Role of Ion Beam Irradiation and Annealing Effect on the Deposition of AlON Nanolayers by Using Plasma Focus Device

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Abstract AlON nanolayers are synthesized on Al substrate by the irradiation of energetic nitrogen ions using plasma focusing. Samples are exposed to multiple (5, 10, 15, 20 and 25) focus shots. Ion energy and ion number density range from 80 keV to 1.4 MeV and 5.6×1019 m−3 to 1.3×1019 m−3, respectively. Moreover, the effect of continuous annealing (473 K and 523 K) on an AlN surface layer synthesized with 25 focus shots is also examined. The main features of the X-ray diffraction (XRD) patterns with increasing focus shots are: (i) variation in the crystallinity of AlN along (111), (200) and (311) planes, (ii) increasing average crystallite size of AlN (111) plane, and (iii) stress relaxation observed in AlN (111) and (200) planes. The crystallinity of AlN surface layer is comparatively better at 473 K annealing temperature. A broadened diffraction peak related to an aluminium oxide phase showing weak crystallinity is observed for 15 focus shots while non-bounded oxides are present in all other deposited layers. Raman and Fourier transform infrared spectroscopy (FTIR) analysis confirm the presence of AlN and AlOx while non-bounded oxides are present in all other deposited layers. Raman and Fourier transform infrared spectroscopy (FTIR) analysis confirm the presence of AlN and AlOx while non-bounded oxides are present in all other deposited layers. Raman analysis shows that the overlapping of AlN and Al2O3 results in the development of residual stresses. Scanning electron microscope (SEM) results demonstrate that the formation of rounded grains (range from 20 nm to 200 nm) and variations in their microstructures features depend on the increasing number of focus shots. Decomposition of larger clusters into smaller ones is observed.

Keywords: characterization, XRD, focus shots, X-ray diffraction, nucleation, surface layer

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1 Introduction

Nanocrystalline AlN films are currently a subject of great interest due to their novel industrial applications. Owing to wide band gap, low electrical but high thermal conductivity, high optical transmission, and high decomposition temperature, AlN films are used in the microelectronic industry [1−3]. Due to good piezoelectric properties with high acoustic velocity [4−7], AlN films are applied to the fabrication of acoustic devices. However, the performance of acoustic devices depends strongly on crystal orientation and surface roughness. Researchers around the world are depositing AlN films by using various techniques like radio frequency magnetron sputtering [8,9], chemical vapor deposition [10], and reactive molecular beam epitaxy [11].

A plasma focus (PF) is a low energy cost effective device that utilizes a self-generated magnetic field to compress the plasma up to high density (1025−1026 m−3) and high temperature (1−2 keV) for a short time (∼10−7 s) [12]. It is a potential candidate for soft/hard X-rays, neutrons, energetic ions, and relativistic electrons [13−15]. Energetic ions have been used for thin film deposition [16], ion implantation [17], nanostructuring of magnetic thin films [18,19], surface modification of materials [20], thermal surface treatment [21], and deposition of fullerene films on silicon [22]. Films deposited by PF devices show superior surface qualities, such as good adhesion to substrate surface, grain growth of different compounds through nucleation by successive focus shots at room temperature [23], and shorter nitriding time in comparison to other techniques [24,25]. Thus, PF devices have advantages over conventionally used high temperature techniques for thin film deposition because of their simplicity, cost effectiveness, and the shorter deposition time required to obtain good adhesive films.

The present work reports the crystalline structure, phase identifications, surface morphology, and vibrational and phonon modes of AlN surface layer synthesized on Al substrates by the irradiation of energetic nitrogen ions emanated during the radial collapse phase of PF device. The film deposited for 25 focus shots is called as-deposited surface layer, which is used to further study the annealing effect on AlN surface layer.

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2 Experimental details

AlN surface layers are synthesized on Al substrate by nitrogen ion irradiation by using a low energy PF device \cite{12,14,15,17,19}, powered by single Maxwell 30 µF, 15 kV fast discharging capacitor with maximum storage energy of 3.3 kJ. The schematic diagram of the PF device is shown in Fig. 1. The electrode system comprises a hollow copper anode surrounded by six copper rods which serve as the cathode. A Pyrex glass insulator sleeve is placed between the electrodes to facilitate the formation of current sheath. The electrode assembly is housed in a stainless steel vacuum chamber, which is evacuated by a turbo pump attached with rotary vane pump down to the pressure of 10\(^{-4}\) mbar and is then filled with nitrogen gas at an optimum pressure of 1.25 mbar. The details of the PF device can be found elsewhere \cite{12}. A BPX65 photodiode detector placed 10 cm from the anode tip is used to characterize the nitrogen ions emitted during one focus shot. The details of the BPX65 photodiode detector, along with a parametric study of nitrogen ions, are described in Ref. [26]. Al samples are prepared in the form of a disk of 10 mm in diameter and 3.5 mm thickness.

![Fig.1 Schematic diagram of the plasma focus device](image1)

The substrate surface is polished with SiC abrasive paper and then cleaned ultrasonically in de-ionized water for 15 minutes. Samples are mounted at 10 cm along the anode axis (at 0\(^\circ\) angular position) and are exposed to multiple focus shots (5, 10, 15 and 20). A stainless steel shutter is arranged to avoid sample exposure during a few weak focus shots. An aperture assembly was used to stop any copper debris that may be ablated from rim of hollow copper anode. Different surface properties of AlN surface layer synthesized with multiple focus shots are examined by using X-ray diffractometer (XRD), scanning electron microscope (SEM), Raman spectroscopy (RS), and Fourier transform infrared spectroscopy (FTIR). Additionally, the annealing effect (for 1 h in air) on the crystallinity and surface morphology of the as-deposited surface layer is also examined.

3 Results and discussions

3.1 Characterization of ion beam and growth of AlN surface layer

The energetic nitrogen ions emanated during the radial collapse phase of PF operation are characterized by employing BPX65 photodiode detector and then used to deposit AlN films. Fig. 2 illustrates a typical ion beam signal recorded with a BPX65 photodiode detector (placed 10 cm from the front of the anode) along with a high voltage signal. The protective glass of the BPX65 photodiode is removed to enable the ions and soft X-rays photons to reach its active area. The photodiode is covered with a foil of 10 µm thickness having an aperture of diameter 600 µm with an area of 0.28 mm\(^2\) to limit the ion beam flux striking the detection area of the photodiode. The ion induced electrical signal is transmitted to the oscilloscope through 50 Ω coaxial transmission line. The dotted line represents the high voltage signal while the solid line indicates the photodiode signal. The first peak in the diode signal is normally referred to as the photopeak and is attributed to the signal to the photons from the pinch plasma column while the subsequent delayed peak(s) are attributed to the ions that enter into the detector. The photopeak serves as the reference point at which the ions of all different energies are assumed to be emitted simultaneously.

![Fig.2 Typical ion beam signals recorded by a BPX65 photodiode detector and high voltage probe](image2)

The time of flight technique (TOF) is used to estimate the ion parameters. The velocity of nitrogen ions is calculated by taking the ratio of the distance (from the ion source to the substrate surface) to the flight time of ions from the source to the detector. The flight time of ions is estimated using the photopeak as the reference point. The estimated ions velocity (v), the detector distance (10 cm) divided by the time of flight, is used to calculate the energy of the ions reaching the detector at various times by the relation \(E = \frac{1}{2}mv^2\), where m is
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the atomic mass of nitrogen ions. The ion number density \(N_\text{i}\), at a particular time instant corresponding to ions of particular energy, is estimated using the relation
\[
N_\text{i} = \frac{V}{qA},
\]
where \(v\) and \(q\) are the velocity and charge of nitrogen ions and \(A\) is the area of the aperture whereas \(V\) is the voltage of the ion pulse developed across the resistor \(R\) at that instant. The estimated ions energy, by employing the above mentioned equations, ranges from 80 keV to 1.4 MeV and the ion number density of ion beam pencil at the entrance hole of the detector is respectively from 5.6×10^{19} \text{m}^{-3} to 1.3×10^{20} \text{m}^{-3} corresponding to the ion energy value. It may be noted that the ion number densities mentioned above are for ions of specific energy values at the 600 \(\mu\text{m}\) diameter entrance hole of the detector. A simple estimate shows that at each energy value there are about 10^{12} ions in the part of the ion beam pencil inside the detector and hence summing over the entire energy range the number of ions along the anode axis that move through the entrance hole of the detector is about 10^{15}, similar to the results obtained by others [27~29]. It may be mentioned that the estimates presented here are based on some simple assumptions and simple analysis; for the detailed ion emission characteristics of PF device, readers are advised to refer to the recent work by LEE and SAW [27]. The ions having energy and number density in the above-mentioned range are used to deposit AlN films. It is known that ions emitted from the focused plasma column are in a fountain like geometry with anisotropy in their angular distribution. Most of the ions are emitted in a small solid angle along the anode axis and their flux decreases with increasing angle [30].

The growth mechanism of an aluminium oxy-nitride surface layer on Al substrate by using PF device can be attributed to the combination of two mechanisms: (i) nitrogen ion implantation into the aluminium substrate (incorporation of nitrogen ions into the Al lattice interstitially and ion induced collision cascades creating point defects and vacancies that result in distortion of the Al lattice); and (ii) reaction of background nitrogen ions (coming from the next bunches of the same focus shot) with Al ions that might be ablated by energetic nitrogen ions from the Al substrate to form AlN that may redeposit onto the Al substrate. It is, however, extremely difficult to say which of the two mechanisms is dominant in the formation of AlN surface layers. Moreover, during annealing in open air, oxide also react with Al ions to form an aluminium oxide phase. Thus, an aluminium oxy-nitride surface layer is formed.

3.2 XRD analysis

Fig. 3 exhibits the XRD patterns of Al and AlN surface layers deposited with multiple (5, 10, 15, 20 and 25) focus shots. For 5 focus shots, XRD patterns [Fig. 3 (a)~(c)] show the emergence of AlN (111), AlN (200) and AlN (311) planes [Ref. code 00-046-1200] at 2θ values of 38.55°, 44.8° and 78.3°, respectively. The XRD results show that the peak intensity of AlN (200) and AlN (311) planes increases up to 20 focus shots, while it increases for AlN (111) plane up to 10 focus shots, and then it starts to decrease for 15 and 20 focus shots, after that, it starts to increase again for 25 focus shots. However, a weak peak intensity of AlN (311) plane is observed for 25 focus shots. This indicates that recrystallization and re-arrangement of atoms along AlN (311) plane are observed with increasing focus shots. Obviously, for 25 focus shots, the crystallinity of AlN (111) and AlN (200) planes is more than the crystallinity of AlN (311) plane. It is well known that peak intensity of any compound is directly associated with its crystallinity or crystal growth. Thus, the crystal growth behavior of AlN surface layer along different planes depends on the number of focus shots. A small shoulder peak (related to an Al2O3 phase, appeared at 2θ values of 78.5°) [Ref. code 01-073-1199] (for 15 focus shots) confirms the presence of oxides in the AlN surface layer. The development of this phase is due to native oxides being developed during the deposition process. It is clear that the aluminium oxide phase is comparatively more crystalline for the layer formed for 15 focus shots, while peaks related to aluminium oxide for other focus shots are broader, showing poor crystallinities. However, oxides are present in the deposited layers both in crystalline and amorphous forms depending on the number of focus shots. Thus the appearance of AlN and Al2O3 phases confirms the deposition of AlON nanolayers. Moreover, the peak broadening indicates the formation of nanocrystallites, and the peak broadening varies with increasing focus shots. Thus, the incorporation of native oxides is responsible for the creation of micro-strains and vacancies, resulting in the decrease of peak intensity of AlN (311) plane.

The crystallite size of different planes plays a vital role in the mechanical and other surface properties of the materials. It is well known that crystallite size is inversely related with the full width half maximum of the diffraction peaks. Thus, the average crystallite size of AlN (111) plane is evaluated by using the Scherrer formula [31]

\[
\text{Crystallite Size} = \frac{k\lambda}{\beta \cos \theta},
\]

where \(k = 0.99\), \(\lambda = 1.54\ \text{Å}\) is the wavelength of the X-rays source, \(\beta\) is the full width half maxima and \(\theta\) is the Bragg’s angle. Fig. 4 depicts the average crystallite size of AlN (111) plane as a function of focus shots. It is found that the average crystallite size increases from 52±2 nm to 116±4 nm with the increase of focus shots.

This increase in crystallite size is due to the increasing rapid thermal annealing of the sample surface irradiated during energetic ion bombardment. However, rapid thermal annealing is related with ion energy flux, which is attributed to the increasing focus shots. The stress relaxations (Fig. 5) observed in AlN (111) and AlN (200) planes is responsible for the increase of the average crystallite size, which results in improved crystallinity.
The XRD data is also employed to determine the residual stresses present in the deposited aluminium nitride/oxide surface layers. Up and down shifting in diffraction peaks from their corresponding stress-free data indicates the presence of residual stresses (compressive and tensile). Nitrogen ion implantation in the interstitial positions of Al and AlN matrix generates lattice distortion (strain) and defects (creation of vacancies), resulting in residual stresses. The incorporations of native oxide are also responsible for the development of residual stresses. Compressive stresses are generally developed due to ion implantation while tensile stresses are caused by thermal shocks developed during ion implantation\cite{32}. Residual stresses present in AlN (111) and (200) planes can be deduced by multiplying strain ($\Delta d/d$) with the elastic constant of AlN. The strain developed in AlN (111) and (200) plane is evaluated using the formula given below\cite{33}

$$\frac{\Delta d}{d} = \frac{d(\text{observed}) - d(\text{PDF})}{d(\text{PDF})},$$

where ($\Delta d/d$) indicates the strain developed in the AlN surface layer.

Fig. 5 shows the residual stresses developed in AlN (200) and AlN (111) planes as a function of focus shots. Compressive stress ($-0.05\pm 0.014$ GPa) is observed in the AlN (200) plane for 5 focus shots, and increases gradually ($-0.169\pm 0.012$ GPa) up to 15 focus shots and then starts to decrease for 20 and more focus shots of ion irradiations. It is clear that compressive stresses observed in AlN (111) plane are transformed into tensile stresses when ion doses increases from 5 to 10 focus shots. This result is consistent with the results given by CHIEH et al.\cite{34}. They reported that compressive stress first increases up to temperature (400$^\circ$C) and then decreases with the further increase in temperature and transforms to tensile stress at higher temperatures.

In the current experiment, the temperature of the irradiated samples is associated with ion energy flux. The total number of incorporated ions increases with increasing focus shots, resulting in the increase of sample surface temperature. It is clear that an increasing trend in both types of stresses is observed and a stress relaxation is observed with further increase of focus shots. Thus, for lower temperature (fewer focus shots), more diffusion of species takes place, resulting in more lattice distortion and point defects which increases the stresses. For higher focus shots, when the sample surface temperature becomes significant, a
stress relaxation is observed which is due to rapid thermal annealing. However, rapid thermal annealing is linked with the ion irradiation process and is responsible for the enhancement of the crystal growth, resulting in the increase of crystallite size (as previously discussed) of various diffraction planes. Increasing focus shot irradiation thus acts in a similar way as increasing annealing temperature. Hence the ions irradiation provides a mechanism equivalent to thermal annealing although on much shorter time scale.

Now we investigate the annealing effect on the crystallinity of the as-deposited AlN surface layer for two different annealing temperatures (473 K and 523 K) in open air for 1 h. Fig. 6 exhibits the XRD patterns obtained for: (i) unexposed aluminium, (ii) an as-deposited AlN surface layer, and (iii) an AlN layer annealed at 473 K and 523 K. The peak intensity of AlN (111), AlN (200) and AlN (311) planes increases for lower annealing temperatures. A new diffraction plane (220) of AlN appears at 2θ values of 65.17° which is comparatively more crystalline. Additionally, a diffraction peak related to Al overlapping with AlN (111) confirms the formation of a solid solution which is due to the incorporation of native amorphous oxides since no diffraction of oxide appears in the XRD pattern hindering the growth of Al and AlN separately. Moreover, after annealing, diffraction peaks are still shifted from their stress-free values, thereby indicating that residual stresses are present even in the annealed surface layer. Thus, stresses produced during ion irradiations could not be removed at this annealing temperature.

A decrease in the crystallinity of AlN (111) and (311) diffraction planes is observed for higher annealing temperature. The higher annealing temperature leads to re-crystallization which is due to more diffusion of native amorphous oxides. However, the increase in the crystallinity of AlN (200) diffraction plane may be another reason causing the aforementioned decrease. Additionally, overlapping between Al and AlN (111) diffraction planes is still present, resulting in the formation of their solid solution. This overlapping is responsible for the shift of the diffraction planes from their standard values and causes the development of stresses. Moreover, AlN (220) diffraction plane formed at lower annealing temperature vanishes in the case of higher annealing temperature. Thus, AlN (220) is stable for lower annealing temperature because all the diffraction planes appearing during ion irradiation process are still present. It is concluded that AlN diffraction planes formed during the ion irradiation process are more stable even at higher annealing temperature and have cubic structures. PARK et al.\textsuperscript{[35]} have reported that the peak intensity of AlN/CrN diffraction planes is better at 700 °C and decreases with a further increase of annealing temperature. In the present work, the crystallinity of AlN diffraction planes obtained at lower annealing temperature is greater than the crystallinity of AlN diffraction planes observed by PARK et al.\textsuperscript{[35]}. Thus, it is concluded that PF assisted AlN surface layers show better crystallinity at lower annealing temperature as compared to the crystallinity of AlN surface layers reported in Ref. [35], which is due to the transient surface annealing during successive focus shot irradiation prior to continuous conventional annealing.

Fig. 6 XRD patterns of the as-deposited AlN and AlN layers annealed at (i) 473 K and (ii) 523 K

Moreover, the XRD patterns show that there is no diffraction plane of aluminium oxide but the overlapping and shifting of diffraction planes confirms the presence of amorphous oxides which come from the open air during annealing. It is clear that these oxides do not have sufficient energy to react with aluminium and nitrogen to form their respective oxide.

Fig. 7 exhibits the influence of annealing temperature on stresses developed during the ion irradiation process. Tensile stress (0.06±0.015 GPa) observed in the as-deposited surface layer increases slowly for 473 K while it increases rapidly (1.0±0.019 GPa) for 523 K annealing temperature. The increasing trend in tensile stress with increasing annealing temperature is due
to more diffusion of amorphous oxides. Moreover, the growth of different phases is linked with the texture coefficient (TC), which is evaluated for AlN (200) diffraction plane by the following relation [36].

$$TC_{(hkl)} = \frac{I_{(hkl)}}{I_0} \times \frac{\sum{Z_{(hkl)}}}{N},$$

where $I$ is the measured intensity, $I_0$ is the standard intensity and $N$ is the number of diffraction planes of AlN.

The TC of AlN (200) plane as a function of annealing temperature is shown in Fig. 8. It is obvious that the TC of AlN (200) plane first decreases (≈1.4) for 473 K and then increases (≈1.7) for 523 K annealing temperature. For lower annealing temperature, the TC of AlN (200) plane decreases, which is due to the increase in crystallinity of AlN (111) and AlN (311) planes. The appearance of AlN (220) plane may be the other reason. For higher annealing temperature, the TC of AlN (200) plane increases, which is due to the decrease in the crystallinity of AlN (111) and AlN (311) planes; however, the disappearance of AlN (220) plane may be another reason. Moreover, the adatom mobility of AlN increases after the energy absorption by the substrate during continuous annealing. This increase in adatom mobility overcomes the barrier and causes rearrangement of the atoms of AlN, which results in its crystal growth. Adatom mobility is related with surface free energy, strain energy, and interfacial energy. Moreover, the growth of AlN (200) plane is governed by surface free energy [34,37]. It is inferred that at lower annealing temperature, adatom mobility is sufficient to grow the crystal along the (111) and (311) orientations, which hinders the growth of AlN along (200) orientation. For higher annealing temperature, adatom mobility is sufficient to grow the crystal along (200) orientation of AlN, which hinders the crystal growth of AlN along the (111) and (311) orientations. Thus, it is concluded that higher annealing temperature is required to decrease the surface free energy and thereby results in the crystal growth of AlN along (200) orientation.

Now, we further investigate the deposited surface layer annealed at 473 K temperature (due to its better crystallinity) for the purpose of confirming the formation of AlN and presence of oxide via Raman and FTIR techniques.

### 3.3 Raman analysis

Raman analysis is carried out to provide an additional assessment of the structural quality and to confirm the presence of oxides in the annealed AlN surface layer. Raman analysis of AlN surface layer annealed at 473 K is shown in Fig. 9. Two broad bands, centered at 716 cm$^{-1}$ and 836 cm$^{-1}$, are observed. The width of the two bands ranges from 665 cm$^{-1}$ to 747 cm$^{-1}$ and from 762 cm$^{-1}$ to 890 cm$^{-1}$, respectively. The first band is related to the phonon mode [E$_1$(TO)] of AlN while the second band again consists of two peaks; one is associated with the phonon mode [A$_1$(LO)] of AlN while the second is related to aluminium oxide [38]. Again the existence of AlN and Al$_2$O$_3$ bands confirms the deposition of AlON nanolayers. Table 1 shows the existence of E$_1$(TO) and A$_1$(LO) bands of AlN reported by different authors. It is clear that the positions of E$_1$(TO) and A$_1$(LO) bands vary respectively from 665 cm$^{-1}$ to 684 cm$^{-1}$ and 870 cm$^{-1}$ to 894 cm$^{-1}$, depending on the substrates and energy of the irradiated ions. Table 1 also indicates that the E$_1$(TO) phonon mode of AlN shifts towards higher wave numbers (from 667 cm$^{-1}$ to 684 cm$^{-1}$ just by changing the orientation of silicon [Si (111) to Si (100)] substrate, whereas the existence of the A$_1$(LO) phonon mode of AlN varies from 870 cm$^{-1}$ to 894 cm$^{-1}$ for different substrates. Moreover, they have used comparatively low energy ions to deposit the AlN surface layer [39–43], whereas in the present work, high energy ions ranging from 80 keV to 1.4 MeV are used to deposit the AlN surface layer. During ion surface interaction, high energy ions transfer enough energy to the substrate surface, which results in repeated rapid thermal annealing during multiple fo-
cus shots exposure and in turn affects the appearance of the phonon modes of AlN. Moreover, the overlapping of two bands is also responsible for the shifting of the bands from their corresponding stress-free values. This overlapping of bands is due to the presence of oxides coming from the open air during continuous annealing. Raman analysis confirms the existence of oxides, while XRD analysis reveals no diffraction peak related to oxides, thereby indicating that the XRD technique is not appropriate for detecting the oxides since oxides are present in amorphous form or in lower contents.

3.4 FTIR analysis

FTIR is an effective technique for investigating the characteristic vibrational modes of lattice and bond formation. Fig. 10 exhibits the FTIR spectrum of the deposited surface layer annealed at 473 K. An absorption peak related to AlN is observed around 667.8 cm\(^{-1}\) wave number. It has been reported that peaks related to AlN were observed around 670 cm\(^{-1}\) and 673 cm\(^{-1}\). However, shifting of peaks from 681 cm\(^{-1}\) to 691 cm\(^{-1}\) depends on discharged powers and the substrate materials [44\textasciitilde46]. The peak shifting to lower wave number indicates the development of residual stresses which are associated with the diffusion of oxides interstitially and more diffusion of oxide results in enhanced residual stresses. Thus, diffusion of oxides plays an important role in the enhancement of residual stresses. A weak peak at about 692 cm\(^{-1}\) related to aluminium oxide is observed, confirming the existence of oxides. A chemical reaction takes place between aluminium and oxygen and results in the formation of aluminium oxide. Thus, the appearance of absorption peaks related to AlN and Al\(_2\)O\(_3\) phases confirms the deposition of AlON nanolayers. Such a type of chemical bond has been reported in the literature [47].

3.5 Surface morphology

Fig. 11 exhibits the surface microstructure of AlN surface layers deposited with multiple (5, 10, 15 and 20) focus shot ion irradiations. All the samples are placed at 10 cm in front of the central anode. Fig. 11(A) shows the formation of rounded grains of different sizes ranging from ~20 nm to 70 nm, which are distributed over
the entire surface. Moreover, some dark regions of irregular shapes are also observed. The sizes of the particles observed in the dark regions are comparatively smaller in dimension.

Fig. 11(B) and (C) again exhibit the formation of rounded grains but they are larger, ranging from 40 nm to 200 nm. These dark and bright regions make the surface rough. Careful investigation reveals the formation of a double layer structure since dark and bright regions have particles size of different dimensions. Fig. 11(D) again demonstrates the formation of rounded grains ranging from 25 nm to 75 nm. However, many large agglomerates are observed showing rounded grains. It is quite clear that these agglomerates are empty in the center but compact at the boundaries. This infers that the growth of agglomerates propagates from the outer to the inner regions. The formation of larger agglomerates on the top surface makes the surface rough. Fig. 11 reveals that the film surface roughness (from its appearance) increases with the increase of ion dose. Moreover, the substrate temperature is associated with the ion dose; as the ion dose increases, substrate temperature increases, resulting in the growth of particles in the form of rounded grains as well as agglomerates.

Fig. 12 demonstrates the surface morphology of the as-deposited and annealed AlN surface layers. Careful investigation shows the formation of a net type microstructure of circular pores (∼50 nm) along with the formation of large but compact agglomerates (300∼500 nm). The large agglomerates comprise smaller grains (∼40 nm). However, the microstructure of the annealed (473 K) AlN surface layer indicates the formation of smaller agglomerates which is due to the rearrangement of nanoparticles by gaining energy during continuous annealing. It is obvious from the microstructure’s appearance that the annealed AlN layer is comparatively compact and smooth in nature. Thus, uniform distribution of smaller agglomerates and formation of a smooth and compact surface layer are due to sufficient energy absorption during continuous annealing.

4 Conclusions

AlON nanolayers are synthesized on aluminium substrates by the irradiation of energetic nitrogen ions emanated from PF device. Ion energy and number density vary from 80 keV to 1.4 MeV and $5.6 \times 10^{19} \text{ m}^{-3}$ to $1.3 \times 10^{19} \text{ m}^{-3}$, respectively. The substrate temperature is associated with ion energy and ion number density; more ion number density, more will be the substrate temperature. The average crystallite size of AlN (111) plane increases while stress relaxation in AlN (111) and (200) planes is observed with increasing number of focus shots. Stress transformation (from compressive to tensile) in AlN (200) plane is also observed for 10 focus shots. Lower annealing (473 K) temperature gives better crystallinity of AlN while it decreases at higher annealing (523 K) temperature. Moreover, AlN (220) plane appears at 473 K, but vanishes at higher annealing temperature (523 K). The diffraction peaks appearing during ion irradiation are more stable than the diffraction peak obtained at 473 K. The texture coefficient of AlN (200) plane is associated with annealing temperature. Raman analysis reveals the existence of two broad bands (centered at 716 cm$^{-1}$ and 836 cm$^{-1}$) which are attributed to AlN. These bands are shifted from their stress-free values due to the presence of non-bounded oxides, which results in the development of stresses. FTIR analysis shows the existence
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of absorption peaks at 667.8 cm$^{-1}$ (related to AlN) and at 692 cm$^{-1}$ (related to aluminium oxide). The results confirm the formation AlON nanolayers. SEM micro-structural features, such as grain size, formation of dark region, bright region, agglomerates ($\sim$300 nm to $\sim$500 nm) and their decomposition, are related with the number of focus shots and the annealing temperature. A net type microstructure consisting of circular holes of diameter $\sim$50 nm is also observed for 25 focus shots.

References


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