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24.5% efficient GaAs p-on-n solar cells with 120 μm h^{-1} MOVPE growth

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Abstract
By enhancing the transport of gas precursors though the chemical boundary layer using a narrow-channel reactor, the growth rate (GR) of GaAs by metal-organic vapor-phase epitaxy has been increased to 120 μm h^{-1} without saturation with an increase in TMGa supply. The minority hole lifetime in n-GaAs was clearly shortened with an increase in GR from 20 to 120 μm h^{-1}. Consequently, the short-circuit current density as well as conversion efficiency slightly decreased with an increase in GR. Nevertheless, a conversion efficiency of 24.48% could be realized for a GaAs cell with the n-type base layer grown at 120 μm h^{-1}.

Keywords: III–V semiconductors, metal-organic vapor phase epitaxy, GaAs, solar cells

(Some figures may appear in colour only in the online journal)

The rapid increase in worldwide power consumption has inspired intensive research on photovoltaic cells as a renewable energy source. During recent years, III–V multiple junction solar cells (MJSCs) have been the focus of research attention because of their high conversion efficiency, for which a record of 46% has been achieved by 4-junction structure under a concentration light of 508 sun [1]. At present, the cost of III–V semiconductor solar cells is still relatively high as compared to that of silicon solar cells, and limits their usages to some special tasks, including concentrating photovoltaic (CPV) [2, 3] and space applications [4, 5]. Much effort has been devoted recently to reduce the cost of GaAs based solar cells including the epitaxy cost, generally by metalorganic vapor phase epitaxy (MOVPE). This can be achieved by either increasing the GR (decrease machine overhead time) or improving material (source) utilization [6–8]. From price per watt analysis, the reduction in the overhead time of MOVPE reactors, together with the improvement in material utilization efficiency, can reduce the cost of GaAs single junction solar cells by 74%, when the GR of GaAs is boosted from 14 to 56 μm h^{-1} with performance loss less than 1% [7, 8]. Recently, GaAs n-on-p solar cells grown at a speed of 100 μm h^{-1} have been recently reported with a high conversion efficiency of 23.6% under AM1.5G condition [9]. They also demonstrated that the GR of GaAs could reach 140 μm h^{-1} with more than 50% of Ga source utilization. Our research group has previously demonstrated that the GR of GaAs in our MOVPE system could be enhanced to 90 μm h^{-1} [10, 11], by shrinking the boundary layer thickness for the mass transport of a Ga precursor [12]. In addition, GaAs p-on-n solar cells exhibited insignificant degradation of cell efficiency which their n-base layers were deposited at 80 μm h^{-1} using a V/III ratio of 40 as compared to the growth at 20 and 60 μm h^{-1} [10]. A high V/III ratio can plausibly be a reason for no degradation in the cell performance up to 80 μm h^{-1}. In addition, several reports have shown that the hole lifetime is less sensitive to the increase in the threading dislocation density in GaAs than the electron lifetime [13, 14]. This may make a p-on-n device more robust against an increase in defect density. In other words, the defect-tolerant structure of p-on-n GaAs solar cells may allow the growths of GaAs with a lower V/III ratio with negligible performance degradation [15]. In this work, the GR of GaAs was further enhanced to 120 μm h^{-1} which was limited by the mass flow controllers of trimethylgallium (TMGa) bubblers in our MOVPE system. The qualities including surface morphology and minority carrier lifetime of GaAs epitaxial layer at this extremely fast GR were evaluated and compared with those of GaAs grown at a lower GR. Later on, influences of GaAs GR on conversion efficiency of p-on-n solar cells were investigated.

The crystal growth in this study were carried out in a MOVPE reactor (Taiyo Nippon Sanso, HR3335) using standard precursors, including TMGa, trimethylindium (TMIn), trimethylaluminium (TMAl), arsine (AsH_3), phosphine (PH_3), diethylzinc (DEZn) and disilane (Si_2H_6). Si-doped GaAs (2–3 $\times$ 10^{18} cm^{-3}) wafers, having (0 0 1) surface with 2° misorientation toward ⟨1 1 0⟩, were used as substrates. First, unintentionally doped (ud) GaAs layers, 4 μm in thickness, were deposited with different GRs of 20, 90 and 120 μm h^{-1} with a V/III ratio of 20. Atomic force microscopy (AFM) was employed to observe the surface morphology of epitaxial layers. In this work, since a GaAs p-on-n solar cell structure will be later investigated, the hole minority lifetime in n-doped GaAs is one of the important parameters to be evaluated. The time-resolved photoluminescence (TRPL) was utilized in order to characterize the impacts of GR on the properties of minority carriers in Si-doped n-type GaAs (active carrier concentration: ~4 $\times$ 10^{17} cm^{-3}) in the n-AlGaAs/n-GaAs/n-AlGaAs doublehetero (DH) structure. Finally, GaAs p-on-n solar cells were fabricated, in which a 2 μm-thick n-GaAs base layer was grown at a speed of 20, 90 and 120 μm h^{-1}, respectively, under a V/III ratio of 20. The current–voltage (*I*–*V*) characteristic under AM1.5G illumination and the external quantum efficiency (EQE) spectrum of each solar cell with ZnS/SiO_2 anti-reflection coating (ARC) were evaluated and compared.

In an ordinary MOVPE reactor, the GR at a moderate growth temperature is limited by the diffusion of precursor fluxes from the gas phase through the mass-transport boundary layer [12]. By reducing the channel height of the flow liner in the reactor, the mass transfer of precursors to the wafer surface and the GaAs GR could be enhanced. The GR of GaAs is plotted as a function of TMGa supply in figure 1 using the growth temperature, reactor pressure and V/III ratio of 680 °C, 6 kPa, and 20, respectively. A good linear relationship can be clearly observed, suggesting that the GaAs GR could be further enhanced. In this work, the amount of TMGa supply was limited by the capability of the mass flow controllers. Figure 2 shows 2 $\times$ 2 μm^2 AFM surface images of 4 μm-thick ud-GaAs grown at a GR of (a) 20 (b) 90 and (c) 120 μm h^{-1}, respectively. Ridge-like textures were observed for all the samples along [0 0 1] direction in parallel with the step edges of the vicinal substrate. A faster GR results in a rougher surface morphology presumably due to a shorter migration time for Ga adatoms. The background hole concentrations in undoped GaAs were 1.6 $\times$ 10^{15}, 2.0 $\times$ 10^{16}, and 3.0 $\times$ 10^{16} cm^{-3} for GR of 20, 90, and 120 μm h^{-1}, respectively, as evaluated by electro-chemical voltage (ECV) measurement.

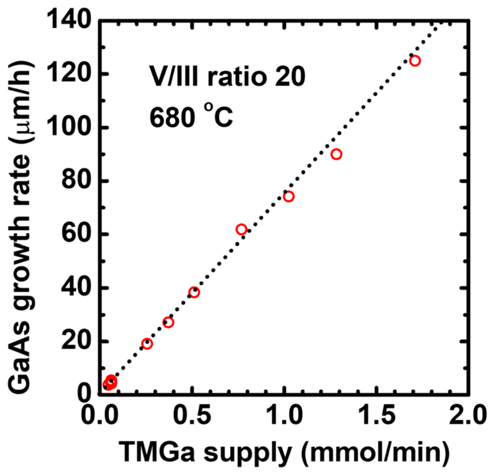


Figure 1. Dependence of GaAs GR on TMGa supply.

The AlGaAs/GaAs double hetero (DH) structure in figure 3(a), fabricated with various GaAs GRs using the common growth temperature, reactor pressure and V/III ratio of 680 °C, 6 kPa and 20, respectively, was utilized for the characterization of minority carrier lifetime. The AlGaAs hetero barriers were grown with a GR of 5 μm h^{-1} under a V/III ratio of 40. A test AlGaAs layer was fabricated and examined by AFM measurement having an RMS roughness of less than 0.2 nm. The decay transients of room temperature photoluminescence under 780 nm excitation were plotted in figure 3(b). The decay time of each transient was extracted using a single exponential model. The extracted values are given in this graph. With a faster GR, minority hole lifetime was remarkably shortened, most likely due to an increase of non-radiative carrier trap density in the n-GaAs. It should be noted here that the AlGaAs/GaAs interface quality can be affected by GaAs GR even though AlGaAs GR was kept constant and the interfacial recombination velocity can be affected by GaAs GR accordingly, which could be a reason of this degradation in minority carrier lifetime. The radiative recombination was plausibly not responsible for this result considering the majority carrier concentrations, which should dominate the radiative recombination lifetime, were controlled to be the same value, 4 $\times$ 10^{17} cm^{-3}, for all GRs as confirmed by ECV measurement. The radiative carrier lifetime calculated using a radiative recombination efficiency of 3.5 $\times$ 10^{-10} cm^3 s^{-1} [16] was 7.1 ns for the doping concentration of 4 $\times$ 10^{17} cm^{-3}. The decay times of 36.0, 21.8 and 9.8 ns for GR of 20, 90 and 120 μm h^{-1} were longer than the radiative lifetime. However, radiative lifetime should not depend on GR because the majority carrier concentration was kept at the same value. We believe that self-reabsorption of emitted photons prevented radiative recombination lifetime from being observed in transient photoluminescence. The dependence of carrier lifetime on the GR is plotted in figure 3(c).

Figure 4 shows the schematic drawing of the GaAs p-on-n solar cell investigated in this work. The structure was similar to that reported in our previous study [10], except that an AlGaAs back surface field (BSF) layer was employed as a rear hetero barrier instead of a highly doped GaAs layer. The reactor pressure was maintained at 6 kPa, while the growth

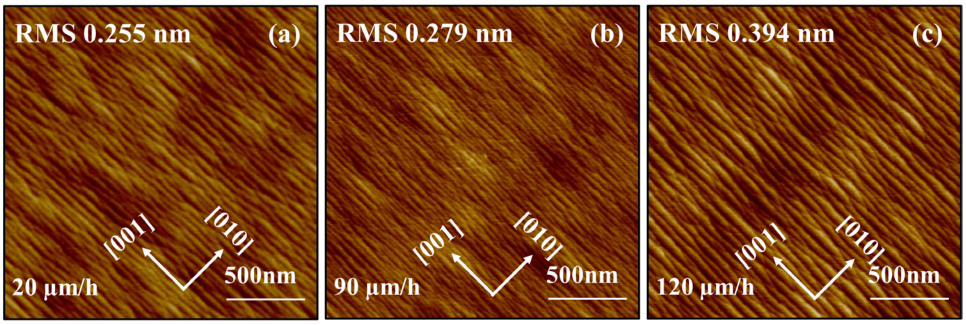


Figure 2. $2 \times 2\ \mu m^2$ AFM images depicting surface morphologies of (a) 20, (b) 90, and (c) 120 μm h^{-1} grown 4 μm-thick undoped GaAs layers.

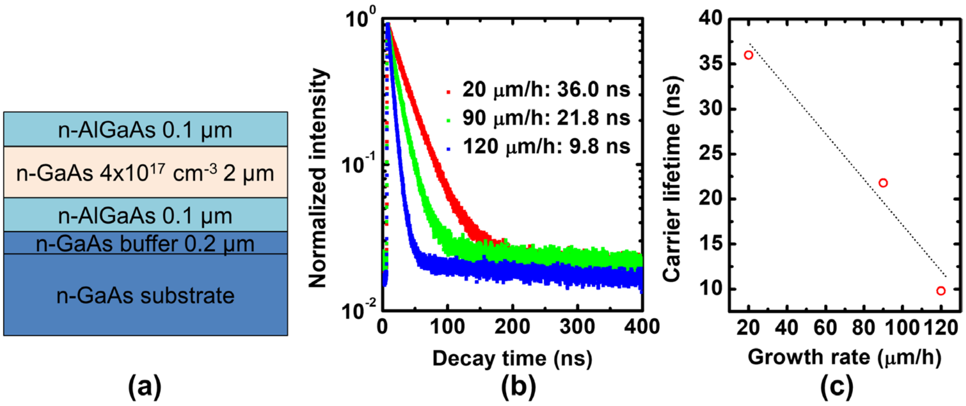
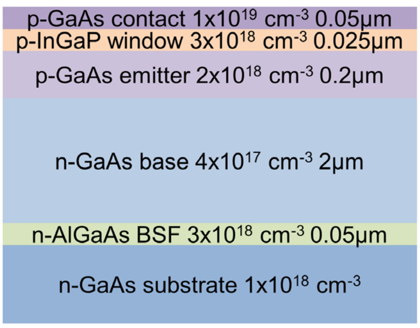


Figure 3. (a) A double-hetero (DH) layer structure for transient PL measurement, (b) PL decay curves for the DH samples with the core n-GaAs grown at GRs of 20, 90 and 120 μm h^{-1}, and (c) minority hole lifetime in n-GaAs as a function of GaAs GR.



Layer	GR. (µm/h)	V/III	T_g (°C)	P_g (kPa)
Contact	5	20	600	6
Window	1.9	100	600	6
Emitter	20	20	680	6
Base	**20/90/120**	20	680	6
BSF	5	40	680	6
Buffer	5	20	680	6

Figure 4. The layer structure of GaAs p-on-n solar cells and the growth conditions of each layer.

temperature was decreased from 680 °C to 600 °C after the growth of the emitter layer for growth of InGaP window and GaAs contact layers. The InGaP window was intentionally Ga-rich in order to decrease light absorption and increase barrier height. Effects of GR on the performance of GaAs solar cell were investigated with the 2 μm-thick n-GaAs base layer grown at various GRs, 20, 90 and 120 μm h^{-1}. The area of GaAs solar cell was approximately 0.043 cm^2, determined by the mesa area. This included 31% of metal coverage, resulting in an effective area of about 0.032 cm^2.

Figures 5(a) and (b) respectively show the current–voltage (I–V) characteristics under AM1.5G (100 mW cm^{-2}) and EQE spectra of GaAs p-on-n solar cells with the n-GaAs base layers grown at various GRs. The surfaces of the cells were with ZnS/SiO_2 ARC. The results here were evaluated using the effective area of 0.032 cm^2, excluding the area covered with the contact metal. A decrease in EQE at long wavelength range, which was brought about by high GRs, evidenced the degradation of photo-carrier correction efficiency. This EQE degradation is responsible for the reduced short circuit current density (J_{sc}) with enhanced GRs, particularly, from 90 to 120 μm h^{-1}. Assuming that the minority hole mobility in n-GaAs with the electron concentration of 4×10^{17} cm^{-3} is comparable to the corresponding majority hole mobility in p-GaAs [17], the minority hole mobility was approximately 210 $cm^2\ V^{-1}\ s^{-1}$ estimated by using the low-field mobility model [18]. Applying this value together with the lifetimes from figure 3(b), the hole diffusion lengths in n-GaAs were calculated to be 4.42, 3.44 and 2.29 μm for GR of 20, 90 and 120 μm h^{-1}, respectively. By this assumption, however, the carrier diffusion lengths

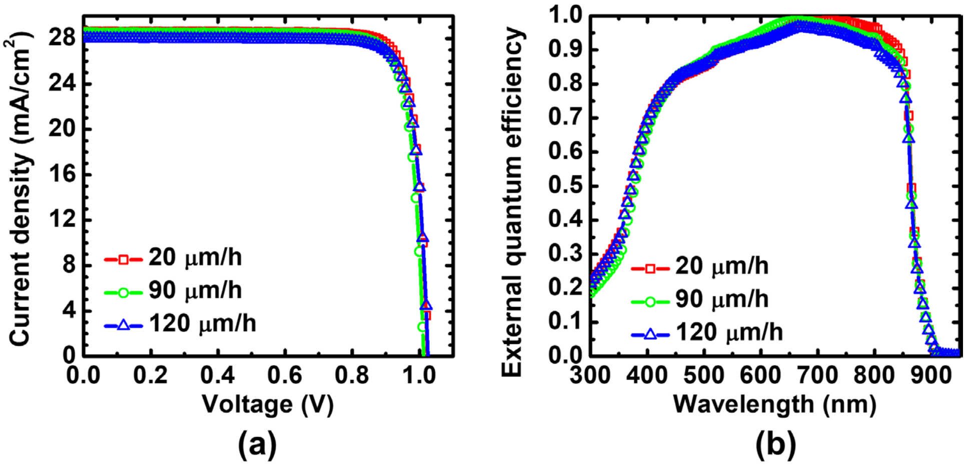


Figure 5. (a) *I–V* characteristics under AM 1.5G illumination and (b) EQE spectra of GaAs solar cells with the n-GaAs base layer grown at 20, 90, and 120 μm h^{-1}. The devices had the effective area of 0.032 cm^2 and evaluated with ZnS/SiO$_2$ ARC.

Table 1. Average values of J_{sc}, V_{oc}, fill factor (FF), and efficiency (η) of the GaAs p-on-n solar cells grown with different GR evaluated using an effective area of 0.032 cm^2.

GR (μm h^{-1})	J_{sc} (mA cm^{-2})	V_{oc} (V)	FF	η (%)
20	28.38	1.023	0.84	24.51
90	28.37	1.013	0.84	24.05
120	27.59	1.023	0.84	23.77

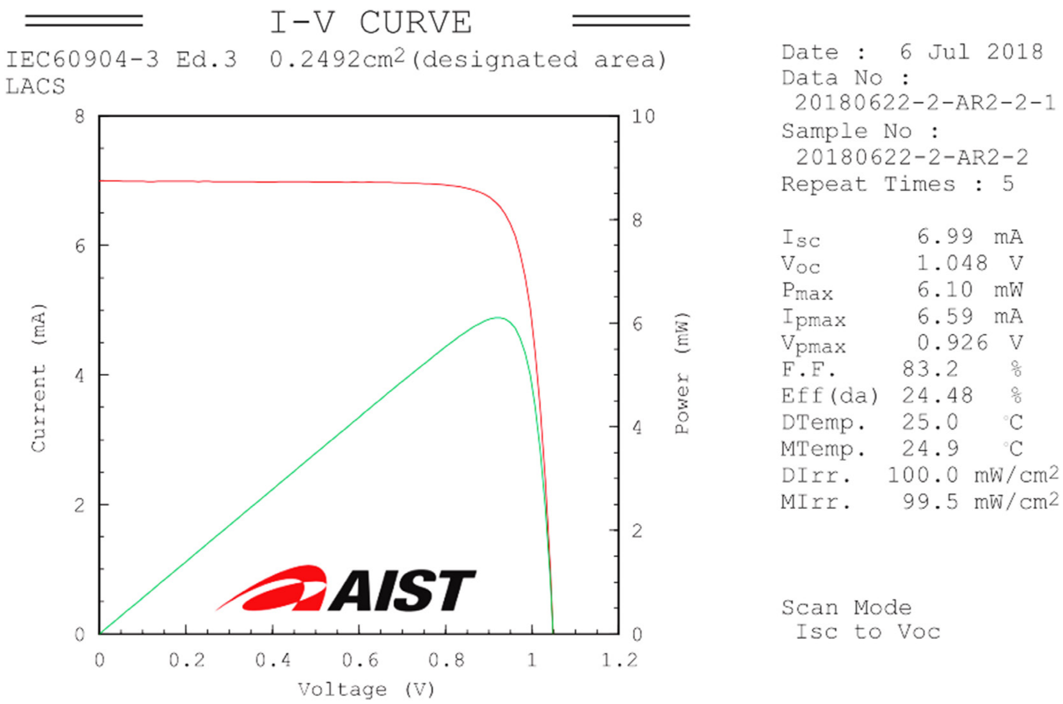


Figure 6. *I–V* characteristic of a 120 μm h^{-1} grown GaAs p-on-n solar cell with optimized fabrication processes certified by Calibration, Standards, and Measurement team of Research Center for Photovoltaics, AIST.

were longer than the thickness of n-GaAs base layer which is not consistent with the EQE results. Therefore, it is reasonable to assume that the minority hole mobility was degraded with enhanced GRs. The background carbon impurity and n-type compensation doping can lead to a faster carrier scattering rate resulting in a shorter diffusion length. Indeed, the carrier diffusion lengths were considered to be less than 2 μm for the GRs of 90 and 120 μm h^{-1} as supported by the decrease in EQE at long wavelength range.

In this work, the open circuit voltage (V_{oc}) seemed to be independent of the GR although the 90 μm h^{-1} grown cell had the lowest value owing to a fabrication failure, i.e. about 8.7 mV decrease in V_{oc} was observed in this sample after ARC deposition. This V_{oc} tolerance is ascribed to both p-on-n cell design and a high doping concentration in the n-type base layer which potentially enhances the radiative recombination under open-circuit condition, but decreases carrier collection efficiency as a drawback. However, typical GaAs p-on-n solar cells grown by MOVPE can achieve a V_{oc} of 1.03–1.06 V [19, 20], higher than the values obtained in this experiment. It is presumable that there were some other factors that limited the cell efficiency in this experiment including the area of the investigated solar cells, in which the effects of the perimeter of the cell mesa is relatively large [21, 22]. Moreover, immaturities in fabrication processes can also be a reason. Table 1 summarizes the average J_{sc}, V_{oc} and conversion efficiency (η) of all GaAs p-on-n solar cells. The efficiency of GaAs p-on-n solar cells, averaged over nine cells, decreased slightly from 24.51% to 23.77% when the GR of the n-GaAs base layer was increased from 20 to 120 μm h^{-1}.

To demonstrate the full potential of the high-speed growth, 120 μm h^{-1}, for high-efficiency GaAs cells, the grid area was expanded to $5 \times 5\,\text{mm}^2$ with only 3% shadow loss. The fabrication procedure was also improved for high efficiency although it follows a standard process of GaAs cell fabrication. The precise *I–V* curve measurements were carried out in the Calibration, Standards, and Measurement team of Research Center for Photovoltaics, AIST. Figure 6 shows the *I–V* characteristic for the GaAs cell with the base layer grown at 120 μm h^{-1}. The layer structure and the growth conditions follow the ones in figure 4. The V_{oc} was increased from 1.023 to 1.048 V while almost no improvement in J_{sc} was obtained compared to the cells with the smaller mesa area. Some possible reasons of this gain in V_{oc} were the larger cell area compared to its perimeter and a decrease in dark current by an improvement of fabrication procedures. The resultant conversion efficiency was 24.48%, which is a remarkable value considering the GR of the base layer exceeding 100 μm h^{-1}.

In conclusion, the GaAs GR could be increased to 120 μm h^{-1} and showed almost no saturation with an increase in TMGa supply. Such fast growth induced lattice imperfection and deteriorated carrier lifetime. Its impact was, however, limited in the performance of GaAs single-junction solar cells and a 24.48% efficiency could be realized for a GaAs solar cell, in with the 2 μm-thick n-type base layer was grown in only 1 min at a GR of 120 μm h^{-1}. This achievement in high-speed epitaxial growth will open a way to the drastic cost reduction in the production cost of III–V photovoltaic cells.

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