instance, in case of a bias voltage of 30 eV with a plasma potential of 10 eV, the impinging ions are accelerated up to 40 eV and then the reflected neutral atoms keep a half of the energy, i.e., about 20 eV.

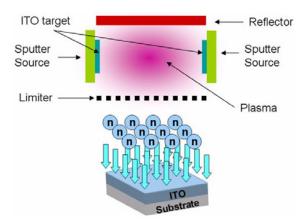


Fig 1. Diagram of the neutral beam source

The HNB source chamber is evacuated down to pressure  $2 \times 10^{-6}$ Torr and Ar partial pressure of 1mTorr were introduced during deposition. The RF biases are  $500 \sim 1200$ W, DC biases for magnetron sputter are  $100 \sim 250$ W for sputter ITO target and reflector biases are  $0 \sim 120$ V. We have designed experimental

conditions to survey effect of

- A. HNB energy dependence: DC bias forwards to reflector from 0V to -120V to see how HNB influences deposition process.
- B. ITO composition dependence: InSn alloy target varies with following ratio of Sn/In as 0, 1/99, 10/90, 15/85 and oxygen partial pressure was controlled to keep minimum resistivity for each conditions.
- C. Ar HNB post-treatment as Time dependence: post-treatment by Ar HNB would work same as post thermal-annealing effect. But the effective thickness would be limited by thickness of ITO, because HNB can influence only few monolayers of ITO film; ITO thickness can be controlled by sputter on-off time modulating. While turning on sputter power, ITO thin films are deposited on glass. And during turning off sputter power, Ar HNB incident to the deposited ITO thin film in order to post treatment. Total ITO thickness is controlled by total number of sputter on-off cycle. The Ar HNB gives its energy to transform the phase of ITO thin film.

The film Thicknesses were measured with a long scan profiler (Tencor P-2). The sheet resistance of ITO film were measured by 4-point probe system with current power supply (Keithley 236) and voltage meter (Agilent 34401A) and Hall Measurement. The Transparencies were measured by scanning spectrophotometer (UV-2101PC). The microstructures of the film were analyzed by field emission transmission electron microscope (FE-TEM).

#### 3. Results and Discussion

#### A. Effect of HNB energy

Fig 2 shows that the carrier types and density of ITO films vary with HNB energy which can be controlled by reflector bias [6]. The resistivities of these samples are same as reverse of Fig 2. It would be the doping effect of additional impurities from reflector; at high reflector bias condition some of metallic components would be sputtered out from reflector.

If carrier type change cause by mainly impurities, the Fermi edge must be change. But the work function of these ITO films do not change (4.45~4.57eV, measured by UPS). So we would rather consider other reasons of change of their carrier type; HNB energy would increase carrier density of ITO thin film, then the conduction band (In:5s band) becomes valence

band. Or increase of HNB energy would enforce transforming abnormal phase of indium oxide and tin oxide. Because of the two oxide's heat of formation energies are different. [10]

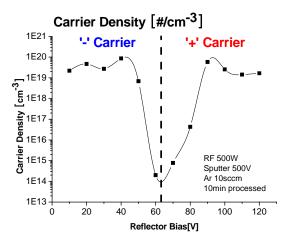


Fig 2. n-type to p-type transition

#### B. Effect of ITO composition

Fig 3 shows that ITO resistivity strongly depends on the ITO composition and HNB energy. The lowest resistivity can be achieved  $6.75e^{-4}$  [ $\Omega$  cm] by the optimized deposition conditions; reflector bias is -35V and the composition ratio of In : Sn is 99 : 1. Transmittance of HNB ITO is little bit lower than that of conventional poly ITO, but their difference is not more than 5% as shown Fig 4. The HNB energy would enhance differently forming ITO as each species.

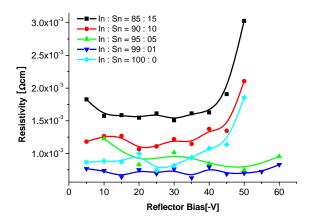


Fig 3 Resistivity as various In:Sn composition

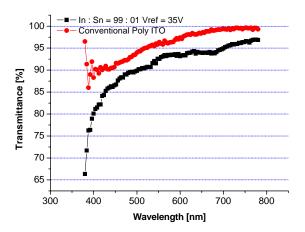


Fig 4. Transmittance of In: Sn = 99:01 and Conventional Poly ITO

Fig 5 shows TEM images and diffraction patterns of ITO film deposited by HNB. For the HNB processed ITO, the TEM image and diffraction patterns represent not poly-crystalline, but nanocrystalline structure with very small grain. This structure might increase conductivity cause by increase of mobility.

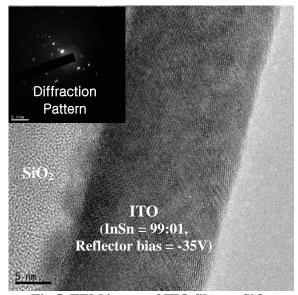


Fig 5. TEM image of ITO film on SiO<sub>2</sub>

### C. Effect of Ar HNB post treatment as Time variation

Fig 6 shows the resistivity of ITO thin film varies with Ar HNB treatment time. The lowest resistivity can be achieved  $4.22e^{-4}$  [ $\Omega$  cm] by the optimized deposition conditions; reflector bias is -30V, the

composition ratio of In: Sn is 99:1 and sputter on-off time ratio of 60-120. Resistivity strongly depends on the time of HNB post treatment during sputter turn off time. The Ar HNB can deliver kinetic energies into the ITO film and enhance crystalline and tin doping concentration in indium oxide. Other experiments for Ar HNB post treatment, increase sputter on time, there are less effect of Ar HNB treatment. That is Ar HNB influence less than 10~20Å of ITO film.

## Sputter On-Off time controll

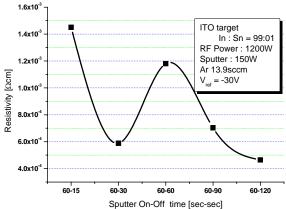


Fig 6. Dependence of Ar HNB treatment time

#### 4. Summary

In this study, we have studied the difference between the ITO films deposited by various conditions of HNB system. We deposited ITO films on glass substrate with various HNB energies, compositions of ITO and Ar HNB post treatment time.

The currently lowest resistivity of ITO films is of  $4.22 \times 10^{-4} [\Omega\text{-cm}]$  with followed optimized process conditions; reflector bias of 30V and target composition ratio for In/Sn of 99/01 and sputter onoff time ratio of 60-120. In this condition, the transmittance is over 90% above visible light region. The TEM images and diffraction pattern show the ITO deposited by HNB sputter might nanocrystalline structure. The hyper thermal energy particle enhances forming the nanocrystalline structure; longer HNB post treatment time drives get lower resistivity. HNB processes can deliver high quality ITO film at the room temperature.

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