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REDUCED CONTACT RESISTANCE Ti/Al OHMIC CONTACTS TO VERTICALLY ALIGNED ZnO NANOROD

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Abstract

An ohmic contact is required for achieve long lifetime execution of optical and electrical devices. In this study, we are investigate on the structures and properties of titanium(Ti)/aluminium(Al) bi-layer contacts on undoped ZnO nanorod/AlN/Si(111). Firstly, high temperature grown buffer layer AlN (about 200nm) were grown on silicon (111) substrates by radio frequency nitrogen

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plasma assisted molecular beam epitaxy (PAMBE). Following by undoped ZnO nanorod layers were grown on AlN buffer layer using a hydrothermal approach. All the films were detected to have *c*-axis (0001) selected orientation adjusted with normal to the substrate surface. The structural and electrical stability of the contacts at thermal treatment (200ºC-500ºC) were studied, as low resistance metal semiconductor contacts are necessary for high quality electronic devices. Scanning electron microscopy (SEM) is applied to investigate the reformations in the surface morphology properties of the contacts on thermal annealing. The transmission line method (TLM) and current-voltage (I-V) measurements were used to determine the specific contact resistivity. The nonalloyed ohmic contact could be associated to the thermal annealing, which forms nitrogen vacancies that result in a heavily *n*-type surface whereby generate a tunneling junction. The nonalloyed Ti/Al contact on as-grown ZnO nanorods surfaces reveals non-ohmic behaviour. After alloying at 500ºC for 15 min, Ti/Al contacts ZnO nanorods surfaces have specific contact resistances around 0.062. Nonalloyed Ti/Al ohmic contacts would be extraordinarily helpful for fabricating high breakdown, recessed gate field effect transistors on ZnO since the moderate post annealing condition changes only the near surface layer to heavily *n*-type.

1. Introduction

Last few years, wide bandgap semiconductor materials, for example, ZnO, ZnSe, and GaN have captivated much concentration for the growth of short wavelength optoelectronics devices, for example, high efficiency low threshold blue light emitting diodes (LEDs) and ultraviolet-blue laser diodes [6, 13, 24]. ZnO takes a vital place in materials physics because of its interesting novel electrical and optical characteristics. In special, ZnO has some remarkable characteristics of the large bond vitality and the ultimate substantiality of excitons, contributing the sight of usable lasers with low threshold current density even at the temperature higher than room temperature [1].

Extraordinarily, undoped ZnO films are generally utilized as a translucent electrode, due to its lack of sophistication however also for its high mobility, an ultra short carrier lifetime, and a high transmittance in the long wavelength province distinguished to doped-ZnO films [24]. Seeing that the performances of devices perceptive rely on the quality of the ohmic contacts, the progress of reduce resistance contacts to the semiconductor device has captivated notable concentration [12, 25, 28, 31, 35]. Nevertheless, not much result for ohmic contact formation on ZnO was reported [3].

In semiconductor devices, if a high contact resistance happens between a metal and a semiconductor, the device achievement could be deteriorated by thermal stress, which might cause device failures. Multilayer metallization has been intriguing to researchers due to its ability of creating phase formation, which supports good ohmic contacts. Ti/Al bilayer metallization is an efficacious ohmic contact for wide bandgap semiconductors due to the synthesis of the compromising phase of $\mathrm{Al}_3\mathrm{Ti}$ [30]. Consequently, it is foreseen that a low-resistance ohmic contact can be executed applying Ti/Al bilayer metallization on ZnO. It was revealed that for the Ti-and Al-based ohmic contacts, interplay between metals and ZnO plays a vital part in shaped good ohmic behaviours [15, 16]. Nevertheless, this interfacial interaction consequence in the constitution of either Ti or Al-oxide possesses high sheet resistance, as a result heading to disagreeable thermal stability [22].

Lately, prominent advance has been formed in expanding ohmic contacts to *n*-type ZnO and to *p*-type ZnO. A large amount of metallization design, for example, Ti/Au, Al/Pt, Ti/Al/Pt/Au, and Al were studied as ohmic contact materials for *n*-type ZnO [10, 17, 20]. Newly, non-alloyed Al schemes were studied to reveal an ohmic property due to its taking part in the interfacial oxide constitution synthesize $\mathrm{Al}_2\mathrm{O}_3$, which promote the tunneling of carries [18, 20, 32].

For low resistance ohmic contacts on *n*-ZnO, besides Ta and Ru, Ti- and Al-based metallization schemes have been extensively studied [9, 23]. Such as, Ti/Au layers were frequently applied as *n*-type bilayer ohmic electrodes for devices, for example, *n*-ZnO/*p*-diamond heterojunction and ZnO homostructural *p*-*i*-*n* junction LEDs. To fabricate *n*-ZnO/*p*-doped (Zn, Mg)O junction and ZnO-based ultraviolet photodetectors, Ti/Al/Pt/Au layers and Al layers were applied as *n*-type ohmic contacts [11, 22]. Nevertheless, there is insufficiency of investigations on the interfacial effect between metal Al and ZnO.

ZnO occupy at the marginal between covalent and ionic semiconductors. Consequently, fabrication of ohmic contacts to ZnO with low contact resistivity can be accomplished by reducing the barrier height to increase thermionic emission (TE). In order to form an ohmic contact to *n*-type ZnO, the work function of the contact metal should be less than that of ZnO's electron affinity (4.2-4.35eV). Therefore, Al and Ti have been used for ohmic contacts to *n*-ZnO as their work functions to ZnO are 4.28eV and 4.33eV, respectively [14]. In this work, electrical and microstructural investigations on Ti/Al bilayer ohmic contact to *n*-ZnO deposited by an RF magnetron sputtering technique are studied in completely. The structural and electrical stability of the contacts at different annealing temperatures were studied.

2. Experimental Procedure

In this work, AlN (about 200nm) films were grown on three-inch silicon substrates by plasma-assisted molecular beam epitaxy (PAMBE) as the buffer layer [4]. ZnO nanorod were synthesized by using a hydrothermal method on silicon (Si) substrate. The ZnO seed layer solution was prepared using 0.005M zinc acetate dehydrates dissolved in 50ml absolute ethanol. For the purpose of enhance the wetting of the seed layer on silicon substrates, the substrates were illuminated by UV lamp for 20 min. The formation of the seed layer on silicon substrate was conducted via drop coating technique. A seed layer is vital in the synthesis of aligned vertical nanorods as the seed will perform to lower the nucleation barrier. After ZnO seed layer coating on the silicon substrates, the substrates were post annealed in air at 400ºC for 30 min. Silicon substrates with seed layer were then vertically inserted into the aqueous precursor solution.

Then ZnO nanorods were grown on the seed layer by using hydrothermal method in 0.025M aqueous precursor solution of zinc acetate dehydrate $(\mathrm{Zn(Ac)}_2 \cdot 2\mathrm{H}_2\mathrm{O}$, Sigma-Aldrich, 99%) and hexamethyltetramine (HMTA, Sigma-Aldrich, 99%) dissolved in 200ml deionized water at low temperature (90°C) for 4 hours. After nanorod growth, the substrate was separated from the chemical dissolvent, accompanied by rinsing with DI water, and followed by dried in air. The length of ZnO nanorods can be self restrained by the number of repetitive chemical reaction and time parameters.

An undoped ZnO nanorods with a thickness of 0.2mm was grown. The electron concentration was $2 \times 10^{18}\text{cm}$, and the electron mobility was $40.2\text{cm}^2 / \text{Vs}$, determined by Hall measurements. The native oxide was eliminated in the $\text{NH}_4\text{OH} : \text{H}_2\text{O} = 1 : 20$ solution for 5 min, then rinsed with deionised water. Consequently, the samples were dipped into $\text{HF} : \text{H}_2\text{O} = 1 : 50$ for 10s, then rinsed with deionised water. The cleaned samples were then chemically etched in boiling aqua regia of $\text{HCl} : \text{HNO}_3 = 3 : 1\ (5\,\text{min})$ to diminish the quantity of carbon contamination of the ZnO surfaces. Wafers were then blown dry with compressed air after cleaning and are ready for the next fabrication step [5]. Firstly, titanium (Ti) with 300nm thickness was sputtered onto the ZnO nanorods through a metal mask, followed by 100nm capping layer of aluminium (Al). The substrates were retained at room temperature during the sputtering.

The characteristics of using this mask in fabricating the mask is that no mesa isolation is required to be fabricated. Figure 1 will additional refine on how the structures are fabricated. The cross dimension of samples applied for the linear TLM structures have to be shorter than the distance across of the metal mask. The contacts were patterned in TLM structures, which start of six rectangular contacts separated by known distance (see Figure 1). The transmission line method (TLM) pads were designed to be 2mm (Z, width) × 1mm (d, length) in size and the spacings, L between the pads were 0.3, 0.4, 0.6, 0.9, and 1.3mm [5]. The specific contact resistivities, ρc were ascertained from the plot of the measured resistances versus the spacing between the TLM pads. The linear-square technique was applied to fit a straight line in the experimental data [7].

Thermal annealing was accomplished in a typical chamber furnace by heating the samples. The gas is flow at a mass flow tempo of roughly 4L min^{-1}. The samples were annealed under flowing nitrogen atmosphere between 200°C and 500°C for 5 min in a typical tube furnace. Alike heat treatments were accomplished for further annealing times of 10 and 20 min to inquire into the thermal stable of the interface contacts. All of the measurements were made in atmosphere at room temperature. The novelty in the surface morphology of the contacts on annealing was studied applying scanning electron microscopy (SEM) [5].

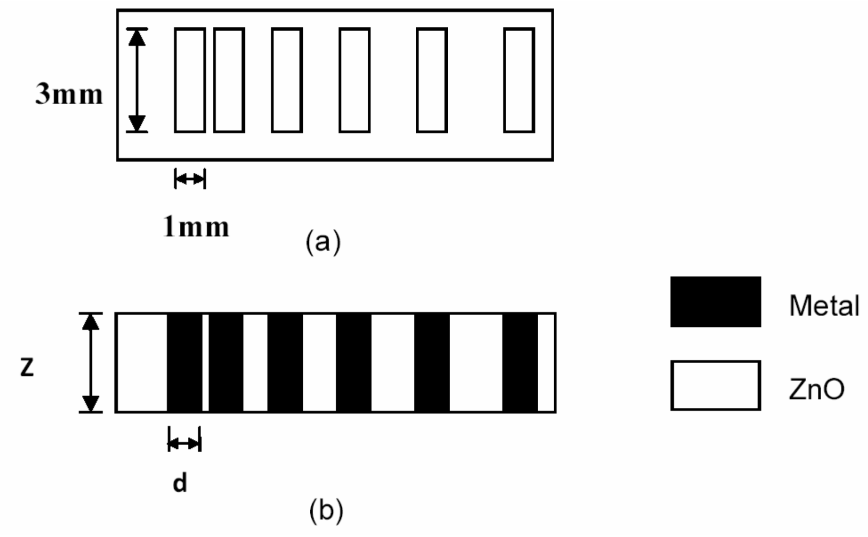


Figure 1. (a) Metal mask used to fabricate the linear TLM pads and (b) samples after metallization with fabricated linear TLM pads [5].

3. Results and Discussion

The surface morphology of the hydrothermal sample was studied by field emission scanning electron microscope (FE-SEM). They exhibit a tendency to grow perpendicular to the silicon surface with AlN as buffer layer. ZnO nanorods with diameters of 100-140nm with a narrow wire-

wire distance can be shown in Figure 2. No other nanostructures are acquired exclude the ZnO nanorods, it shows the high purity. The SEM images obvious proof that continues nanorods grown on the silicon substrate surfaces with high density. One of the most satisfactory features of SEM analysis is energy dispersive X-rays analysis (EDX). Figure 3 is the corresponding EDX spectrum of the ZnO nanorods grown on a silicon surface with AlN as buffer layer.

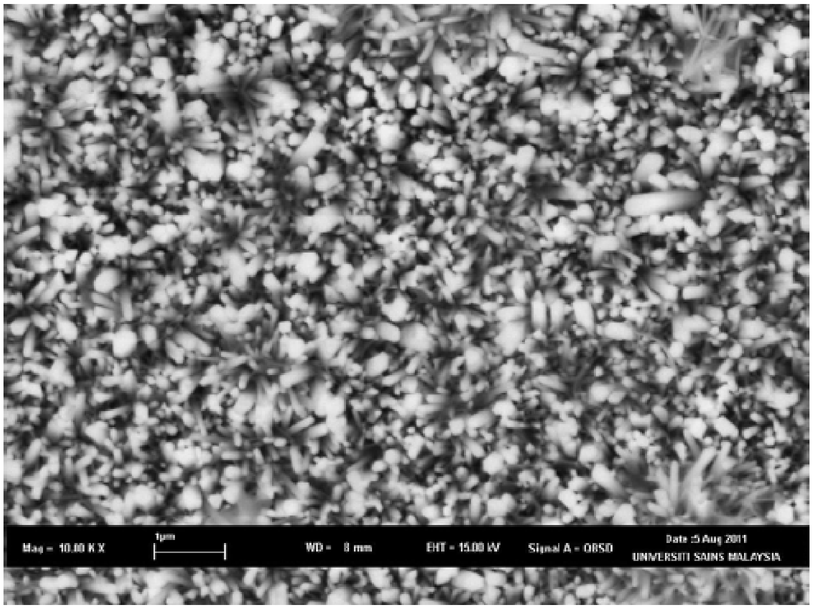


Figure 2. ZnO nanorod grown on a silicon surface with AlN as buffer layer.

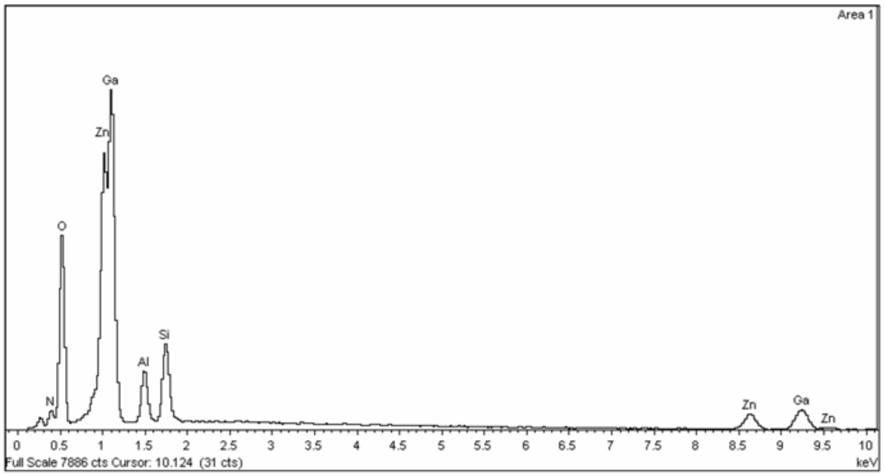


Figure 3. The corresponding EDX spectrum of the ZnO nanorods grown on a silicon surface with AlN as buffer layer.

The outcomes confirmed that as-deposited contacts proved Schottky characteristics. The surface oxide is disclosed to decrease effective Schottky barrier height and to worsen the metal/ZnO interface, foremost to the degradation of ohmic contacts. In the literature review, the Ti metallization was quoted as a wetting representative, however, it was deduced that Al has additional character involving increase of the surface oxides and reacting with ZnO at low temperatures to form binary and ternary compounds that might be electrically vital.

With the TLM approach, the specific contact resistivity (ρ_c) is calculated from a measurement of the resistance (R_i) between two contacts with spacing l_i, is given by [8]

$$R_i = \frac{R_{sh} l_i}{W} + \frac{2 R_{sk} L_t}{W}, \tag{1}$$

$$R_i = \frac{R_{sh} l_i}{W} + 2R_c, \tag{2}$$

where W is the width of the pad, R_c is the resistance because of the contact, R_{sh} is the sheet resistance of the semiconductor layer exterior the contact district, R_{sk} is the sheet resistance of the layer instantly concealed by the contact, and L_t is the transfer length. In the planar contact form, almost all of the current pass in the semiconductor throughout a tiny area at the boundary of the contact. The plot of R_i as a function of l_i will yield a straight line with a slope of R_{sh} / W, and $2R_c$ is yielded from the intercept at y-axis [29]. The intercept at x-axis, will give L_x, where

$$L_x = \frac{2R_{sk}L_t}{R_{sh}} \approx 2L_t, \tag{3}$$

with the presumption that $R_{sh} = R_{sk}$. As one point of view, the hypothesis of an electrically elongated contact $d >> L_t$ well-filled the relationship $\rho_c = R_{sh}L_t^2$ to be called, which leads to $\rho_c = R_c W L_t$ [7].

Thermal treatments were applied to figure tunneling contacts by alloying between the contact metal and a semiconductor. In Table 1, specific contact resistivities studied by the TLM, pointed out that higher annealing temperatures inducement to the furtherance of the contact resistances. The specific contact resistivity decreases with the rise of annealing temperature up to 500ºC. It strength be associated to the formation of $\mathrm{Al_3Ti}$ [30].

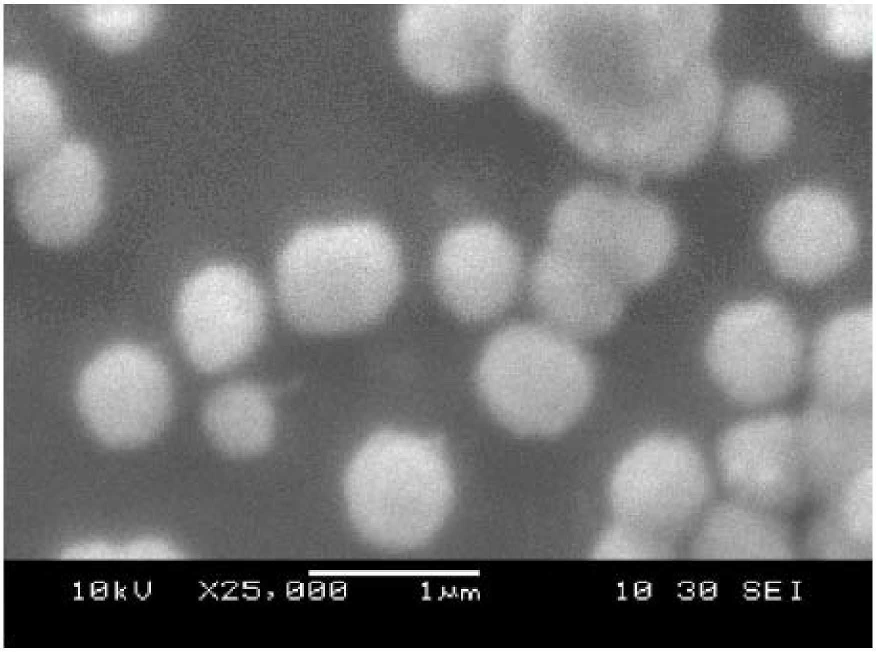
Table 1. The specific contact resistivities at different annealing temperatures and times

Annealing temperature (ºC)	Specific contact resistivities (Ω - cm^2)		
	Time / (cumulated time)		
	5 min	10 min/ (15 min)	20 min/ (35 min)
200°C	nearly ohmic	1.013	1.008
300°C	1.021	0.721	–
400°C	0.389	0.213	–
500°C	0.221	0.062	–

For annealing time of total 15 minutes, samples revealed develop decrease of the contact resistances for annealing temperatures between 300°C to 500°C. The annealing convinced enhancement of the electrical properties of the Ti/Ag contacts could be associated to elimination of agglomeration and interfacial voids, which were ascertained in the single contacts. The utilization of the interlayer was able to obstacles the generation of interfacial voids at the metal interfaces. The synthesis of AlO_2 layer could enrich the bond by associate with metal Ti and it enhance adhesion between the ZnO and metal Al layer. From the literature, non-alloyed Al schemes were investigated to reveal an ohmic characteristics due to its engaging in the interfacial oxide constitution yielding Al_2O_3, which promote the tunneling of carries [18, 20, 32].

The creation of interfacial voids will lessen contact areas and instantly will increases contact resistivity. In this work, a notable furtherance in the characteristics of sample, which was annealed at 500°C was captured. The contact characteristic of all the samples was depreciated by the following 35 minutes heat treatment.

The surface morphologies of Ti/Al contacts on ZnO with various anneal temperatures and duration were acquired. SEM images in Figure 4 shows that the surface morphology of contacts annealed in nitrogen ambient for 35 minutes experience ‘balling up’ effect at 400°C as distinguished to a smooth-faced surface of the as-deposited sample. The noted surface morphology evolve into smoother as the sample annealing temperature is rised to 500°C, which in turn degraded later after annealing at 600°C and 700°C. Interdiffusion in the metal part of stack after annealing caused the film surface to become rougher as compared with the as-deposited sample [29].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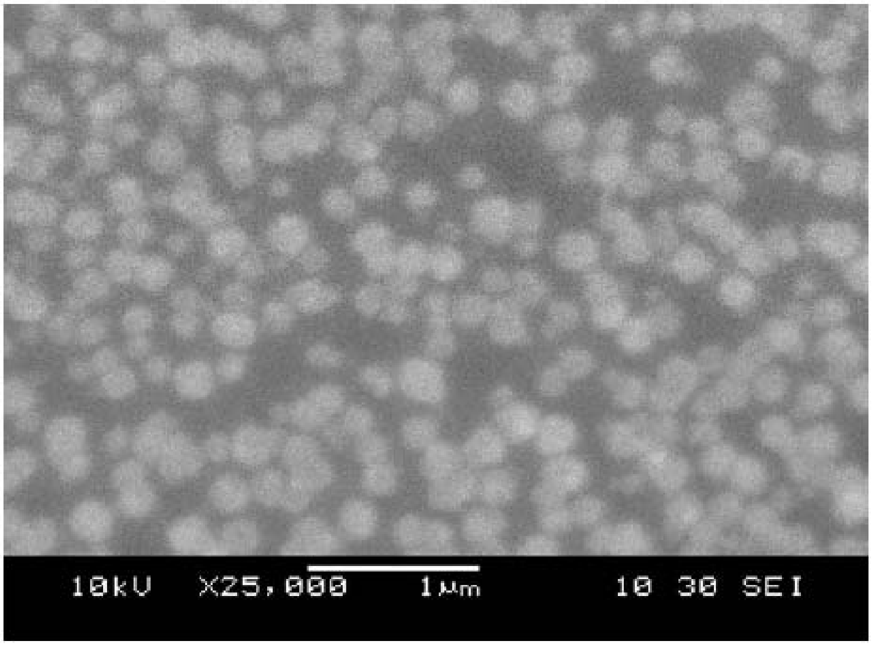
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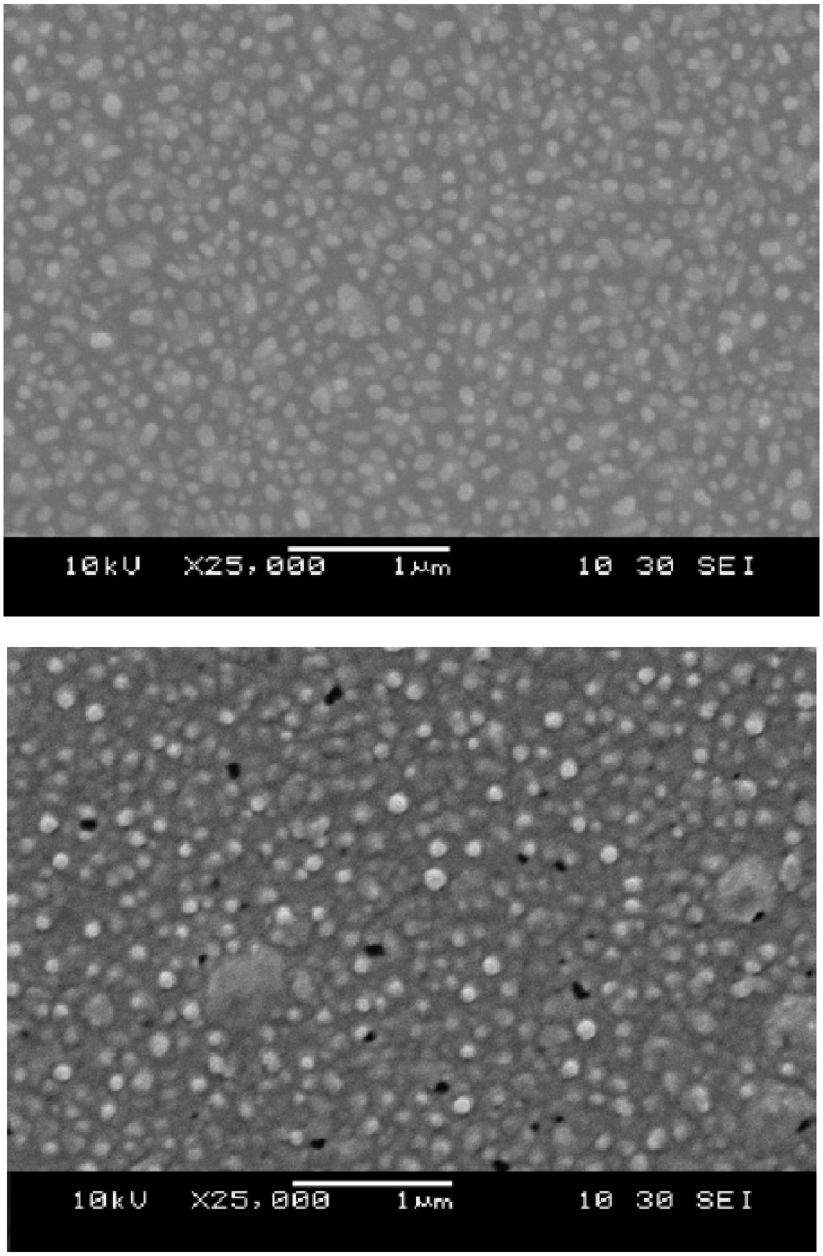

Figure 4. Ti/Al contacts on ZnO annealed for 35 minutes with (a) 200ºC; (b) 300ºC; (c) 400ºC; and (d) 500ºC in nitrogen ambient.

When aluminium or titanium is close contact with ZnO, it can involve with oxygen to form interface defects that enlarge subsurface doping density and deteriorate contact resistance. This contact has positive barrier heights at the metal-semiconductor interface, but the width of the potential barrier is thin sufficient for carriers to get over. Titanium has a higher affinity with oxygen than Zn [34]. As the contact forms, oxygen atoms move toward the Ti layer, which could have resulted in the production of oxygen vacancies at the ZnO surface. In consideration of the oxygen vacancies act as electron donors inside ZnO, these reactions advance the *n*-type carrier concentration of the ZnO surface layer, hence reducing the barrier width. This out-diffusion of oxygen is additional improved by thermal annealing [2].

While annealing, the oxygen atoms can drift from the ZnO lattice pointing to the metal layer, removing oxygen vacancies near the surface of ZnO [2]. The gain in carrier concentration of the *n*-type ZnO effect a decrease of the depletion region width. Consequently, the possibility of tunneling is increased and specific contact resistivity is dramatically reduced [16].

In fabrication alloyed ohmic contacts, the annealing temperature is vital. Annealing performed over the optimum temperature causes out-diffusion of both Zn and O atoms pointing to metal layer, effect severe dissociation of the uppermost ZnO layer. In addition, metals are quickly oxidized at high temperature, finally turning them into insulators, leading to notable increasing the specific contact resistivity. The interdiffusion of dissimilar metal layers also deteriorates the characteristics of ohmic contact because of the formation of different intermetallic compounds and enhance the roughness of metal surfaces. The alloyed ohmic contact with low specific contact resistivity and good thermal stability is vital for wide bandgap semiconductors, for example, ZnO, particularly for high power and high temperature utilizations [2, 16, 34]. Table 2 shows a brief of reported results for ohmic contacts to ZnO.

Table 2. Reported ohmic contact schemes on *n*-type ZnO

Contact schemes	Thermal treatment (ºC)	Specific contact resistivity $(\Omega\text{-cm}^2)$	Annealing condition	Authors
Ti/Au	–	4.3×10^{-5}	Nonalloyed	Lee et al. (2001) [26]
Al/Au	200	1.4×10^{-4}	N_2	Kim et al. (2008) [19]
Ti/Au	300	2×10^{-4}	N_2	Kim et al. (2001) [14]
Ta/Au	300	5.4×10^{-6}	N_2	Sheng et al. (2003) [33]
Ti/Al	300	9.7×10^{-7}	N_2	Kim et al. (2002) [21]
Re/Ti/Au	700	1.7×10^{-7}	N_2	Kim et al. (2005) [23]
In/Zn	550	2.4×10^{-6}	Ar	Zhuge et al. (2005) [36]
Ni/Au	450	2.06×10^{-4}	N_2	Lu et al. (2008) [27]

4. Summary

In conclusion, Ti/Al contact structures on undoped ZnO were fabricated and characterized. For its specific contact resistivity (SCR), it was perceived that the as-deposited contacts showed nonlinear ohmic characteristics, but became ohmic upon annealing. This possible corresponds to the creation of the alloy phases, which aid to enhance the doping. The specific contact resistivity of this Ti/Al scheme was sensitive to the alter of annealing temperatures and durations. Al and Ti have been used for ohmic contacts to *n*-ZnO as their work functions to ZnO are 4.28eV and 4.33eV, respectively. The results indicate that the Ti/Al bi-layer scheme as a dormant essential scheme for the high achievement flip-chip light emitting diodes.

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