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A Comparative Study of $p^{\dagger}n$ and $n^{\dagger}p$ InP Solar Cells Made by a Closed Ampoule Diffusion

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ABSTRACT

The purpose of this study was to demonstrate the possibility of fabricating thermally diffused p⁺n InP solar cells having high opencircuit voltage without sacrificing the short circuit current. The p⁺n junctions were formed by closed-ampoule diffusion of Cd through a 3-5 nm thick anodic or chemical phosphorus-rich oxide cap layer grown on n-InP:S(N_D = 3 x 10¹⁶ and 5 x 10¹⁷ cm⁻³) Czochralski LEC grown substrates. After thinning the emitter from its initial thickness of 1 to 2.5 μ m to 0.06 - 0.15 μ m, the maximum efficiency was found when the emitter was 0.2 to 0.3 μ m thick. Typical AMO, 25° C values of 854-860 mV were achieved for V_{oc}. J_{sc} values were from 27.5 to 29.1 mA/cm² using only the P-rich passivating layer left after the thinning process as an anti-reflection coating.

For solar cells made by thermal diffusion we expect the p^tn configuration to have a higher efficiency than the n^tp configuration. Based on this study we predict that the AMO, BOL efficiencies approaching 19% should be readily achieved providing that good ohmic front contacts could be realized on the p^t emitters of thickness lower than 1 μ m. If, as we expect, the p^tn structures prove to be at least as radiation tolerant as n^tp structures, then p^tn InP solar cells made by thermal diffusion can become attractive for space applications, due to a relatively low fabrication cost, for this method of fabrication.

INTRODUCTION

Owing to its potential low cost, reduced complexity and adaptability to large scale batch processing thermal diffusion is a desirable technique for p^+n or n^+p InP junction formation.

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Most research on InP solar cells, so far has concentrated on the n⁺p configuration. However, there is no apriori reason why the n⁺p configuration should yield higher BOL efficiency and/or higher radiation tolerance than the p⁺n configuration.

 $n^{+}p$ InP solar cells made either by closed ampoule diffusion of sulfur into InP:Zn [1] or InP:Cd [2] substrates or by open tube diffusion [3] have been extensively studied. Good quality p⁺n InP solar cells on the other hand, have been fabricated only by epitaxial techniques [4,5]. This may be so because of the difficulty of 1) fabricating thin p⁺ emitters in a controlled manner, 2) forming good ohmic contacts to p⁺ surfaces and 3) passivating p⁺ surfaces.

The results we report here are part of a larger experimental effort undertaken in an attempt to optimize the processing of InP homojunction solar cells made by thermal diffusion, so as to achieve high-efficiency low cost InP solar cells by this method of junction formation.

We have previously reported on limitations of n⁺p InP solar cells made by closed ampoule diffusion [2,6]. Therefore, most emphasis here is on the p⁺n configuration. Specifically, the objective of this preliminary work was to demonstrate the possibility of fabricating thermally diffused p⁺n InP solar cells having high open-circuit voltage (V_{oc}) without sacrificing the short circuit current (I_{sc}).

In developing high-efficiency, radiation resistant p⁺n InP solar cells made by thermal diffusion our short-term effort, still in progress, consists of:

- 1. Investigate the effects of i) various surface preparation procedures including chemical treatments and anodic or chemical oxidation, ii) choosing between Zn and Cd diffusants, in elemental form or as Zn_3P_2 and Cd_3P_2 . and iii) diffusion through bare and capped surfaces, on the characteristics, reproducibility and quality of p⁺n InP structures made by closed ampoule thermal diffusion of Zn or Cd into Czochralski LEC grown n-InP:S substrates;
- 2. Design optimum front contact grid pattern and form good ohmic contacts on the emitters of thickness lower than 1 $\mu\text{m};$
- 3. Choose from among the following processes for thinning the p^+ InP emitter and optimize the processing parameters:
 - a) anodic dissolution;
 - b) anodic oxidation-removal cycles;
 - c) chemical etching,

so that the residual oxide left as a result of thinning i) is stable, ii) has good passivating properties at the $p^+/oxide$ interface, and iii) can possibly be used as a first layer AR coating.

- 4. Find an appropriate second layer AR coating.
- 5. Radiation tolerance measurements.

Based on previous reports on n^+p InP solar cells made by closedampoule diffusion [1, 2, 6] and this work, we evaluate the limitations of the n^+p and p^+n configurations in order to choose the structure and the fabrication procedure of solar cells made by thermal diffusion with efficiencies approaching those of epitaxially grown structures [5, 7].

EXPERIMENTAL

Cd and Zn diffusions into n-InP:S ($N_p = 3.5 \times 10^{16}$ and 4.5×10^{17} cm⁻³) were performed by a closed ampoule technique, using high purity Cd and Zn or Cd₃P₂ and Zn₃P₂ [8]. Diffusion temperatures were from 480 to 550° C in the case of Zn and from 550 to 600° C in the case of Cd diffusion. Diffusion times were from 5 to 75 minutes. The substrates were Czochralski LEC grown with EPDs from 3 x 10⁴ to 7 x 10⁴ cm⁻².

Diffusions were performed through bare surfaces or by using cap layers of: i) resistively evaporated SiO_2 (50-100Å thick) [9]; ii) phosphorus rich anodic [10] and chemical [11] oxides (35-50Å thick).

The quality of $p^{+}n$ InP structures was investigated from:

- Inspection of the post-diffused surface topography using Nomarski and SEM microscopy;
- Revealing the surface and deep precipitates after thinning down the p⁺ layer to different depths by anodic dissolution using the FAP electrolyte, and SEM inspection [12].
- 3. Investigation of surface and deep etch pit density revealed electrochemically using the FAP electrolyte at high current densities (1-3 mA/cm²) under illumination [12].
- Electrochemical C-V; 1/C²-V, G-V and I-V characteristics by using a Polaron PN4200 profiler.
- 5. Performance parameters of p'n InP solar cells fabricated on selected p'n structures.

RESULTS AND DISCUSSION

p⁺n InP JUNCTION FORMATION

Using elemental Zn and Cd as diffusants, the p^* surfaces were seriously pitted even when phosphorus-rich oxide capping was employed.

Using high purity Zn_3P_2 and Cd_3P_2 sources, significant improvements in the surface and p⁺n diode quality were achieved. Due to space limitation we are going to refer here only to diffusions using the compound sources.

Diffusion through bare surfaces has led to unacceptable levels of surface defect densities, e.g., as high as 10^9cm^{-2} in the case of Zn-diffusion and of up to about 5 x 10^7cm^{-2} in the case of Cd-diffusion.

The surface quality could be improved by diffusion through resistively evaporated SiO₂ cap layers. In the case of Cd-diffusion EPDs values as low as 8 x 10^5 cm⁻² were recorded, after removing the SiO₂ cap layer and the contaminated front emitter layer (about 100Å) from the surface using the FAP electrolyte [12]. However, from electrochemical C-V, $1/C^2$ -V, G-V and I-V characteristics, in this case, it was found that relatively good diode characteristics were obtained only after removing the front 0.2-0.3 µm layer from the surface.

Using phosphorus-rich anodic and chemical oxide cap layers, the quality of $p^{+}n$ structures was dramatically improved.

Anodic oxides were grown in $o-H_3PO_4:H_2O:ACN$ (1:200:300) under strong illumination ($\approx 100 \text{ mWcm}^{-2}$) at a constant current density of 0.6 mA/cm⁻² [10]. Prior to diffusion the front In-rich oxide was removed by a short dip in dilute HF (5%) solution, and the thickness of P-rich interfacial oxide layer used as a cap was varied from about 25 to 50Å (cell voltage: 30 to 40 V).

Chemical oxides were grown using a newly developed etchant, called "PNP" [11] with general formula: $(o-H_3PO_4)_u:(HNO_3)_v:(H_2O_2)_t:(H_2O)_{100-(u+v+t)}$, which grows an interfacial layer rich in $In(PO_3)_3$ of thickness which is proportional to the etching time and depends on the composition of the etchant.

In diffusion due to its higher diffusivity as compared to Cd diffusion is hard to control. Furthermore, not only do the surface precipitates have a high density but these precipitates extend deep into the p⁺ emitters. The surface precipitates have a density ranging from 5 x 10^4 to about 5 x 10^8 cm⁻² depending on the diffusion temperature, amount of evaporating material and temperature gradient between the end of the quartz ampoule containing the source and the substrates region, and the P-rich diffusion cap being used. We observed that keeping the source region at a lower temperature (about 10° C below the substrate region) improves the diode quality by reducing the surface precipitates.

In the case of Cd diffusion the precipitates which had a density ranging from 4×10^2 to 6×10^7 cm⁻², depending on the diffusion conditions, appear to be superficial. After removal of about 50Å from the surface (below the oxide layer), no deep precipitates could be revealed.

Figure 1 shows a typical view of p^+ surface topography for Cd and Zn diffusion through capped and uncapped surfaces. As seen, specular surfaces are obtained in the case of Cd diffusion through the thin P-rich oxide cap layer.

Surface dislocation densities as seen in Figure 2 revealed either electrochemically using the FAP electrolyte [12] at a high current density or chemically using the Hubert etchant, have shown that extremely low EPDs are achieved in the case of Cd diffusion using P-rich anodic or chemical cap layers. The lowest EPD of 400-800 cm⁻² in this case was obtained at a diffusion temperature of 560°C with a temperature gradient between the source and substrates of about 15°C. EPDs were about one order of magnitude higher if no temperature gradient was used (see Figure 2B) and as high as 10° cm⁻² in the case of Zn diffusion through bare surfaces (Figure 2C).



Figure 1. SEM view of surface topography of p'n InP structures made by diffusion of Cd at 560°C (A & B) and Zn at 540°C for 30 min (C) through about 35 Å thick chemical oxide cap layer (A & B) and bare surface (C). Temperature gradient between the surface and the substrates: A) 15°C; B) 0°C; C) 15°C.



Figure 2. EPD's revealed electrochemically (A & B) using the FAP electrolyte at 2 mA/cm² under illumination and C) chemically using $HBr:o-H_3PO_4$ (1:2) for structures in Figure 1.

From electrochemical C-V, $1/C^2-V$, G-V, and dark and illuminated I-V characteristics, which were done for

all p⁺n structures, the diode quality was found to be the best for Cd diffusion through the P-rich oxide cap layers. An example of these characteristics after removing about 0.03 μ m is shown in Figure 3. As seen quasi-ideal characteristics are obtained in this case even at a diffusion temperature of 600° C. The diode characteristics in this case were found to be near perfect even after removing just the front oxide layer, implying an absence of a front dead layer, which proves that the P-rich oxide is a very good choice as a diffusion cap layer.

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Figure 3. Electrochemical C-V, $1/C^2-V$ and G-V characteristics using the FAP electrolyte [12] for a p⁺n InP structure, Cd diffused at 600°C for 10 min, after removing 0.03 μ m from the surface.

After Zn-diffusion, in the best case (diffusion at 540° C) reasonably good diode characteristics were obtained only after the removal of about 0.4 to 0.5 μ m from the surface.

Electrochemical C-V (EC-V) profilings performed using the FAP electrolyte [12] have shown that the net acceptor concentration, (N_A-N_D) , at the surface, in the case of Cd-diffusion for diffusion temperatures of 550 to 600°C, was from 9 x 10^{17} to 2 x 10^{18} cm⁻³, depending on the diffusion conditions. In Figure 4 are shown two EC-V profiles after Cd diffusion through about 40Å P-rich chemical oxide cap layer into InP:S of two doping levels. As expected the diffusivity of Cd

into the highly doped substrate appears lower than in the low doping substrate.

p'n InP SOLAR CELL PERFORMANCES

Small area (0.48 cm^2) p⁺n InP solar cells were fabricated on structures diffused at 560°C, the diffusion temperature at which the



Figure 4. EC-V profiles of two p*n InP structures made by Cd diffusion into n-InP:S through a In(PO₃)₃-rich chemical oxide cap layer (~40 Å thick).

surface dislocation density is minimum.

Since Au-Zn-Au front contacts melt as far deep as 2 μ m into InP during sintering at 430°C, p'n structures with thick emitters were fabricated. Therefore, after front contact sintering at temperatures from 390°C to 430°C, the emitters had to be thinned down over the uncontacted areas. Anodic oxidation-removal cycles and chemical etching were considered. Since none of the etchants known could be used, to us a new etchant which we call the "PNP" etchant wad developed [11]. Resulting surfaces using this etchant are very smooth as seen in Figure 5 and a P-rich oxide grows during the dissolution

process. Performance parameters were recorded at CSU after each thinning





Figure 5. Nomarski micrograph of a surface after thinning the p^{*} layer from ~2.5 μ m to ~0.1 μ m using the "PNP" etchant, (u = 15, v = 45, t = 1) [11].

step (see figure 6a). Up to 150 steps **Figure 6**. Illuminated I-V characteristics were used in the case of chemical of cell #33 using a) an ELH lamp and b) under AMO, 25°C conditions. emitter thickness. For selected cells, illuminated I-V (see Figure 6b) dark I-V, I_{sc} -V_{oc} and spectral response measurements were done under AMO conditions at NASA-LERC.

Dark I-V, I_{sc} -V_{oc} characteristics, have shown that the diode quality factor calculated at the maximum power point was from 1.03 to 1.08 and the dark saturation current density from 8 x 10⁻¹⁷ to 2 x 10⁻¹⁶ A/cm².

As seen in Figure 7, I_{sc} values increase with decreasing emitter thickness and reaches a maximum at 0.15 to 0.25 μ m. η_{max} was found for emitter thickness of 0.2 to 0.3 μ m. This means that most of the current comes from the low defect density emitter and space charge regions. The low FF values are due to the large values of R_s due to an inappropriate front grid design. The decrease of FF as a result of thinning the emitter is directly related to an increase in R_s due to a large increase in sheet resistance once the surface concentration goes below 9 x 10¹⁷ cm⁻³.

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Figure 7. Relative variations of I_{sc}, V_{oc}, FF, η , R, and R_{sh} as functions of junction depth of a p⁺n InP solar cell (#17).

Table 1 shows the variation of the short circuit current density J_{sc} and the open circuit voltage V_{oc} for several p'n InP solar cells fabricated by closed ampoule diffusion of Cd into InP:S substrates after thinning the emitter from about 2.5 μ m to 0.06-0.15 μ m by anodic and chemical oxidation, as a function of the oxide type and thickness. As seen, by using the PNP-2 etchant [11] the largest J_{sc} and V_{oc} values have been obtained. The high $V_{\rm oc}$ values for these thermally diffused p'n InP solar cells correlate well with the low surface state density minimum $N_{ss,min}$ of 3 x 10^{10} cm⁻² eV⁻¹ recorded after dissolution of a p'n InP structure using the PNP-2 etchant, from 2 μ m down to \approx 0.25 μ m. The oxide thickness in this case was \approx 120 nm. The N_{ss} values were calculated from the conductance-voltage (G-V) data at different frequencies using a Polaron PN 4200.

In-rich surface layers have previously been identified to be a possible cause of the high density of interface states [13]. From the large enchancement of Photoluminescence Intensity as a result of removing the In-rich surface oxide layer [14], it will appear that this is the case. This might explain the data in Table 1 which show an increase in $V_{\rm oc}$ of anodic oxide covered solar cells after

Table 1. J_{sc} and V_{oc} of selected p⁺n InP Solar cells after thinning the p⁺ emitter by anodic oxidation-removal and chemical etching with PNP etchant [11].

Cell #	Oxide type	Approx. oxide thickness (nm)	Before removing the front oxide layer (b)		After the fro laye	J _{sc} (b)/J _{sc} (a) (そ)	
			J _{sc} (b) (mA/cm ₂)	V _{oc} (b) (mV)	J _{sc} (a) (mA/cm ₂)	V _{oc} (a) (mV)	
20(c)	anodic	120	25.69	849	23.33	854	90.8
22 (d)	chemical	. 850	27.00	859	22.43	852	83 1
23(e)	chemical	. 150	27.65	859	22.33	850	80.0
26(f)	anodic	130	20.80	846	21.22	851	102 0
27 (g)	chemical	400	27.96	859	23.04	853	82.4
33(h)	chemical	. 120	29.10	860		_	-

* Illuminated I-V characteristics measured at NASA LeRC, under AMO, 25°C conditions; (c) grown in 3% citric acid in ethylene glycol (1:3) buffered to pH7 with 20% NH4OH solution; (d), (e) and (g) grown in PNP-1 etchant; (d): (u=0, v=60, t=1); (e): (u=15, v=45, t=1); (g): (u=5, v=60, t=1); (f) grown in o-H_3PO_4:H_2O:ACN (1:200:300) [10]; (h) using PNP-2 etchant: (u=10, v=30, t=1); where the PNP etchant is: $[(o-H_3PO_4)_u: (HNO_3)_v: (H_2O_2)_t: (H_2O_1)_{100-(u+vt)}]$

3-8

removing the front In-rich oxide layer. As seen, after removing the front oxide layers, the solar cell performance parameters are comparable for the cells made using the two thinning processes, namely, the PNP-1 etchant and anodic oxidation. However, the cell performance parameters are even better using the PNP-2 etchant. Since the anodic oxidation-removal process is very laborious and hard to control [10] as compared to a simple chemical etching process, the latter seems to be the best answer.

The above $V_{\rm oc}$ values, measured at NASA LeRC under the AMO, 25° C conditions, are higher than any previously reported $V_{\rm oc}$ values for InP solar cells made by thermal diffusion.

Structure	Junction formation technique	Dopant	Approx. Junction depth (µm)	Voc (mV)	JSC mA/cm ²	FF (%)	ग (%)	References
p*-i-n	LPE	Mg/S	0.6	823.7	37.6 (active area)	75.4	17.2 (active area)	Itoh et al., NTT [4] (Measured)
p'(InGaAs)/	LPE		0.7	866	29.25	81	15	Shen, et al. [5]
p ⁺ (InP)/n(InP)/ n ⁺ (InP)	MOCVD	Zn/S		864	32.84	76.7	15.9	(Measured)
n ⁺ -p	Closed Ampoule	S/Zn	0.2-0.3	828	33.7	81.6	16.6	Okazaki et al., NTT [1] (Measured)
n*-p	Closed Ampoule	S/Cd	0.15 0.08-0.1	806 840	30.5 36.5	80.1 84	14.35 18.8	Faur, et al., CSU [2] (Measured) (Predicted)
p [*] -n (Cell 33)	Closed Ampoule	Cd/S	0.25 0.25 0.25-0.3	860 880 890	29.1 34.5 37.4	52.2 83 84	9.52 18.36 20.4	Faur, et al., CSU, (Measured) This work. (Predicted) (Predicted)

Table 2. Measured or Predicted AMO, 25°C p/n or n/p InP Solar Cell Performances



In Table 2 is shown a review of performances of p^{*}n InP solar cells including those measured and predicted for our high performance p^{*}n junctions. For comparison, performances of n^{*}p InP solar cells made by closed-ampoule sulfur diffusion into Zn and Cd doped substrates are also given in this table.

One can see that our measured $V_{\rm oc}$ and $I_{\rm sc}$ values are higher or very close to

Figure 8. External Quantum Efficiency plots of selected p⁺n InP solar cells measured under AMO, 25°C conditions at NASA LeRC.

previously reported values of epitaxially grown p^+-i-n [4] or $p^+(InGaAs)/p^+$ InP/n(InP)/n^+ InP [5] solar cells.

From the external quantum efficiency plots, seen in Figure 8, an increase of 15 to 25% in I_{sc} can be expected by optimizing the passivating layer and using a second layer AR coating. From the $I_{sc}-V_{oc}$ characteristics such an increase in I_{sc} will produce an increase of about $I_{sc} - V_{oc}$ 10 mV in V_{oc} . By further optimizing the diffusion process we estimate that a V_{oc} value of 880 mV is readily achievable using these high performance thermally diffused p'n InP structures. Assuming that by reducing R_s , FF values of 83% can be achieved, a simple computation based on presently measured $V_{\rm oc}$ and $I_{\rm sc}$ values indicates that AMO efficiencies of over 18% are readily achievable. By further reducing the external losses (i.e. 4% grid coverage and below 5% overall reflexivity), using better quality substrates (i.e. defect density below 10⁴cm⁻²), optimizing base doping, further optimizing the diffusion process and the quality of the passivating layer, AMO, 25° C V_{oc} values as high as 890 mV for solar cells fabricated by this method of junction formation should be possible. The $J_{sc,max}$ in this case is of about 37.4 mA/cm². Assuming a FF of 84%, the expected maximum AMO efficiency is of 20.4%.



Figure 9. Variation of I_{ph} and majority carrier concentration (N_p) as function of depth after photoanodic dissolution using the FAP electrolyte [12] of an n⁺p InP structure made by thermal diffusion [2]:

By comparison, the maximum reported AMO efficiency for an n⁺p InP solar cell made by thermal diffusion is 16.6% [1]. Voc in this case is the principal limiting parameter. Based on our thorough investigation on n†p InP thermally diffused structures [2, 6] we estimate that the maximum achievable V_{oc} is about 840 mV which is in close agreement with previously predicted values [1]. The maximum J_{sc} in this case is limited 36.5 to mA/cm^2 . Assuming a similar FF value of 84%, the maximum AMO efficiency in this case is of about 18.8%. These values were calculated

for an n⁺p InP solar cell with similar minimal external losses as described above, for a stucture fabricated by sulfur diffusion into Cd-doped InP substrates, using a thin $In(PO_3)_3$ diffusion cap layer and removing the phosphorus depleted dead layer from the surface (400Å).

As seen in Figure 9 which plots the variation of I_{sc} at the FAP electrolyte/n⁺ InP interface and N_d as a function of the dissolution depth, however the highest I_{sc} value is found after removal of about 400Å from the surface, which corresponds to a surface concentration N_d of about 2 x 10^{18} cm⁻³. The dislocation density minimum at this level was about 6 x 10^{5} cm⁻², as revealed electrochemically. This explains the relatively low V_{oc} and I_{sc} predicted values. However, by further thinning the emitter, I_{sc} and V_{oc} both increase and reach a maximum at about 500Å. Due

to the graded nature of the diffusion profile [6], in this case the $R_{\rm s}$ drastically increases due to an increase in sheet resistance. Because of this the maximum efficiency in this case was found for emitter thickness of 800 to 1000Å, which offers an explanation of why the $V_{\rm oc,max}$ is only 840 mV, and the $J_{\rm sc}$ is below than that expected from a well designed p⁺n InP solar cell.

Very few and contradictory comparative radiation resistance measurements of the two n⁺p and p⁺n InP configurations have been reported [1,15]. While Okazaki, et al. [1] assume the radiation tolerance scale for InP solar cells of different configurations to be: $p^+-n = n^+-p-p^+ > n^+-p^+ > p^+-i-n$, Weinberg, et al. [15] on the contrary predict the n⁺p configuration to be more radiation resistant than the p⁺n configuration. An explanation of this is that radiation tolerance evaluations were not done on structures fabricated under similar conditions. Therefore more insight in this area is necessary. We plan to do radiation resistance measurements on p⁺n InP solar cells but only after the external losses are drastically reduced, i.e. an AMO efficiency greater than 16%.

CONCLUSIONS

From our preliminary investigation it appears that optimal conditions for high quality thermally diffused p⁺n structures are: i) Cd_3P_2 as source; no added phosphorus; ii) diffusion through thin (25-40Å thick) $In(PO_3)_3$ -rich chemical oxide; iii) diffusion temperature: 560° C and iv) about 15°C temperature gradient between the source and substrates.

For solar cells made by thermal diffusion we expect the p⁺n configuration to have higher efficiency than the n⁺p configuration, due especially, to an increased V_{oc} .

Based on this study we predict that p⁺n InP solar cells with AMO, BOL efficiencies approaching 19% should be readily achievable providing that good ohmic front contacts could be realized on the p⁺ emitters of thickness lower than 1 μ m. The maximum expected AMO efficiencies for the p⁺n configuration is about 20.4%, while for the n⁺p configuration, it is about 18.8%.

If, as we expect, the $p^{+}n$ structures prove to be at least as radiation tolerant as the $n^{+}p$ structures, then $p^{+}n$ InP solar cells made by thermal diffusion can become very attractive for space applications due to a potential low cost, reduced complexity and adaptability to large scale batch processing.

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3-12