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Effect of barrier layers on the properties of indium tin oxide thin films on soda lime glass substrates

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Abstract

In this paper, the electrical, structural and optical properties of indium tin oxide (ITO) films deposited on soda lime glass (SLG) haven been investigated, along with high strain point glass (HSPG) substrate, through radio frequency magnetron sputtering using a ceramic target (In_2O_3:SnO_2, 90:10 wt.%). The ITO films deposited on the SLG show a high electrical resistivity and structural defects compared with those deposited on HSPG due to the Na ions from the SLG diffusing to the ITO film by annealing. However, these properties can be improved by intercalating a barrier layer of SiO_2 or Al_2O_3 between the ITO film and the SLG substrate. SIMS analysis has confirmed that the barrier layer inhibits the Na ion's diffusion from the SLG. In particular, the ITO films deposited on the Al_2O_3 barrier layer, show better properties than those deposited on the SiO_2 barrier layer.

Keywords: Indium tin oxide (ITO); Soda lime glass (SLG); RF-magnetron sputtering; Barrier layer; Diffusion

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ABSTRACT

In this paper, the electrical, structural and optical properties of indium tin oxide (ITO) films deposited on soda lime glass (SLG) haven been investigated, along with high strain point glass (HSPG) substrate, through radio frequency magnetron sputtering using a ceramic target (In_2O_3:SnO_2, 90:10 wt.%). The ITO films deposited on the SLG show a high electrical resistivity and structural defects compared with those deposited on HSPG due to the Na ions from the SLG diffusing to the ITO film by annealing. However, these properties can be improved by intercalating a barrier layer of SiO_2 or Al_2O_3 between the ITO film and the SLG substrate. SIMS analysis has confirmed that the barrier layer inhibits the Na ion's diffusion from the SLG. In particular, the ITO films deposited on the Al_2O_3 barrier layer, show better properties than those deposited on the SiO_2 barrier layer.

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1. Introduction

Indium tin oxide (ITO) thin films, most widely used as transparent conducting oxides (TCOs), have been widely used as optoelectronic devices such as flat panel displays (FPDs), touch panels and solar cells because of their superior conductivity and high transparency [1–4].

Most properties of ITO films depend on the nature of their substrate, the deposition techniques used and the composition of the ITO film. For application in display panels, these films are normally deposited on a substrate having a high strain point (>575°) or borosilicate glass containing a few Na ions. These films must be deposited at a temperature higher than 250 °C and then annealed at a temperature higher than 300 °C in order to have high optical transmittance in the visible region, low resistivity and chemical duration [5,6]. However, the high strain point glass (HSPG) used in FPDs actually blocks the widespread use of large sized FPDs because it is more expensive than commercial soda lime glass (SLG).

If SLG could be used as a substrate for FPDs, then the diffusion of the Na ions from the substrate occurs into the ITO films during annealing or heat treatment during the manufacturing process. This diffusion affects the properties of thin films and so proper care needs to be taken to minimize Na ion diffusion. Several researchers have used SiO_2, SiN_x, TiO_2 and SiO_2–TiO_2 coatings on films as barrier layers [7–10]. However, to date, optimum thickness control, the diffusion coefficient of Na ions into the ITO and barrier films, and lattice stress from the coefficient of thermal expansion (CTE) mismatch between the barrier layer and the SLG have not been considered.

In this study, the electrical, structural and optical properties of ITO films deposited on a SLG, barrier layer (SiO_2 or Al_2O_3) have been investigated, along with a HSPG substrate after annealing. Also, the influence of the barrier layer thickness is discussed and the SiO_2 is

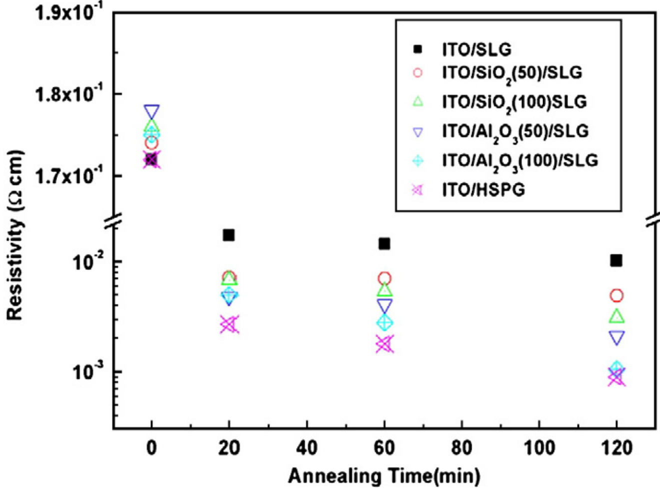

Fig. 1. The resistivity of ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG, and ITO/HSPG films for as-deposited, 20 min, 60 min and 120 min in N_2 atmosphere at 400 °C.

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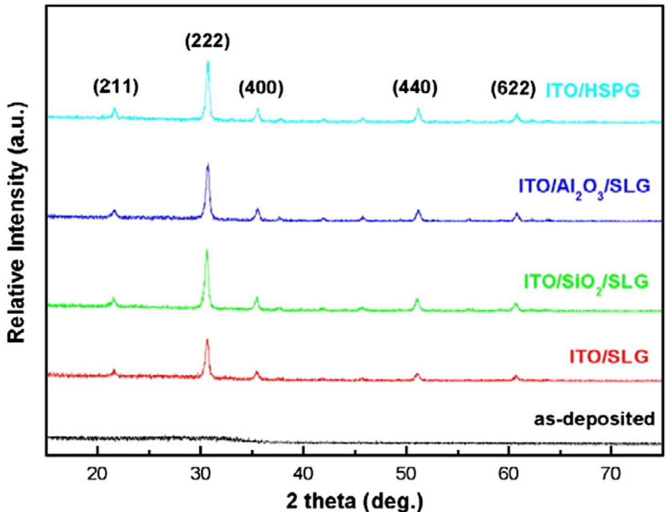

Fig. 2. The XRD patterns of the as-deposited ITO film and ITO/SLG, ITO/SiO_2(100)/SLG, ITO/Al_2O_3(100)/SLG and ITO/HSPG films after annealing for 120 min in a N_2 atmosphere at 400 °C.

compared with the Al_2O_3 as possible barrier layers. The results of this study provide evidence that the barrier layer greatly affects the properties of the ITO films.

2. Experimental details

Commercially available SLG and HSPG (PD200, Ashai glass corp.) containing a few Na ions substrate with a surface area of 20×20 mm^2 were used in this work. The glass substrates were cleaned with an ultrasonic cleaner using acetone, methanol and deionized (DI) water. The barrier layer (SiO_2, Al_2O_3) was deposited with a thickness of 50 nm and 100 nm on the SLG substrate by radio frequency (13.56 MHz) magnetron sputtering in Ar/O_2 with a flow rate of 30 sccm / 5 sccm using SiO_2 and Al_2O_3 targets (LTS Inc.) with a purity of 99.99%. The ITO films were sputtered-deposited on each of the substrates(SLG, barrier layers/SLG, HSPG) using an ITO target (LTS Inc.) with 99.99% purity In_2O_3:SnO_2 (90 wt.% and 10 wt.%, respectively). The sputtering was carried out at Ar flow rates of 20 sccm, a working pressure of 15 mTorr and an RF power of 120 W. After the deposition, annealing of the samples was carried out in N_2 atmosphere, at 400 °C, for 120 min.

The film thickness was measured using an α-step (Tencor 500) and the structural properties of the films were determined using X-ray diffraction measurements (Rigako D/MAX2200) with Cu-Kα radiation. The electrical resistivity of the films was measured by a four-point probe method (Keithley 2420, 2182A). Their optical transmittance was measured in a wavelength range from 200 to 800 nm using an UV–VIS spectrometer (Simazu 2401). The surface morphologies were analyzed using a Field Emission Scanning Electron Microscope (JEOL JSM-6700F). The suppression effect of the barrier layer on the diffusion of the Na ions from the SLG was investigated using secondary-ion mass spectrometry (CAMECA IMS-6f Magnetic Sector).

3. Results and discussion

Fig. 1 shows the resistivity of ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG and ITO/HSPG films for as-deposited, 20 min, 60 min and 120 min in a N_2 atmosphere at 400 °C. When the annealing time is above 20 min., the resistivity of the ITO films sharply decreases and the lowest value obtained after the annealing is found to occur for an annealing time of 120 min. The resistivity of all the samples decreased with increasing annealing time, indicating that the resistivity of the annealed ITO films in the crystalline phase is lower than that of the as-deposited amorphous phase. This result is confirmed by the XRD patterns seen

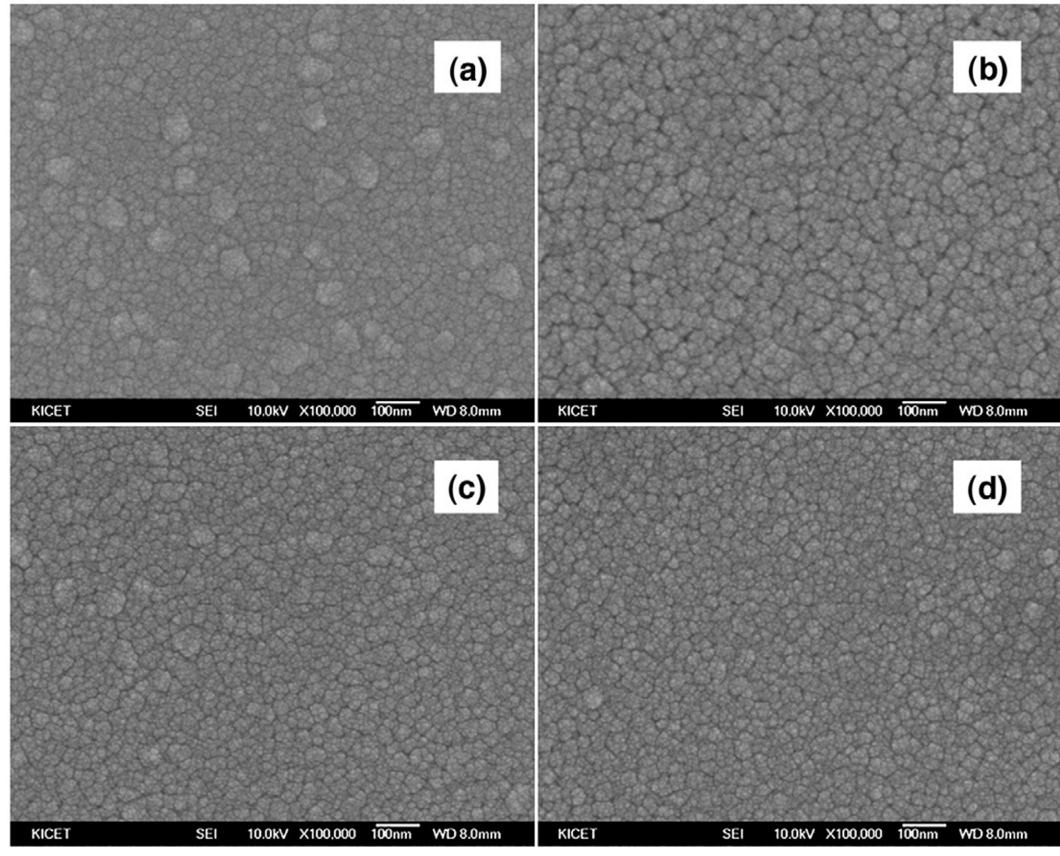

Fig. 3. SEM micrographs of the top surface of (a) ITO/SLG, (b) ITO/SiO_2(100)/SLG, (c) ITO/Al_2O_3(100)/SLG and (d) ITO/HSPG films.

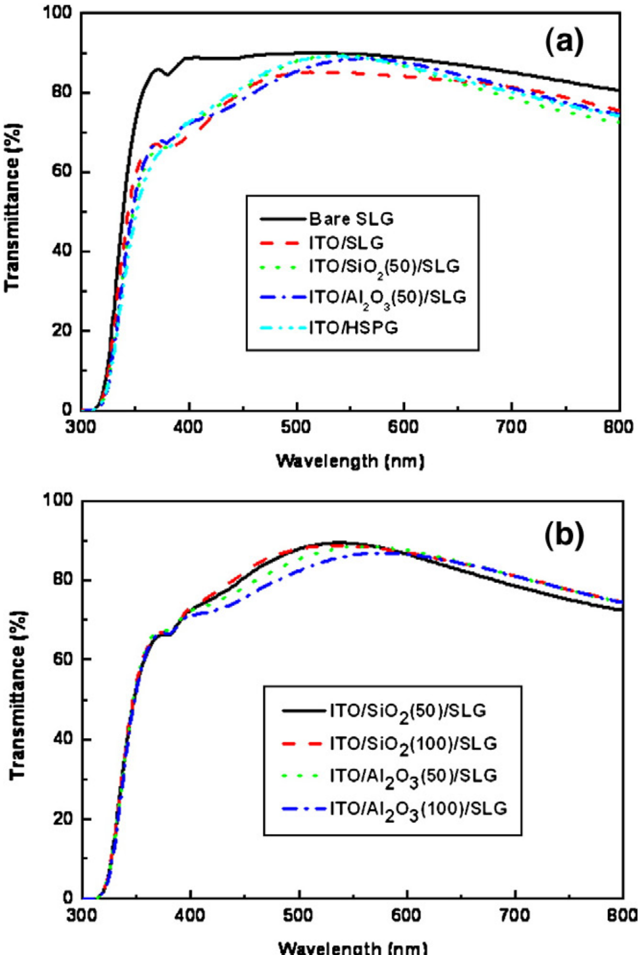

Fig. 4. (a) Transmission spectra of ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG and ITO/HSPG films and (b) Transmission spectra of films according to barrier layer thickness.

in Fig. 2. The ITO/HSPG film has a lowest resistivity of 9.01×10^{-4} Ω cm. The resistivity of the ITO/SLG film has the highest resistivity of the six samples investigated. This poor resistance to thermal annealing is related to the diffusion of the Na ion impurity from the SLG to the ITO films. The resistivity of the ITO is greatly decreased by introducing a barrier layer of SiO_2 or Al_2O_3 between the ITO and SLG. The resistivity (10.2×10^{-4} Ω cm) of the ITO/Al_2O_3(100)/SLG film is most similar to that of the ITO/HSPG film and is assumed that the diffusion of the Na ion from the SLG to the ITO films is suppressed by the intercalation of the barrier layer of SiO_2 or Al_2O_3 films. Also, the effect of using Al_2O_3 with a barrier thickness of 100 nm is excellent.

Fig. 2 shows the XRD patterns of the ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG and ITO/HSPG films before and after annealing for 120 min. at 400 °C in N_2 atmosphere. It can be seen that the ITO films, deposited without annealing, are in the amorphous phase. Crystallization occurs after annealing at a temperature above 400 °C. Most of the annealed ITO films have a cubic bixbyte structure with diffraction peaks corresponding to (211), (222), (400), (440) and (622) orientations. The (222) plane has the most prominent peak of all the investigated samples. This result is also consistent with the work presented by Raoufi [11] and Salehi et al. [12] who obtained the same (222) predominant peak. The amorphous films have many defects in structure and nonstoichiometry in composition. Post annealing can oxidize a nonstoichiometry composition, such as In_2O_{3-x} and SnO_{2-x}, and rearrange the atoms to form stable polycrystalline films [13]. The average crystallite size is calculated from the (222) peak seen in Fig. 2 using the Scherrer formula given by $s = 0.9\lambda / \beta\cos(\theta)$, where θ is the Bragg diffraction angle in degrees, λ is the wavelength of the X-rays (Cu-Kα radiation) and β is the full width at half-maximum (FWHM) in radian [14]. It can be seen that the average grain size is 244 Å for the ITO/SLG film and about 275 Å for the ITO/SiO_2/SLG, ITO/Al_2O_3/SLG and ITO/HSPG films. This result shows that the diffusion of the Na ion impurity from SLG results in poor crystallinity due to the formation of a secondary phase observable in the SEM image of Fig. 3(a). such as Na_2SnO_3 or $Na_2Sn_2O_5$ that suppress normal grain growth and can promote secondary grain growth [15,16].

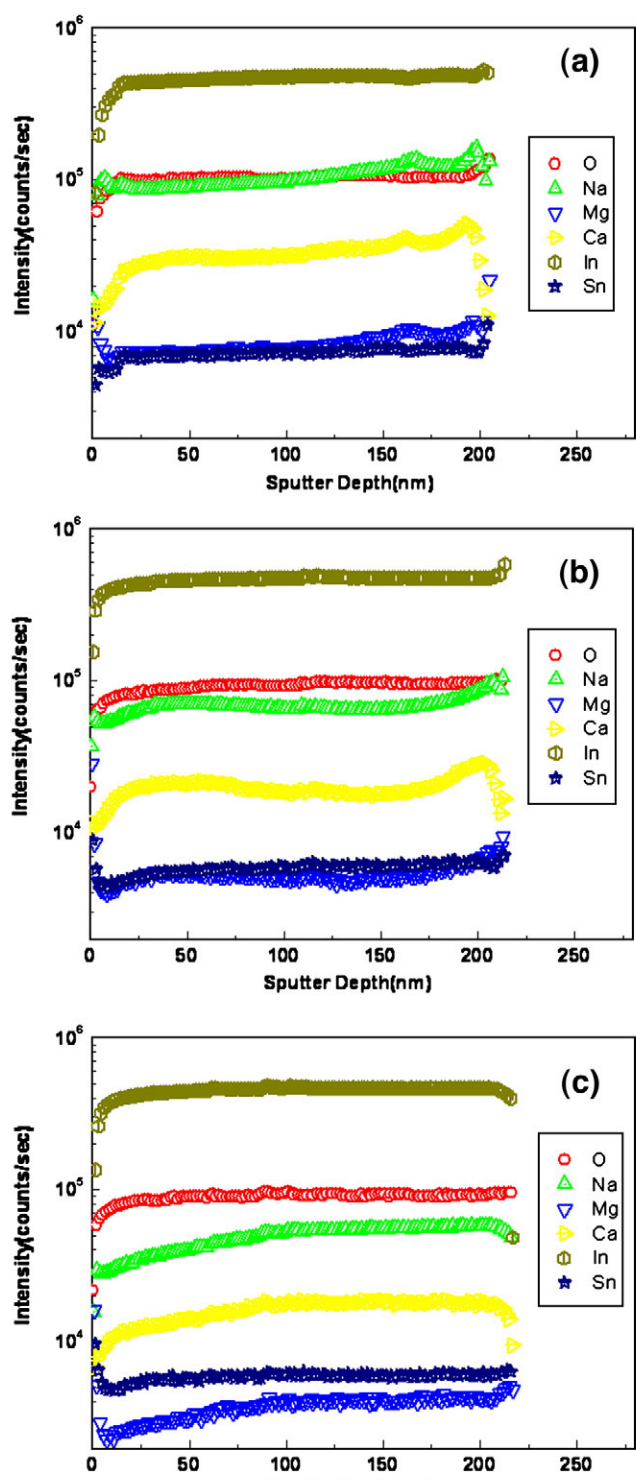

Fig. 5. SIMS concentration profiles as a function of the ITO thickness without (a) and with the intercalation of (b) SiO_2(100) and (c) Al_2O_3(100) barrier layers after annealing at 400 °C.

The grain size of the affected ITO films was increased by the intercalation of a barrier layer and the use of the HSPG substrate. The resistivity of polycrystalline ITO films is described with grain-boundary scattering. The increase in the grain size brought about a decrease in the grain boundary scattering and grain boundary potential [17]. Therefore the resistivity of ITO films with large grain size was reduced.

The SEM micrographs of annealed ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG and ITO/HSPG films are shown Fig. 3 from which it is clear that all films have a polycrystalline structure. A uniform grain distribution is observed in Fig. 3(b–d) and from Fig. 3(a), the formation of a heterogeneous phase by the Na ion diffusion from the SLG substrate can also be seen. The average grain size along the sample surface is about 260 Å, which is also consistent with the grain size estimated by the Scherrer formula. The column boundaries can be seen to widen and crack in the ITO/SiO_2/SLG (Fig. 3(b)) films. This also explains the high thermal tensile stress obtained by the CTE mismatch between the SiO_2 (14×10^{-7}/°C) and the SLG(87×10^{-7}/°C) for annealing. However the Al_2O_3 (84×10^{-7}/°C) on SLG has a little thermal stress since they have a similar CTE.

The transmittance of the ITO films is also important with regard to their application in FPDs. Fig. 4(a) shows the optical transmittance spectra of ITO/SLG, ITO/SiO_2/SLG, ITO/Al_2O_3/SLG, ITO/HSPG films and bare SLG substrates in the wavelength range from 300 to 800 nm. The average transmittance T of the SLG in the visible light range of the spectra ($\lambda=380$ to 780), using air as a reference, is 89%. The transmittance of the ITO films deposited on the SLG and HSPG is 84% and 86%, respectively. In particular, the transmittance of polycrystalline ITO films is affected by grain boundary. When the surface grain size increases, light is less scattered by the decreasing grain boundaries [13], therefore the ITO/HSPG film has high transmittance in the visible light range. Also, flat surface of ITO films reduce the scatter of the light and increase transmittance. The transmittance spectra of the ITO/SiO_2/SLG and ITO/Al_2O_3/SLG films show almost the same transparency when compared with those of ITO/HSPG films. It can be seen that there is no actual decrease of the average transmittance by intercalating a barrier layer between the ITO films and the SLG.

The transmittance spectra of the ITO films deposited on a barrier layer of two different thicknesses (50, 100 nm) are shown in Fig. 4(b). By ignoring the absorption of the films, the transmittance of the multi layer thin films can be estimated: $T=(1-R_1)(1-R_2)(1-R_3)\ldots$, where R is a reflectance that is represented by the refractive index of each of the materials. The SiO_2 film of 50 nm thickness, being a lower refractive index material than that of the Al_2O_3 film and having the desired physical thickness, is possibly the reason for the high visible transmittance of the four types.

Fig. 5 illustrates the concentration profiles of several elements including Na as a function of the depth of the ITO films deposited on the SLG, on the SiO_2(100)/SLG and on the Al_2O_3(100)/SLG substrate annealed at 400 °C. Due to the uncontrollable charging problem, the depth profiles, within the barrier layer and the SLG substrate, are absent. It is observed that the constituent elements In, Sn and O are uniformly distributed throughout the ITO films. For the ITO/SLG, the concentration profile confirms the Na diffusion from the SLG substrate during the crystallization treatment and the contamination is effective through almost the entire thickness of the ITO films. Moreover, the analyses also reveal the presence of other alkaline earth traces such as magnesium and calcium. In contrast, the ITO films deposited on SiO_2/SLG and Al_2O_3/SLG substrates demonstrated the effectiveness of the barrier layers against diffusion of the Na ions. This result also confirms that the diffusion of the Na ions and other impurities through the Al_2O_3 barrier layer into the ITO film is much lower than through the SiO_2 barrier layer. The thicker barrier layers are more efficient in hindering the diffusion of the Na ions but amorphous Al_2O_3 with 100 nm thickness having a lower diffusion coefficient for the Na ion than for SiO_2 [18,19] is most useful.

4. Conclusions

The electrical, structural and optical properties of the ITO films deposited on a SLG, barrier layer (SiO_2 or Al_2O_3) have been investigated using RF-magnetron sputtering. Through an annealing treatment at 400 °C for 120 min in a N_2 atmosphere, transparent, electrically conductive ITO films have been obtained. However, the ITO films, deposited on the SLG substrate, have high electrical resistivity and low crystallization because the alkali metal impurity from the SLG can diffuse to the ITO films through the annealing treatment. The properties of ITO/SLG film can be significantly improved by intercalating a barrier layer of SiO_2 or Al_2O_3 between the ITO layer and the SLG substrate. The Al_2O_3 (100 nm) barrier layer improves the resistivity and crystallization more effectively than using a SiO_2 (100 nm) barrier layer due to the superior suppression of the diffusion of impurities.

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