International Journal of Chemical Sciences

Review | Vol 15 Iss 2

Preparation of Nanocrystalline Aluminum Oxide Thin Films: A Review

Ho SM[*]

Faculty of Information Technology, Maths and Science, INTI International University, Putra Nilai, Negeri Sembilan, Malaysia

[*]**Corresponding author:** Ho SM, Faculty of Information Technology, Maths and Science, INTI International University, Putra Nilai, Negeri Sembilan, Malaysia, Tel: +6067982000; E-mail: soonmin.ho@newinti.edu.my

Received: February 03, 2017; **Accepted:** April 10, 2017; **Published:** April 12, 2017

Abstract

Nowadays, binary compound such as aluminium oxide thin films are well known for applications in a wide range of microelectronics devices and optoelectronics. This work demonstrates that it is possible to prepare aluminium oxide thin films by using various deposition methods including chemical deposition method and physical deposition technique. Characterization of thin films is a broad science. Here, the characterization of the films was done with x-ray diffraction, scanning electron microscopy, X-ray photoelectron spectroscopy, Fourier transform infrared absorption and UV-visible spectrophotometer.

Keywords: Thin films; Aluminium oxide; Semiconductor; Solar cells

Introduction

Recently, there has been growing interest to prepare various types of metal chalcogenide thin films [1-30] and metal oxide films [31-45] by using different deposition methods. Aluminium oxide thin films have been extensively studied due to their promising applications in optoelectronics devices, microelectronics devices, water repellent coating, protective coating, dielectric and sensing layer. These materials have many properties include chemical inertness, good adhesion to glass substrate, transparency over wide wavelength range, mechanical strength, high thermal conductivity, and high dielectric constant.

There are several different deposition techniques either physical deposition method or chemical deposition technique have been applied for the preparation of aluminium oxide thin films such as reactive magnetron sputtering method, thermal evaporation method, pulsed laser deposition, plasma enhanced chemical vapor deposition method, sol gel technique, atomic layer epitaxy and chemical vapor deposition. Here, a critical analysis was carried out in order to prepare aluminium oxide films. Then the obtained films were characterized using X-ray photoelectron spectroscopy, scanning electron microscopy, x-ray diffraction and Fourier transform infrared.

Citation: Ho SM. Preparation of Nanocrystalline Aluminum Oxide Thin Films: A Review. Int J Chem Sci. 2017;15(2):115.

Literature survey

Zhao et al. [46] have reported the synthesis of aluminum oxide thin films by using off plane filtered cathodic vacuum arc system. They figure out that aluminum oxide films could be deposited with high deposition rate (1.5 nm/s to 0.5 nm/s) on quartz and Si (100) substrates at room temperature under various oxygen pressures (0.01 Pa to 0.0373 Pa). The obtained results show that the as-grown films are amorphous with low stress (lower than 0.5 GPa) and good uniformity could be seen. They conclude that the film properties display the potential applications in optical coatings industry. On the other hand, the influence of annealing process on the properties of aluminium oxide thin films has been discussed by Zhao and Tay [47]. It is noticeable that the surface of films remained smooth up to 600°C. The experiment results indicated that crystallization is induced for the films when heated at 900°C. Also, they claim that the refractive index increased with increasing the annealing temperature from 200°C to 900°C.

Sol gel technique has been used to prepare aluminum oxide thin films by Nursen et al. [48]. In the experiment described, acetyl acetone, aluminium sec-butoxide and alcohol solution (ethanol, isopropanol and n-butyl alcohol) was used as a chelating agent, raw materials, and solvent, respectively. The data obtained show that the most stable one was synthesized by using n-butyl alcohol if compared to other solvents. On the other hand, highly transparent, homogeneous and amorphous aluminium oxide films were observed on silicon substrate after annealing the films at 500°C. X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared absorption (FTIR) spectroscopy tests verify that the deposited films were hydroxide free.

Atomic layer epitaxy deposition and chemical vapor deposition were used to prepare aluminium oxide film as proposed by Hiltunen et al. [49]. Atomic layer deposition technique has many benefits include low defect density could be achieved, can produce pin hole free films and desired specified thickness. Meanwhile, chemical vapor deposition method has many advantages such as can deposit films which are hard to evaporate, high growth rates and can grow epitaxial films. In their study, the $AlCl_3$, water, oxygen and aliphatic alcohol were used as aluminium and oxygen source respectively. Finally, the electrical constants and environmental stability analysis show that the obtained films are suitable for applications such as insulating and protecting layers. In other case, Pradeep et al. [50] have synthesized Al_2O_3 films with thicknesses of 400, 300 and 200 nm on silicon and soda lime glass substrates using atomic layer deposition. The optical behavior was studied in the 400 to 1800 nm wavelength range. The obtained outputs reflect that the films grown on glass substrate have higher refractive indices as compared to the films deposited on silicon.

Plasma enhanced chemical vapor deposited method has been used to prepare aluminum oxide thin films as suggested by Wen and Suhr [51]. The choice of this method due to it possesses a number of advantages such as low operation temperature, lower chances of cracking deposited layer and good step coverage. In their experiments, the aluminium oxide films were deposited onto various substrates includes glass, quartz, steel, silicon, and nickel using aluminium acetyl acetonate as precursor. It is interesting to note that the deposits are hard (up to 2370 HK) and indicate good adherence to the substrates.

Aluminum oxide thin films were prepared by using pulsed laser deposition method as described by Arrieta et al. [52]. This deposition method has several advantages such as cost effective, fast and versatile method. They point out that the obtained films are amorphous and slightly deficient in oxygen. Also, these films indicate a smooth surface with dispersed splashed particles with diameters ranging from 0.2 to 0.5 micrometers. In their study, the thermo luminescent properties of films were investigated. They conclude that thermo luminescent curve indicate two peaks at 110°C and 176°C. Furthermore, a linear relationship between absorbed dose and the thermoluminescent response for doses span from 150 Gy to 100 Gy was detected. In another case, Al_2O_3 films of 800 nm thicknesses were prepared by Ion et al. [53] using pulsed laser deposition method. The scanning electron microscopy (SEM) and atomic force microscopy (AFM) analysis show that diameter of grains about 50 nm. Energy dispersive X-ray spectroscopy (EDX) investigation points out that a stoichiometric transfer from target to film. The atomic percentage of oxygen and aluminium is 63.25% and 36.75%, respectively.

Electro spraying method has been applied as a means for the deposition of Al_2O_3 films on a metal substrate. Jaworek et al. [54] have described that electrospray system contained stainless steel capillary nozzle and a heated table of diameter of 120 mm. The SEM micrographs reflect that the particles produce a tight layer of particles without visible agglomerates. They further explain that the bigger grains (estimated about 1 μm to 2 μm) are due to an effect of coagulation of the smaller ones.

Aluminium oxide thin films were prepared using thermal evaporation method on aluminium substrate in open air environment as reported by Khan et al. [55]. The surface morphology has been studied by scanning electron microscopy. The formation of rounded grains (1 μm to 2.5 μm) and irregular patch of particles was detected for 1 and 2 hours. However, the surface morphology displayed the formation of irregular patches distributing uniformly for the 5 hours. They conclude that the formation of microstructure mainly depended on treatment time (1 to 5 hours). On the other hand, the XRD data show that crystallinity of various planes varied with increasing treatment time. Also, XRD patterns display the formation of Al_2O_3, $Al(OH)_3$, and $Al_2O_3.H_2O$ phases.

Solvothermal approach was employed to prepare aluminium oxide thin films as suggested by Duan et al. [56]. This method allows for the precise control over the size, shape distribution and crystallinity of films. The SEM analysis shows that the obtained films are dense and grainy surface morphology. The thickness of the films was estimate to be 150 nm to 300 nm.

Chemical liquid phase deposition was employed for the first time to prepare Al_2O_3 films as described by Jie and Sun [57]. They observe that the growth rate is 12 nm/h with the deposition carried out at 15°C by using 0.0837 M of $Al_2(SO_4)_3$ and 0.214 M of $NaHCO_3$. They also found that post growth annealing not only densifies and purifies the films, but lead to film crystallization as well.

The preparation of Al_2O_3 thin films using electrodeposition method has been proposed by Farzana et al. [58]. The films are electrodeposited at 5°C, at 3 V in deposition time from 30 to 120 minutes. The thickness increases from 50 nm to 250 nm as the deposition time was increased from 30 to 120 minutes. They claim that these materials could be used as barrier coatings due to the surface roughness is in the range of 5.9 nm to 15.4 nm. Lastly, they suggest that the band gap reduces from 4.38 eV to 4 eV with increase in deposition time.

X-ray diffraction (XRD) analysis was conducted in order to learn more about the film structure. Effect of bath temperature on the properties of electrodeposited Al_2O_3 films was investigated and reported by Imran et al. [59]. XRD results confirmed the formation of aluminium oxide at the bath temperature of 5°C to room temperature. Also, XRD analysis indicates that the crystallite size reduces as the bath temperature is increased.

Reactive magnetron sputtering method was used to prepare aluminium oxide thin films on soda glass substrate as described by Koushki et al. [60]. The XRD profile suggests that the formation of cubic aluminium oxide phase. SEM analysis indicates that the average grain size of 25 nm was detected. On the other hand, the influence of introduced oxygen flow on optical properties of films was studied. The optical spectra highlight that the highest transmission value belongs to highest amount of introduced oxygen. Researchers have claim that the deposition process was controlled by target voltage. For example, the maximum deposition rate obtained was 215 nm/minutes, 77% of the rate of metallic aluminium as reported by

Juliet et al. Meanwhile, Kari et al. [61] have observed that the maximum deposition rate was 150 nm/minutes from the value of metallic minimum. In the other case, the influence of annealing temperature on the properties of aluminium oxide films was investigated by Lim et al. [62]. They conclude that the crystallite size was detected to increase, whereas the micro strain and dislocation density were reduced by increasing the annealing temperature. They explain that due to the reduction in the lattice strain.

Conclusions

Aluminium oxide thin films have been prepared by using different deposition methods. The properties of obtained thin films were influenced by various experimental conditions such as annealing temperature, target voltage, oxygen flow and deposition time.

Acknowledgement

Inti International University is gratefully acknowledged for the financial support of this work.

REFERENCES

1. Fekadu GH, Francis KA. Effect of deposition temperature on the structural, morphological and optical band gap of lead selenide thin films synthesized by chemical bath deposition method. Mater Chem Phys. 2016;183:320.
2. Joshi Rk, Kumar P, Sehgal HK, et al. Study of solution grown variable band gap $Pb_{1-x}Mn_xS$ semiconductor nanoparticle films. J Electrochem Soc. 2016;153:C707.
3. Anuar K, Tan WT, Jelas M, et al. Effects of deposition period on the properties of FeS_2 thin films by chemical bath deposition method. Thammasat Int J Sci Technol. 2010;15:62.
4. Lin LH, Wu CC, Lai CH, et al. Controlled deposition of silver indium sulfide ternary semiconductor thin films by chemical bath deposition. Chem Mater. 2008;20:4475.
5. Anuar K, Ho SM, Tan WT, et al. Composition, morphology and optical characterization of chemical bath deposited ZnSe thin films. Eur J Appl Sci. 2011;3:75.
6. Ottih IE, Ekpunobi AJ. Effects of complexing agents on chemically deposited $Mg_{1-x}Ni_xS$ thin films. Moldavian J Phys Sci. 2012;11:209.
7. Anuar K, Nani R, Ho SM. Atomic force microscopy studies of zinc sulfide thin films. Int J Adv Eng Sci Technol. 2011;7:169.
8. Lugo S, Lopez I, Peria Y, et al. Avellaneda, Characterization of $CuInS_2$ thin films prepared by chemical bath deposition and their implementation in a solar cell. Thin Solid Films. 2014;569:76.
9. Alias MFA, Naji IS, Taher BY. Influence of substrate temperatures on the optical properties of thin Cu_3SnS_4 films prepared by CBD. IPASJ Int J Electr Eng. 2014;2:1.
10. Anuar K, Saravanan N, Tan WT, et al. Effect of deposition period and pH on chemical bath deposited Cu_4SnS_4 thin films. Phil J Sci. 2009;138:161.
11. Balasubramanian V, Suriyanarayanan N, Kannan R. Optimization of chemical bath deposited bismuth sulphide thin films on glass substrate. Arch Appl Sci Res. 2012;4:1864.
12. Bicer M, Sisman I. Electrodeposition and growth mechanism of SnSe thin films. Appl Surf Sci. 2011;257:2944.
13. Anuar K, Saravanan N, Zulkefly K, et al. Influence of complexing agent (Na_2EDTA) on chemical bath deposited Cu_4SnS_4 thin films. Bull Chem Soc Ethiop. 2010;24:259.

14. Gopakumar N, Anjana PS, Pillai PKV. Chemical bath deposition and characterization of CdSe thin films for optoelectronic applications. J Mater Sci. 2010;45:6653.
15. Hannachi A, Hammami S, Raouafi N. Preparation of manganese sulfide (MnS) thin films by chemical bath deposition: Application of the experimental design methodology. J Alloys Compd. 2016;663:507.
16. Garcia LV, Loredo SL, Shaji S, et al. Structure and properties of CdS thin films prepared by pulsed laser assisted chemical bath deposition. Mater Res Bull. 2016;83:459.
17. Hatam EG, Ghobadi N. Effect of deposition temperature on structural, optical properties and configuration of CdSe nanocrystalline thin films deposited by chemical bath deposition. Mater Sci Semicond Process. 2016;43:177.
18. Pitchaimani K, Amalraj L. Microstructure, optical and structural characterization of $Cd_{0.98}Fe_{0.02}S$ thin films co-doped with Zn by chemical bath deposition method. Muthukumaran Physica E. 2016;78:56.
19. Anuar K, Ho SM, Saravanan N. Preparation of lead selenide thin films by chemical bath deposition method in the presence of complexing agent (tartaric acid). Turk J Sci Technol. 2011;6:17.
20. Khan MD, Hameed S, Haider N, et al. Deposition of morphology tailored PbS thin films by surfactant enhanced aerosol assisted chemical vapor deposition. Mater Sci Semicond Process. 2016;46:39.
21. Liu R, Tan M, Xu L, et al. Tang, preparation of high quality Cu_2ZnSnS_4 thin films for solar cells via the improvement of sulfur partial pressure using a static annealing sulfurization approach. Sol Energy Mater Sol Cells. 2016;157:221.
22. Anuar K, Ho SM, Lim KS, et al. SEM, EDAX and UV-Visible studies on the properties of Cu_2S thin films. Chalcogenide Lett. 2011;8:405.
23. Hassanien AS, Aly KA, Akl AA. Study of optical properties of thermally evaporated ZnSe thin films annealed at different pulsed laser powers. J Alloys Compd. 2016;685:733.
24. Anuar K, Ho SM, Tee WT, et al. Morphological characterization of CuS thin films by atomic force microscopy. Res J Appl Sci Eng Technol. 2011;3:513.
25. Harizi A, Rabeh MB, Laatar F, et al. Substrate temperature dependence of structural, morphological and optical properties of $Sn_4Sb_6S_{13}$ thin films deposited by vacuum thermal evaporation. Mater Res Bull. 2016;79:52.
26. Chander S, Dhaka MS. Effect of thickness on physical properties of electron beam vacuum evaporated CdZnTe thin films for tandem solar cells. Physica E. 2016;84:112.
27. Ho SM. Influence of complexing agent on the growth of chemically deposited $Ni_3Pb_2S_2$ thin films. Oriental J Chem. 2014;30:1009.
28. Ahmad SM. Study of structural and optical properties of quaternary $Cu_xAg_{1-x}AlS_2$ thin films. Opt Int J Light Electron Opt. 2016;127:10004.
29. Gurav KV, Kim YK, Shin SW, et al. Pulsed electro deposition of Cu_2ZnSnS_4 thin films: Effect of pulse potentials. Appl Surf Sci. 2015;334:192.
30. Ma S, Sui J, Cao L, et al. Synthesis of Cu_2ZnSnS_4 thin film through chemical successive ionic layer adsorption and reactions. Appl Surf Sci. 2015;349:430.
31. Ezema FI, Ekwealor ABC, Osuji RU. Optical properties of chemical bath deposited nickel oxide (NiO_x) thin films. Superficies Vacio. 2008;21:6.
32. Gomaa MM, Boshta M, Farag BS, et al. Osman, Structural and optical properties of nickel oxide thin films prepared by chemical bath deposition and by spray pyrolysis techniques. J Mater Sci Mater Electron. 2016;27:711.
33. Noh S, Lee E, Seo J, et al. Electrical properties of nickel oxide thin films for flow sensor application. Sens Actuators A. 2006;125:363.

34. Han SY, Lee DH, Chang YJ, et al. The growth mechanism of nickel oxide thin films by room temperature chemical bath deposition. J Electrochem Soc. 2006;153:C382.

35. Osuwa JC, Onyejiuwa GI. Structure and electrical properties of annealed nickel oxide thin films prepared by chemical bath deposition. J Ovonic Res. 2013;9:9.

36. Desai SP, Suryawanshi MP, Bhosale SM, et al. Influence of growth temperature on the physico chemical properties of sprayed cadmium oxide thin films. Ceram Int. 2015;41:4867.

37. Uplane MD, Kshirsagar PN, Lokhande BJ, et al. Preparation of cadmium oxide films by spray pyrolysis and its conversion into cadmium chalcogenide films. Indian J Pure Appl Phys. 1999;37:616.

38. Lokhande BJ, Uplane MD. Effect of deposition temperature on spray deposited cadmium oxide films. Mater Res Bull. 2001;36:439.

39. Metodija ZN, Ivan SG, Biljana MS. Oriented cadmium oxide thin solid films. J Mater Chem. 1996;6:761.

40. Urbiola IRC, Bon RR, Vorobiev YV. The transformation to cadmium oxide through annealing of cadmium oxide hydroxide deposited by ammonia free SILAR method and the photocatalytic properties. Thin Solid Films. 2015;592:110.

41. Mahalingam T, Chitra JS, Chu JP, et al. Photoelectrochemical solar cell studies on electroplated cuprous oxide thin films. J Mater Sci Mater Electron. 2006;17:519.

42. Morales J, Sanchez L, Martin F, et al. Use of low temperature nanostructured CuO thin films deposited by spray pyrolysis in lithium cells. Thin Solid Films. 2005;474:133.

43. Mugwang FK, Karimi PK, Njoroge WK, et al. Optical characterization of copper oxide thin films prepared by reactive dc magnetron sputtering for solar cell applications. Int J Thin Films Sci Technol. 2016;2:15.

44. Rachel O, Usha R. Characteristics of electron beam evaporated and electrodeposited Cu_2O thin films-comparative study. Int J Electrochem Sci. 2012;7:8288.

45. Rastkar AR, Niknam AR, Shokri B. Characterization of copper oxide nanolayers deposited by direct current magnetron sputtering. Thin Solid Films. 2009;517:5464.

46. Zhao ZW, Tay BK, Yu GQ, et al. Optical properties of aluminium oxide thin films prepared by room temperature by off plane filtered cathodic vacuum arc system. Thin Solid Films. 2004;14:447-8.

47. Zhao Z, Tay BK. Property study of aluminium oxide thin films by thermal annealing. Phys Status Solid C. 2012;9:77.

48. Nursen A, Philippe FS, Johan L, et al. Dirk, Optical and structural properties of aluminium oxide thin films prepared by a non-aqueous sol gel technique. J Sol Gel Sci Technol. 2011;59:327.

49. Hiltunen L, Kattelus H, Leskela M, et al. Growth and characterization of aluminium oxide thin films deposited from various source materials by atomic layer epitaxy and chemical vapor deposition processes. Mater Chem Phys. 1991;28:379.

50. Pradeep K, Monika KW, Charles HW, et al. Optical properties of Al_2O_3 thin films grown by atomic layer deposition. Appl Opt. 2009;48:5407.

51. Wen Y, Suhr H. Multilayers, aluminium oxide thin films prepared by plasma enhanced chemical vapour deposition. 1992;55:176.

52. Arrieta A, Escobar L, Camps E, et al. Thermoluminescent response of aluminium oxide thin films subject to gamma irradiation. IX International Symposium/XIX Nacional Meeting on Solid State Dosimetry, Mexico city. 29.

53. Ion M, Berbecaru C, Iftimie S, et al. PLD deposited Al_2O_3 thin films for transparent electronics. Digest J Nanomater Biostructures. 2012;7:1609.

54. Jaworek A, Sobczyk AT, Krupa A, et al. Electrostatic deposition of nano thin films on metal substrate. Bull Polish Academy Sci. 2009;57:63.
55. Khan IA, Rashid A, Fatima A, et al. Cost effective deposition of aluminium oxide layers. J Basic Appl Sci. 2013;9:252.
56. Duan X, Tran N, Roberts NK, et al. Solvothermal approach for low temperature deposition of aluminium oxide thin films. Thin Solid Films. 2010;518:4290.
57. Jie S, Sun Y. Chemical liquid phase deposition of thin aluminum oxide films. Chin J Chem. 2004;22:661.
58. Farzana M, Saira R, Shahzad N. Low temperature formation of electrodeposited aluminium oxide thin films. 2nd International Conference on Biotechnology, Nanotechnology and its applications (ICBNA'2013) London (UK). 2013;pp:18.
59. Imran M, Majid F, Riaz S. Optical and structural properties of electrodeposited aluminium oxide at low temperatures. Advances in Nano, Biomechanics, Robotics and Energy Research, Seoul, Korea. 2013.
60. Koushki E, Mousavi SH, Baedi J. Oxygen amount effect on optical properties of aluminium oxide nanostructured films prepared by reactive magnetron. Opt Int J Light Electron Opt. 2016;127:4635.
61. Kari K, Jorma H, Pierre J. Voltage controlled reactive sputtering process for aluminium oxide thin films. Thin Solid Films. 1998;326:189.
62. Lim WQ, Mutharasu D. Variation of structural and surface properties of RF sputtered aluminium oxide (Al_2O_3) thin films due to the influence of annealing temperature and time. Int J Mater Sci Appl. 2014;3:404.