Cumhuriyet Sci. J., 46(2) (2025) 390-395
DOI: https://doi.org/10.17776/csj.1672625

Cumhuriyet Science Journal

| csj.cumhuriyet.edu.tr | Founded: 2002 ISSN: 2587-2680 e-ISSN: 2587-246X Publisher: Sivas Cumhuriyet University

MOVPE Growth and Doping Optimization of n- $Al_xGa_{1-x}As$ Layers for Laser Diode Applications

Gamze Yolcu [1, 2, a], İlkay Demir [2, 3, b, *]

[1] *Department of Metallurgical and Materials Engineering, Sivas Cumhuriyet University, 58140 Sivas, Türkiye*

[2] *Sivas Cumhuriyet University Nanophotonics Application and Research Center-CÜNAM, 58140 Sivas, Türkiye*

[3] *Department of Nanotechnology Engineering, Sivas Cumhuriyet University, 58140 Sivas, Türkiye*

**Corresponding author*

Research Article

History
Received: 09/04/2025
Accepted: 16/06/2025

ABSTRACT

Epitaxially grown n-$Al_xGa_{1-x}As$ layers, which form the basis of the modern semiconductor laser structures, play a critical role in both optical and electrical performance of the device. These layers provide electron injection into the active region and at the same time act as optical waveguides, allowing efficient steering of the laser light. Since Al concentration and doping levels have a direct effect on fundamental properties such as band gap, carrier density and resistive losses, it is of great importance to meticulously optimize these parameters. In this study, n-$Al_xGa_{1-x}As$ layers were epitaxially grown on GaAs substrate by MOVPE (Metal Organic Vapor Phase Epitaxy) method, with n-type doping using SiH_4 (silane) precursor. Here we focused on the effects of increasing Al concentration on doping density in the $Al_xGa_{1-x}As$ layers. The obtained results showed that when Al concentration is above 30%, no significant increase in doping density was observed despite the maximization of SiH_4 flow. This phenomenon is thought to be associated with the formation of DX centers (deep donor levels), which become more prominent at high Al concentrations and can trap free electrons, thereby reducing the effective doping efficiency. Furthermore, the data obtained from Hall and ECV (Electrochemical Capacitance Voltage) measurements exhibited good agreement at low Al concentrations, while significant differences were observed for Al fractions above 0.2. This comprehensive analysis reveals the current limitations of epitaxially grown n-$Al_xGa_{1-x}As$ layers and emphasizes the need for precise control of Al concentration, while providing a more in-depth interpretation by systematically comparing the obtained results with the data reported in the literature.

Keywords: MOVPE method, epitaxial growth, carrier density, n $Al_xGa_{1-x}As$ layers, DX center.

[a] gamzeyolcu@cumhuriyet.edu.tr https://orcid.org/0000-0003-0201-8266 [b] idemir@cumhuriyet.edu.tr https://orcid.org/0000-0002-2224-989X

Introduction

$Al_xGa_{1-x}As$-based semiconductor laser diodes have become prominent in various fields such as scientific research, industrial production, medical devices, and imaging technologies, thanks to their high efficiency, compact size, configurable design possibilities, and capability to operate under harsh environmental conditions [1,2]. These diodes are notable for their low operating current requirement and high electrical-to-optical power conversion efficiency. For instance, in communication systems, where high data transmission speeds and stable signal transmission are required, these low-operating-current $Al_xGa_{1-x}As$-based semiconductor laser stand out as a key components and plays a critical role in both communication application and data transfer. Furthermore, since long-term and stable operation of lasers depends directly on the applied electric current, using devices that provide high performance at low pump currents significantly enhances system efficiency. on the other hand, since applications like optical recording (e.g., rewritable CDs), which rely on high light output for fast writing and erasing speeds and high recording density, require high-power laser diodes [3]. This broad spectrum of performance demands underscores the need for precise optimization of structural parameters in laser diode design.

The performance of high-power semiconductor lasers depend directly on the quality of the epitaxially grown semiconductor material [4]. In this context, the n-type $Al_xGa_{1-x}As$ layers in $Al_xGa_{1-x}As$/GaAs laser diode structures represent one of the fundamental components governing the device's electrical and optical characteristics [5]. The n-type $Al_xGa_{1-x}As$ layers enables the efficient operation of the $Al_xGa_{1-x}As$ based-laser diode by providing the transport of negatively charged electrons and plays a critical role in the formation of optical gain at the p–n junction. [6]. Moreover, accurate control of the doping concentration is essential for maintaining the optimal balance between electronic conductivity and optical losses in the n- $Al_xGa_{1-x}As$ based-laser diode structures.

However, the aluminum (Al) concentration in the $Al_xGa_{1-x}As$ layers has a major impact on the laser's optical and electronic properties, which in turn determines the band gap and refractive index profile of the material [7,8]. Careful optimization of the Al content contributes to the improvement of the waveguide mechanism and the increase of laser efficiency. For high-power laser diodes to operate effectively, precise control over doping levels, accurate definition of layer composition, and minimization of impurity concentrations are essential. Additionally, both composition and doping conditions must be dynamically adjusted during the growth process. In this context, optimization of n-type doping and Al concentration is critical for the development of highly efficient and reliable $Al_xGa_{1-x}As$-based laser diodes. Studies in the literature show that adjusting the carrier density above 30% Al concentration poses a significant challenge for researchers.

For instance, Watanabe et al reported that Silicon (Si) donor levels in $Al_xGa_{1-x}As$ layers showed significant changes depending on the material composition and growth conditions. It was determined that the energy levels of DX centers were related to the Fermi level and that these levels changed with the increase in Al concentration. In particular a rapid decrease in the concentration of DX centers was observed in the $X_{Al} < 0.3$ range, while it was revealed that the shallow donor density increased with the decrease of DX centers in the $X_{Al} > 0.5$ region [9].

In another study conducted by Pfeffer et al., $Al_xGa_{1-x}As$ samples with different aluminum contents (x = 0.10, 0.25 and 0.35) were grown and it was revealed that the Hall carrier concentration decreased rapidly as the Al content increased. These observed changes in carrier density were further supported by Secondary Ion Mass Spectrometry (SIMS) and ECV measurements. Although the Hall mobility measurement ranged from 300 to 900 $cm^2/V \cdot s$, this variation can't serve as an indication of the presence of DX centers as suggested by the author Additionally, it was determined that SIMS, ECV and Hall data were consistent for the $X_{Al} = 0.1$ sample; however, significant discrepancies between the measurements emerged for the sample that exhibit Al concentration of $X_{Al} \geq 0.2$.

Although the presence of DX centers could not be definitively confirmed, the observed decrease in carrier density is suggested to be associated with DX centers, consistent with the band structure model proposed by Kuech et al. The findings reveal that n-type doping levels and Al concentration in $Al_xGa_{1-x}As$ systems require more accurate optimization process for the efficiency and stability of high-power semiconductor lasers [10, 11].

To make it clear, the donor ionization energy increases significantly in $Al_xGa_{1-x}As$ compounds when the Al content exceeds 25% [12]. This causes the ionized carrier density to decrease, reducing the doping efficiency. In addition, high Al concentrations, especially at high flow rates of source gases such as TMAl, lead to increased carbon contamination. Since carbon acts as a acceptor, its incorporation further suppresses the free electron concentration in the material. However, high Al content can cause dopant atoms such as Si to change position in the crystal lattice and become passive DX centers. DX centers emerge as a result of the dopant atom forming a complex with a donor-cation vacancy and are not electrically active [13]. Moreover, the tendency of Al atoms to attract impurities such as oxygen into the crystal promotes the formation of DX centers. Oxygen creates deep-level, nonradiative trap centers, causing donor compensation and a reduction in the material's radiative efficiency [14]. Collectively, these factors contribute to an increase in the density and activity of DX centers, which ultimately reduces the free carrier concentration and degrades electrical conductivity.

Materials and Methods

Epitaxial n- $Al_xGa_{1-x}As$ layers were grown on undoped GaAs substrate by AIXTRON 200/4 RF-S horizontal flow, MOVPE. Trimethylaluminum (TMAl), trimethylgallium (TMGa) and arsine (AsH_3) gases were used as Al, Ga and As sources, respectively, while SiH4 gas was used for n-type doping. In situ optical reflection was measured with a monitoring wavelength of 880 nm to control important parameters such as growth rate and surface quality during growth. In order to determine the crystal quality and Al concentrations of the grown films, Rigaku High Resolution X-Ray Diffraction (HRXRD) system was employed. Carrier concentrations were determined using both Hall effect and ECV measurement systems. All the necessary characterizations were performed in detail.

Findings and Discussion

Extensive growth optimizations have been carried out to obtain the desired doping levels in the n-$Al_xGa_{1-x}As$ layer. During these experiments, it was noticed that the doping level decreased with the increase of the Al concentration ratio. Notably, when the Al concentration exceeded 30%, the carrier concentration plateaued around 5×10^{17} cm^{-3} even when the SiH_4 flow was adjusted to the maximum value. Representative results from these optimization experiments are presented in Table 1.

Table 1. Growth parameters and results obtained for the growth of n-$Al_xGa_{1-x}As$ layers for the optimization of Al concentration and doping level.

Sample Name	Growth Temp. (°C)	TMGa (sccm)	TMAl (sccm)	AsH3 (sccm)	SiH4 (sccm)	Al Conc. (%)	Carrier Conc. (cm-3)
Sample A	650	16	23	20	15	0.25	$1.85x10^{18}$
Sample B	650	16	41	20	20	0.37	$3.1x10^{17}$
Sample C	650	16	38	20	31	0.335	$4.9x10^{17}$
Sample D	650	16	23	50	15	0.225	$3.4x10^{18}$
Sample E	700	8	23	20	31	0.27	$7.2x10^{17}$
Sample F	650	10	23	20	15	0.31	$4.21x10^{17}$
Sample G	650	10	23	20	15	0.35	5x1017
Sample H	650	16	36	20	10 - 30	0.35	$4.1x10^{17}$

For the Samples A, B, and C, in which only the TMAl and SiH_4 flows were changed, it was noticed that as the Al concentration increased, carrier density decreased even though the SiH_4 flow was increased. In order to investigate the effect of increasing the As flow, another experiment was preformed taking Sample A as reference in this experiment, indicated by sampled D, the AsH_3 flow was increased from 20 sccm to 50 sccm. The results showed that although the carrier density increased in Sample D, the Al concentration decreased from 0.25 to 0.22.

Another experiment, referred to as Sample E, was conducted to gain clearer insight into the doping mechanism at high aluminum concentrations. The objective was to increase the Al content by reducing the TMGa and AsH_3 flow, while simultaneously enhancing the carrier density by raising the growth temperature and increasing the SiH_4 flow rate. However, carrier density could not be increased up to $1x10^{18}$ cm^{-3} levels.

In Sample F growth, unlike Sample A, TMGa flow was decreased, and reactor pressure was increased from 100 mbar to 250 mbar. In this growth, carrier density could not be increased even though Al concentration was increased. It is clearly seen from the Figure 1 that the dopant density decreases rapidly with increasing Al concentration and after 30% Al concentration the carrier density was always around $5x10^{17}$ cm^{-3}. When literature studies are examined, it is observed that researchers encounter the same problem. In $Al_xGa_{1-x}As$ layers when $x > 0.25$, it is thought that the increase in donor ionization energy, carbon contamination, deep trap levels originating from oxygen and the formation of DX centers cause a significant decrease in the electron carrier concentration. The increase in donor ionization energy leads to a decrease in the free electron density, while the high TMAl flow rate suppresses electron carriers by increasing carbon contamination. In addition, the interaction of oxygen and aluminum creates deep trap levels, increasing the electrical resistance of the material and decreasing its conductivity. DX centers that occur at $x > 0.2$ levels restrict electron mobility due to their high activation energies and further reduce the conductivity [14].

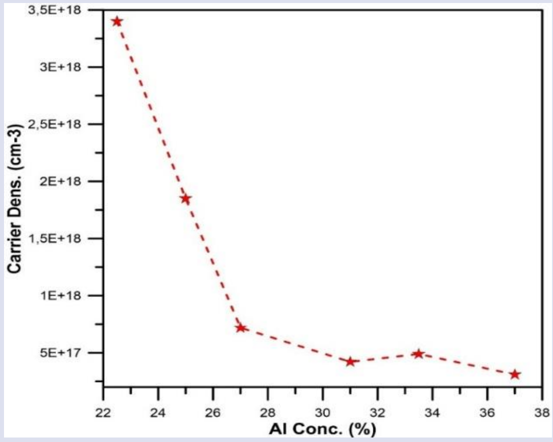

Figure 1. Variation of carrier density in n-$Al_xGa_{1-x}As$ layer versus Al concentration

In order to obtain more comprehensive information about the crystal structure and material quality for these samples, XRD measurements were performed conducted using θ–2θ scans. The Al composition, FWHM (full width-half maximum) values and lattice parameters of each sample were determined using the Global Fit simulation program [15]. Based on the obtained lattice constants, the reference lattice constants a_0 values were calculated with the help of Vegard's law [16]. In this way, the strain in the samples were also calculated and all obtained values are presented in Table II.

Based on the obtained data, the crystal quality and lattice mismatch of the samples were comparatively evaluated depending on FWHM and strain values [17, 18]. Although the FWHM and strain values of the samples are relatively close to each other, Sample B exhibits the highest crystal quality with the lowest FWHM value (113.6 arcsec), while also having the highest strain ($4.50×10^{-4}$), indicating the greatest lattice mismatch. In contrast, Sample D shows the highest FWHM value (231.7 arcsec) and the lowest strain ($1.35×10^{-4}$), suggesting better lattice compatibility despite the presence of more structural defects. These findings suggest that the relationship between FWHM and strain may not be strictly linear or directly correlated.

Table 2. Measured FWHM and Calculated Strain Values of the $Al_xGa_{1-x}As$ Layers

Sample Name	Al Conc. (%)	FWHM (arscec)	a_0 (Å)	$a_{measurement}$ (Å)	Strain (%)
Sample A	0.25	181.1	5.65532	5.65648	2.05×10^{-4}
Sample B	0.37	113.6	5.65629	5.65884	4.50×10^{-4}
Sample C	0.335	158.6	5.65601	5.65808	3.66×10^{-4}
Sample D	0.225	231.7	5.65512	5.65588	1.35×10^{-4}
Sample E	0.27	149.8	5.65548	5.65696	2.62×10^{-4}
Sample F	0.31	154.4	5.65581	5.65716	2.39×10^{-4}

As a last experiment Sample F was taken as reference, and a new experiment was conducted and the reactor pressure was set to 100 mbar again resulting in Sample G. Al concentration increased from 31% to 35% and carrier density increased from 4.20×10^{17} to 5×10^{17} cm^{-3} (Hall measurement). ECV measurement of Sample G was contacted and it was noticed that the carrier density was in the range of 2×10^{18} cm^{-3} (Figure 2). In the studies conducted in the literature, when the Al ratio was around 0.1, a good agreement was observed between ECV and Hall data, but when the Al ratio was greater than 0.2, a serious inconsistency was noticed between the measurements [19].

In the studies conducted on Si-doped $Al_xGa_{1-x}As$, it was reported that there were two types of donor centers: deep and shallow [10]. In general, when measuring n-type materials with ECV, unlike p-type materials, the UV light source used allows the detection of deep donor levels that cannot be detected by Hall measurement. Since all Si atoms are electrically active, ECV measurement was taken into account for n-type layers.

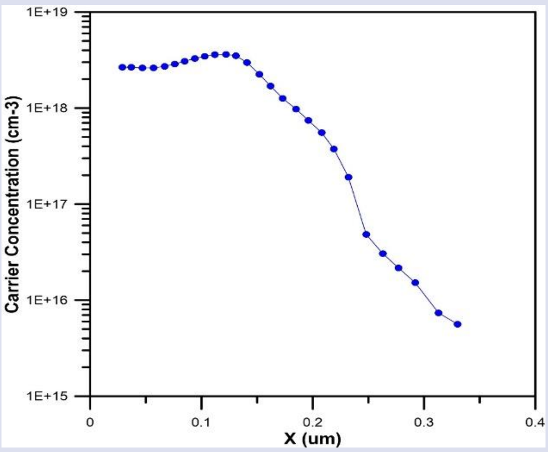

Figure 2. Sample G is the ECV measurement result of the sample

Although an inconsistency was noticed in ECV and Hall measurements in n-$Al_xGa_{1-x}As$ samples, it was also proven by ECV measurements that when the Al concentration exceeded 30%, the carrier density remained constant and could not be increased. The structure of Sample H and ECV measurement are given below (Figure 3). In Sample H, $Al_xGa_{1-x}As$ with 35% Al concentration was grown and the SiH_4 flow was changed as 10, 15, 20, 25, 30, and 35 sccm. The change in carrier density with increasing the SiH_4 flow was investigated. ECV measurement of Sample H is given in Figure 4. Although the SiH_4 flow was increased, the carrier density of the $Al_xGa_{1-x}As$ layer with 35% Al concentration remained constant at 1×10^{18} cm^{-3} levels.

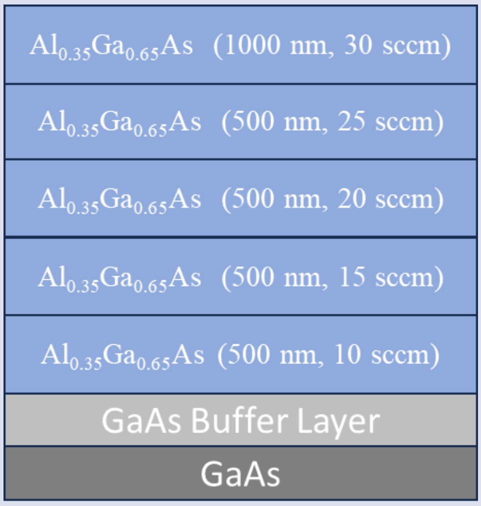

Figure 3. Structure of the Sample H

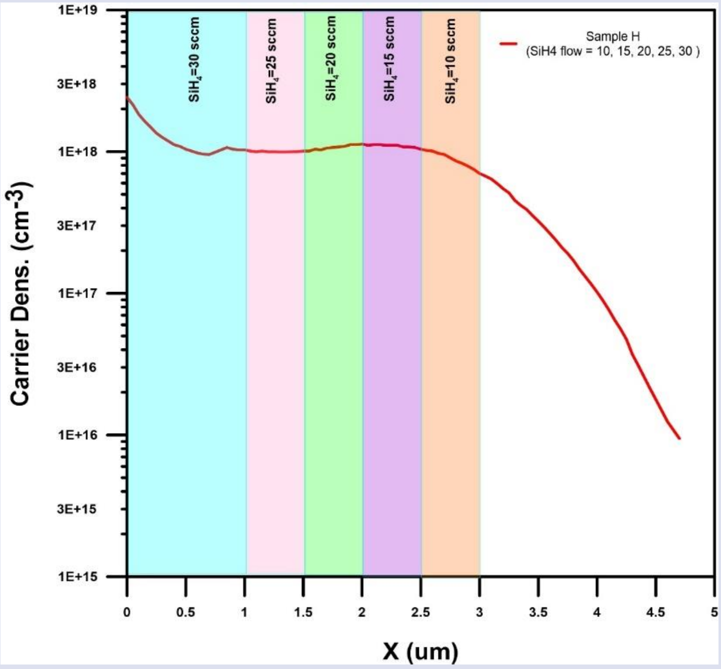

Figure 4. ECV measurement result of Sample G

Conclusions

In conclusion, the optimization of doping density and Al concentration on epitaxially grown n-$Al_xGa_{1-x}As$ layers in this study clearly revealed the main challenges encountered in the production of high-power semiconductor laser diodes. Experimental data showed that when the Al concentration exceeds 30%, the expected increase in doping density could not be achieved despite the adjustment of the SiH_4 flux to the maximum level. While the agreement between Hall and ECV measurements was achieved at low Al ratios, significant differences occurred above 0.2 Al ratio. This situation indicates that the current limitations in n-type doping processes and precise control of Al concentration should be reviewed. The obtained results indicate that the process parameters should be re-evaluated for the development of high efficiency and reliable laser diodes and provide a solid basis for further research by providing data consistent with similar studies reported in the literature.

Conflicts of interest

There are no conflicts of interest in this work.

Acknowledgement

This work is based on results obtained during the ongoing Ph.D. thesis of the first author, titled "Development of a GaAs-based pump laser diode structure (λ = 88x nm)". Gamze YOLCU is supported by the YÖK (Council of Higher Education) 100/2000 Ph.D. Scholarship Program and The Scientific and Technological Research Council of Turkey (TUBITAK) 2211/A Program. The authors acknowledge the usage of the Sivas Cumhuriyet University Nanophotonics Application and Research Center-CUNAM facilities. This study supported by The Scientific Research Project Fund of Sivas Cumhuriyet University, Turkey under the Project number M-2023-857.

References

[1] Ladugin M.A.E., Marmalyuk A.A.E., Effect of (Al)GaAs/AlGaAs quantum confinement region parameters on the threshold current density of laser diodes, *Quantum Electron.*, 49 (6) (2019) 529.

[2] Saeed S., Usman M., Ali S., Ali H., Mustafa L., High performance near-infrared III-Arsenide laser diodes with p-AlGaAs barriers, *Opt. Mater.*, 139 (2023) 113809.

[3] Kim K.C., Kim T.G., Sung Y.M., Choi Y.C., Park Y.J., Han I.K., Park J.I., Performance improvement of high-power AlGaAs lasers, *J. Korean Phys. Soc.*, 47 (2005) S572.

[4] Zappe H., *Laser Diode Microsystems*, Springer Science & Business Media, (2004).

[5] Knauer A., Wenzel H., Erbert G., Sumpf B., Weyers M., Influence of oxygen in AlGaAs-based laser structures with Al-free active region on device properties, *J. Electron. Mater.*, 30 (2001) 1421–1424.

[6] Ito M., Kimura T., Carrier density dependence of refractive index in AlGaAs semiconductor lasers, *IEEE J. Quantum Electron.*, 16 (9) (1980) 910–911.

[7] Perkitel I., Kekül R., Altuntas I., Gür E., Demir I., Influence of Highly Efficient Carbon Doping on $Al_xGa_{1-x}As$ Layers with Different Al Compositions (x) Grown by MOVPE, *J. Electron. Mater.*, 52 (9) (2023) 6042–6051.

[8] Gullu H.H., Yıldız D.E., Yıldırım M., Demir I., Altuntas I., Electrical characteristics of Al/AlGaAs/GaAs diode with high-Al concentration at the interface, *J. Mater. Sci.: Mater. Electron.*, 35 (2) (2024) 189.

[9] Watanabe M.O., Morizuka K., Mashita M., Ashizawa Y., Zohta Y., Donor levels in Si-doped AlGaAs grown by MBE, *Jpn. J. Appl. Phys.*, 23 (2A) (1984) L103.

[10] Pfeffer T.L., et al., The use of diethylsulphide for the doping of $Al_xGa_{1-x}As$ grown by chemical beam epitaxy, *J. Cryst. Growth*, 146 (1–4) (1995) 399–403.
[11] Kuech T.F., Tischler M.A., Potemski R., Cardone F., Scilla G., Doping and dopant behavior in (Al, Ga)As grown by metalorganic vapor phase epitaxy, *J. Cryst. Growth*, 98 (1–2) (1989) 174–187.
[12] Ishikawa T., Saito J., Sasa S., Hiyamizu S., Electrical properties of Si-doped $Al_xGa_{1-x}As$ layers grown by MBE, *Jpn. J. Appl. Phys.*, 21 (11A) (1982) L675.
[13] Oh-hori T., Itoh H., Tanaka H., Kasai K., Takikawa M., Komeno J., Donor–cation vacancy complex in Si-doped AlGaAs grown by metalorganic chemical vapor deposition, *J. Appl. Phys.*, 61 (9) (1987) 4603–4605.
[14] Shieh H.M., Wu T.S., Hsu W.C., SiH_4-doped AlGaAs epilayers formed by MOCVD, *J. Cryst. Growth*, 121 (4) (1992) 665–670.
[15] Kalyon G., Mutlu S., Kuruoğlu F., Perkitel İ., Demir İ., Erol A., InGaAs-based Gunn light emitting diode, *Mater. Sci. Semicond. Process.*, 159 (2023) 107389.
[16] Kaynar E., Sayrac M., Altuntaş İ., Demir İ., Determination of optical properties of MOVPE-grown $In_xGa_{1-x}As$/InP epitaxial structures by spectroscopic ellipsometry, *Braz. J. Phys.*, 52 (5) (2022) 184.
[17] Perkitel İ., Altuntaş İ., Demir İ., The effect of Si (111) substrate surface cleaning on growth rate and crystal quality of MOVPE grown AlN, *Gazi Univ. J. Sci.*, 35 (1) (2022) 281–291.
[18] Pürlü K.M., Koçak M.N., Yolcu G., Perkitel İ., Altuntaş İ., Demir İ., Growth and characterization of PALE Si-doped AlN on sapphire substrate by MOVPE, *Mater. Sci. Semicond. Process.*, 142 (2022) 106464.
[19] Nanometrics, ECVPro: Electrochemical C-V Profiler User Manual, P/N: 9PROF-UM01 Rev. B.