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Green Synthesis of Aluminium Oxide Nanoparticles by using Aerva Lanta and Terminalia Chebula Extracts

P. Duraisamy[1]
[1]*Assistant professor, Department of Physics, Thiruvalluvar Govt. Arts College, Rasipuram-637401, Tamilnadu, India*

Abstract: ***Eco-friendly synthesis of aluminium oxide nanoparticles was achieved by a novel, facile green route method using aerva lanta and terminalia chebula as a reducing agent. Aluminium oxide nanoparticles (Al_2O_3) were synthesized from aluminium nitrate using extracts of aerva lanta leaves and terminalia chebula seeds. Nanoparticles produced by plants are more stable and rate of synthesis is faster than that in other organisms. The present investigation was carried out to green synthesis of alumina nanoparticles by using medicinal plants. Deionised Water was taken as solvent medium. The formations of AlNP were initially monitored by the colour changes occurring in the reaction mixture during the incubation period. The successful formation of alumina nanoparticles has been confirmed by X-ray diffraction, UV-visible, FTIR and FE-SEM analysis. The X-ray diffraction data revealed the average size of the Al-NPs as 70 nm. The AlNP were found to be spherical agglomeration in shape in case of aerva lanta leaves extract with a size of 50–70 nm and to be spherical shaped in case of terminalia chebula seed extract with an average size of 50–100 nm.***

Keywords: ***Aluminium oxide nanoparticles, green synthesis, hydrothermal, aerva lanta, terminalia chebula***

I. INTRODUCTION

Nowadays, metal oxide nanoparticles are attracting noteworthy interest as they can change the viable unconventional to conventional materials in various fields of solid state. Metal oxide nanoparticles are being fundamentally used as a heterogeneous nanocatalyst in a variety of organic transformations as they contained high surface area than their bulk counterparts [1-2]. Besides, colloidal and monodisperable nanomaterials have also paying tremendous attention because of their small size effects particular in catalytical and biological applications [3-4]. Aluminium oxide is a compound of aluminium and oxygen with chemical formula of Al_2O_3. It is the most commonly occurring of several aluminium oxides, and specifically identified as aluminium (III) oxide. It is commonly called alumina. The oxides of aluminium materials are widely used in ceramics, refractories and abrasives due to their hardness, chemical inertness, high melting point, non-volatility and resistance to oxidation and corrosion [1-5]. Predominantly, aluminum oxide nanoparticles commonly known as alumina (Al_2O_3 NPs) have trapped the awareness of many researchers due to its great catalytical activities. An alumina nanoparticles can be synthesized by using many techniques including ball milling, spray combustion, hydrothermal, sputtering, sol-gel, microwave and laser ablation [5-10]. Out of all the methods, hydrothermal method proved more helpful to obtain well shaped materials with designed texture and composition at low processing temperatures[11-12]. The literature survey reveals that Al_2O_3 was synthesized by hydrothermal method using different precursor . Research on aluminium oxide nanoparticles have shown many different applications. In general, alumina has many interesting properties such as catalyst, as high stability and hardness insulation, surface protective coatings, as composite materials with tuneable mechanical properties, etc [6]. It can be used as biomaterial [3] but also at wastewater treatment [9] are just a few of these. This paper aims to obtain alumina with controlled nanoparticles size into a simple and efficient manner. Metal oxide nanoparticles have been extensively developed in the past decades. They have been widely used in many applications such as catalysts, sensors, semiconductors, medical science, capacitors, and batteries [1–6].

The plant Avera lanta(mountain knotgrass) known colloquially as *Sirupeelai.* It is a medicinal plant that has an excellent diuretic and lithontriptic property. Terminalia chebula, commonly called as kadukkai in Tamil and harde whole in English is a medical wonder and has many health benefits and medicinal uses. Terminalia chebula has been regarded very highly from ancient times by healers and is rightly called the mother of herbs. Jatropha curcas plant can grow in wastelands and grows on almost any terrain, even on gravelly, sandy and saline soils. Jatropha oil is produced from the seeds of the Jatropha curcas and the oil is considered to be an excellent source of bio-diesel.

II. EXPERIMENTAL

A. Preparation of the Aerva lanta leaves extract and terminalia chebula seeds extract

Fresh aerva lanta leaves were collected from Thiruvalluvar Govt Arts college, Rasipuram campus. The leaves were washed several times with water to remove the dust particles and then sun dried to removes the residual moisture. The extract was prepared by placing 50g of washed dried fine cut leaves in 250ml beaker along with 100ml of deionised water. The mixture was then boiled for 60 minutes. The extract was cooled to room temperature and filtered using filter paper. Terminalia chebula seeds were collected from local chidha medical shop. 20g of grinded terminalia chebula seed powder was dissolved in 100ml of deionised water. The mixture was then boiled for 60 minutes. The extract was cooled to room temperature and filtered using filter paper. These extract were stored in a refrigerator in order to be used for further experiments.

B. Preparing of Jatropha curcas seed extract

Jatropha curcas seeds were collected from local source. 50g of seeds washed and cleaned with double distilled water and dried with water absorbent paper. Then it was crushed with the help of motar and pestle and dispensed in 100ml of deionised water and heated for 1 hour. After filtration clear seed extract was obtained for further use.

C. Preparation of aluminium oxide nanoparticles using Aerva lanta extract

For synthesis of Al NPs, 50ml of aerva lanta leaves extract was taken and boiled to 60-80 degree celsius using a stirrer heater. 10ml of aluminium nitrate(0.1M) aqueous solution was added to the stirring extract solution. Then Jatropha curcas seeds extract (5ml) was added drop wise to the stirring solution This mixture is then boiled it reduced to deep yellow coloured paste. This paste was then collected in a ceramic crucible and heated in a muffle furnace at 500 degree Celsius for 2 hours. A light yellow coloured powder was obtained and this was carefully collected and packed for characterization.

D. Preparation of aluminium oxide nanoparticles using terminalia chebula extract

For synthesis of Al NPs, 50ml of terminalia chebula seeds extract was taken and boiled to 60-80 degree Celsius using a stirrer heater. 10ml of aluminium nitrate(0.1M) aqueous solution was added to the stirring extract solution. . Then Jatropha curcas seeds extract (5ml) was added drop wise to the stirring solution. This mixture is then boiled until it reduced to deep yellow coloured paste .This paste was then collected in a ceramic crucible and heated in a muffle furnace at 500 degree Celsius for 2 hours. A light yellow coloured powder was obtained and this was carefully collected and packed for characterization.

III. CHARACTERIZATION OF ALUMINIUM OXIDE NANOPARTICLES

A. UV-Visible absoorption Spectra analysis

Absorption in the near ultraviolet region arises from electronic transitions associated within the sample. UV–Vis absorption spectra of prepared and annealed AlNP are shown in Fig.1(a,b). For as aerva lanta extract mediated synthesized Al_2O_3 nanoparticles, the strong absorption band at low wavelength near 271 nm. correspond to bandgap energy of 2.65 ev (figure 1a). The UV–Visible spectrum of AlNP synthesysized from terminalia chebula seeds extract shows the peak at 273nm(figure.1b).

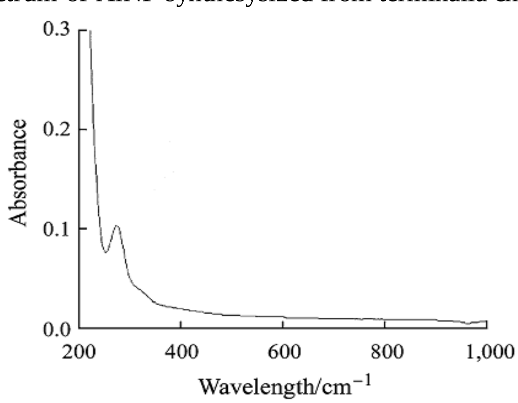

Fig.1(a). UV–Vis of AlNP using Aerva lanta extract

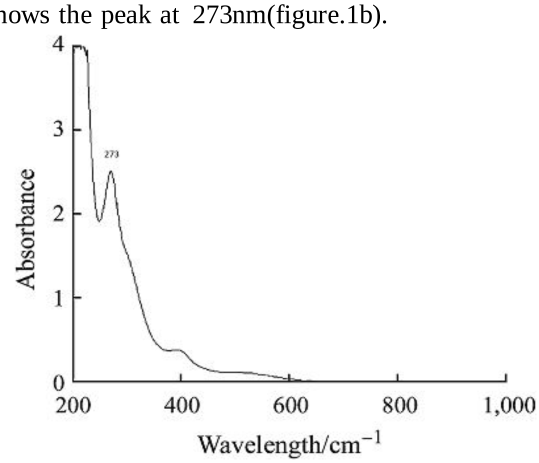

Fig.1(b) UV–Vis of AlNP using terminalia chebula extract

B. XRD Analysis

X-ray diffraction (XRD) is a powerful nonde structive technique for characterizing crystalline materials**.** It provides information on crystal structure, phase, preferred crystal orientation (texture), and other structural parameters, such as average grain size, crystallinity, strain, and crystal defects. X-ray diffraction peaks are produced by constructive interference of a monochromatic beam of X-rays diffracted at specific angles from each set of lattice planes in a sample. The peak intensities are determined by the distribution of atoms within the lattice. The determination of the crystalline phases was carried out by using X-ray diffraction (XRD) patterns which were obtained with Cu K radiation (λ = 1.5406 A = 20-80) atθO and room temperature. By using scherrer's equation the average crystallite size of obtained powders was estimated from XRD pattern. The structure of synthesized particles was investigated by XRD measurement. To identify the structural difference between AlNP with Aerva lanta extract and AlNP with terminalia chebula extract , the XRD measurement for AlNP target was carried out in fig.2. The observed peaks in figure 2 could be indexed based on AlNP with Aerva lanta extract and AlNP with terminalia chebula extract pure Al and Al_2O_3 phase in Joint Committee on Powder Diffraction Standard-International Center for Diffraction Data (JCPDS-ICDD) Card nos. 85-1327 and 29-0063, respectively. It is seen from the XRD spectra that all particles obtained from the synthesis are Al_2O_3. This confirms that Al transformed into Al_2O_3 after the synthesis. Moreover, it is clearly observed that the intensity of XRD peaks increased in AlNP with terminalia chebula extract. The peaks obtained in all the samples at 2θ = 31.9^o **,**36.5^o, 44.6^o,,54.3^o,57.6^o, 63.5^o corresponds to the lattice planes (220), (311), (400), (422), (511), (400) respectively..

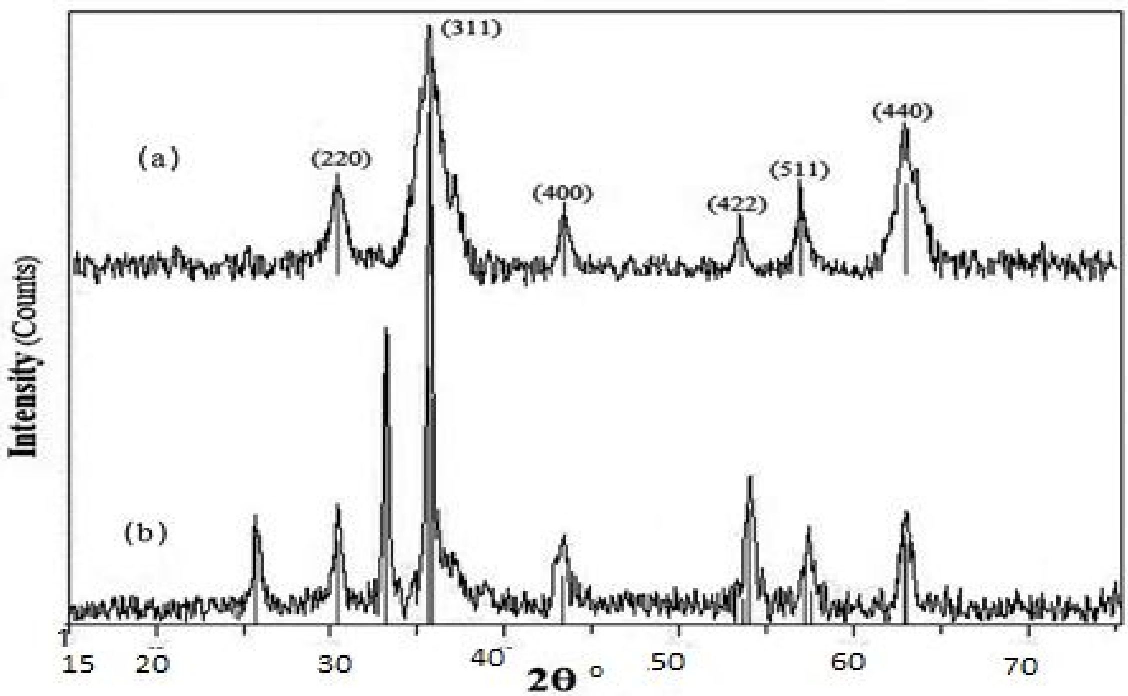

Fig. 2. XRD pattern of Al NPs (a) using Aerva lanta extract (b) using terminalia chebula extract

X-ray diffraction is a convenient method for determining the mean size of nano crystallites in nano crystalline bulk materials. The first scientist, Paul Scherrer, published his results in a paper that included what became known as the Scherrer equation in 1981 .This can be attributed to the fact that "crystallite size" is not synonymous with "particle size", while X-Ray diffraction is sensitive to the crystallite size inside the particles. From the well-known Scherrer formula the average crystallite size,

$$D = k \lambda / (\beta \cos\theta)$$

K
L
λ
β
θ
= (1)

where λ is the X-ray wavelength in nanometer (nm), β is the peak width of the diffraction peak profile at half maximum height resulting from small crystallite size in radians and K is a constant related to crystallite shape, normally taken as 0.9. The value of β in 2θ axis of diffraction profile must be in radians. Scherrer equation, $d = 0.9\lambda/ (\beta \cos \theta)$ was used to estimate grain average sizes of crystallites. The average grain size of prepared sample (a) is 65 nm and for sample(b) is 80nm.

C. Sem Analysis

Scanning Electron Microscopy uses a focused beam of high-energy electrons to generate a variety of signals at the surface of solid specimens. In most SEM microscopy applications, data is collected over a selected area of the surface of the sample and a two-dimensional image is generated that displays spatial variations in properties including chemical characterization, texture and orientation of materials. The SEM is also capable of performing analyses of selected point locations on the sample. This approach is especially useful in qualitatively or semi-quantitatively determining chemical compositions, crystalline structure and crystal orientations. The Scanning Electron Microscopy (SEM) micrographs of AlNP using Aerva lanta extract clearly indicates the formation of spherical AlNP with the size ranging between 50 nm and 70 nm as shown in the Fig.3 (a, b). From the SEM images (Fig. 4. a,b) can be observed the surface topography of sample using terminalia chebula extract. The obtained nanoparticles have regular geometric shapes which can be assimilated with spheres with diameters between 50nm and 100nm.

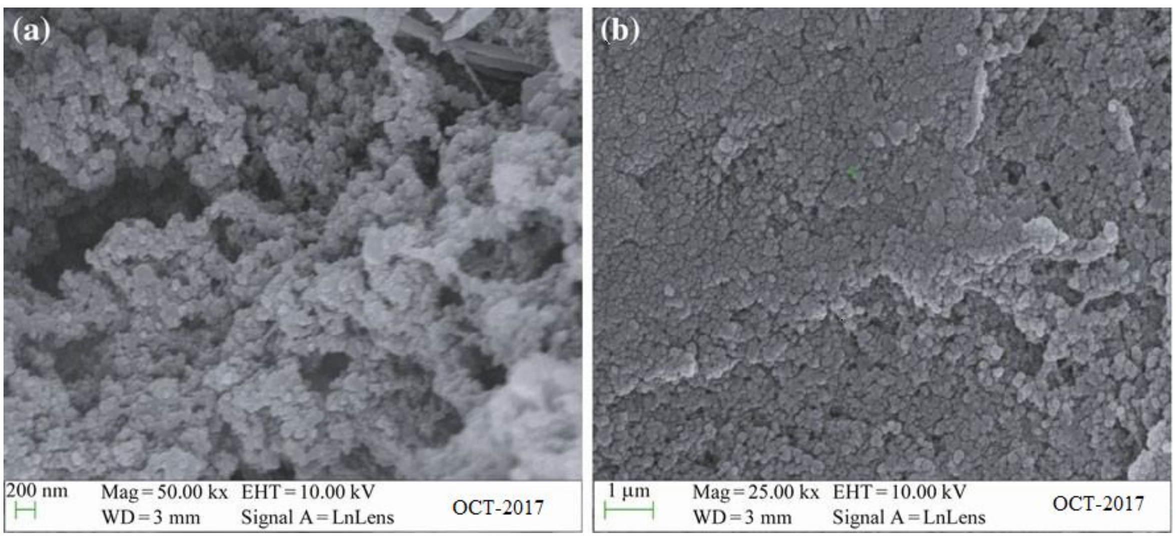

Fig. 3 SEM images of AlNP using Aerva lanta extract

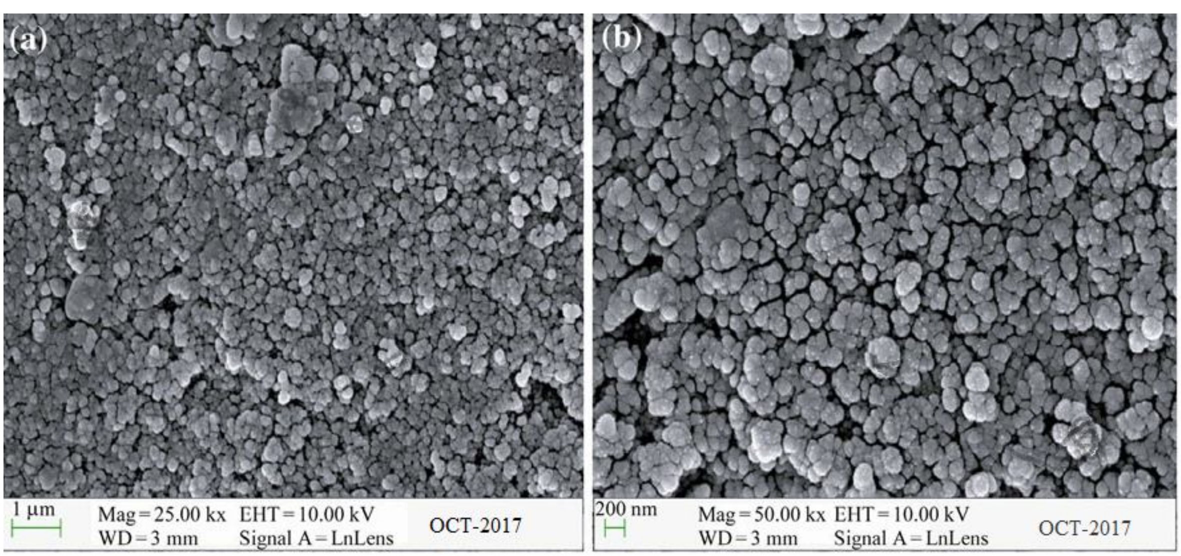

Fig. 4. SEM images of AlNP using using terminalia chebula extract

D. Ftir Spectra Analysis

Fourier transform infrared (FTIR) spectroscopy that deals with the infrared region of the electromagnetic spectrum that is light with a longer wavelength and lower frequency than visible light. FTIR(fig.5) peaks of AlNP using Aerva lanta extract at 1630, 1385 and 1092cm^{-1}. The peak at 1092cm^{-1} indicates the presence of C-N stretching frequency, whereas the germinal ethyl group at 1,383 cm^{-1} and peak at 1,630 cm^{-1} represent the unreacted ketone group, suggesting the presence of flavonones adsorbed on the surface of AlNP. This suggests that influence of water soluble organic moieties of aerva lanta is responsible for the synthesis of AlNP and surface modification. FTIR is shown in Fig. 6, showing peaks at 1,633, 1,380 and 1,080 cm^{-1}. This indicates the secretion of some water soluble organic components, which might contribute for the reduction of aluminium nitrate into AlNP. FTIR spectra enable to identify the material, the biomolecules responsible for the reduction of metal ions and capping of the bio reduced metal nanoparticles synthesized by using plant extract.

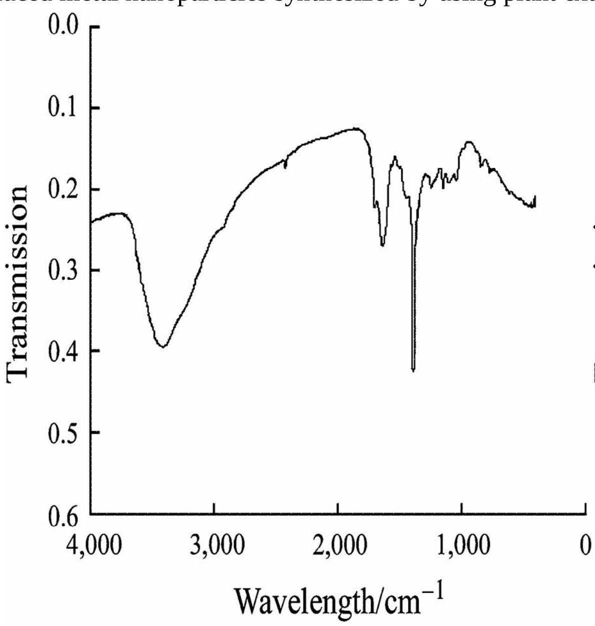

Fig. 5.FTIR of AlNP using Aerva lanta extract

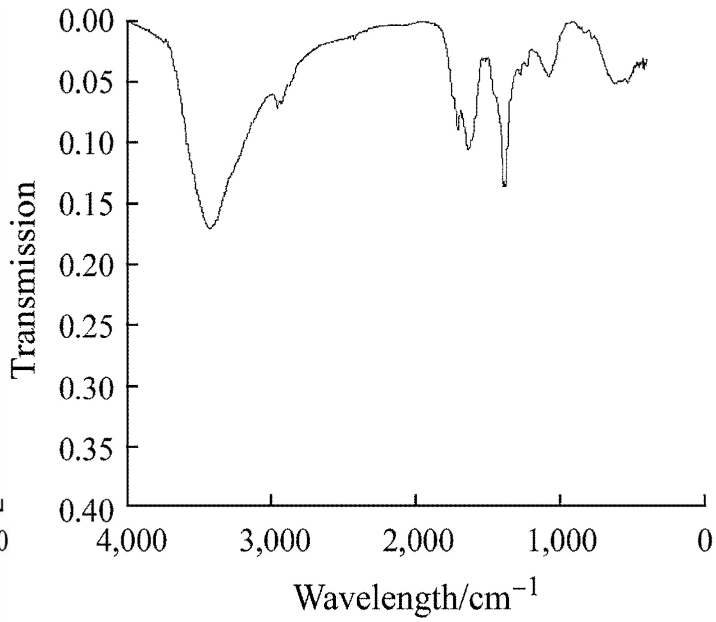

Fig. 6. FTIR of AlNP using terminalia chebula extract

IV. CONCLUSION

Plant synthesized metal nanoparticles are more stable, safe and easily scaled up. For the future developmental point of view genetically modified plants can improve the properties of nanoparicles. Aluminium nanoparticles were successfully obtained from bioreduction of aluminium nitrate using aerva lanata plant leaves extract and terminalia chebula plant seed extract acted as reducing and stabilizing agent. Here the drop wise added Jatropha curcus seed extract solution acted as a bio-fuel. Aluminum oxide nanoparticles was successfully synthesized by hydrothermal method and characterized by various standard spectroscopic techniques to know its particle sizes and shape. Further we effectively developed a method involving the use of alternate, an efficient, safer nanocatalyst for solvent free green synthesis of derivative. To the best of our knowledge, this is the first time reported synthesis of dehydro-aromadendrene in avera lanta and chebulinic acid, methyl ester derivatives in terminalia chebula by using alumina catalyst. The mildness of the conversion, experimental simplicity, excellent yields, and shorter reaction time makes this procedure more gorgeous in synthesizing a variety of derivatives. Along with this a facile approach in recovery and reusability of the alumina catalyst is significant toward environmentally benign procedures. Therefore, alumina nanoparticles can contribute the green synthesis of aromatic organic compounds in solvent free condition. . It is known that these characteristics significantly depend on the type of the fuel used as reagent. In this study, Jatropha curcus seed extract solution used as fuel. Also, it can be used for various organic transformation reactions. The present study investigates synthesis of nanoparticles of alumina with controlled size (smaller than 100nm) using hydrothermal treatment. The morphological and structural analysis of nanomaterial obtained demonstrated the efficiency of the method. The properties (dimension) of nanoparticles thereby justifying further applied research and development of advanced material for sustainable water management. XRD spectrum shows rhombohedral (hexagonal) structure of Al2O3 annealed at 500^{o} C. The X-ray diffraction data revealed the average size of the spherical Al_2O_3-NPs as 70nm. The UV–vis absorption show the small band gap is found to be 2.65 eV.

V. ACKNOWLEDGMENT

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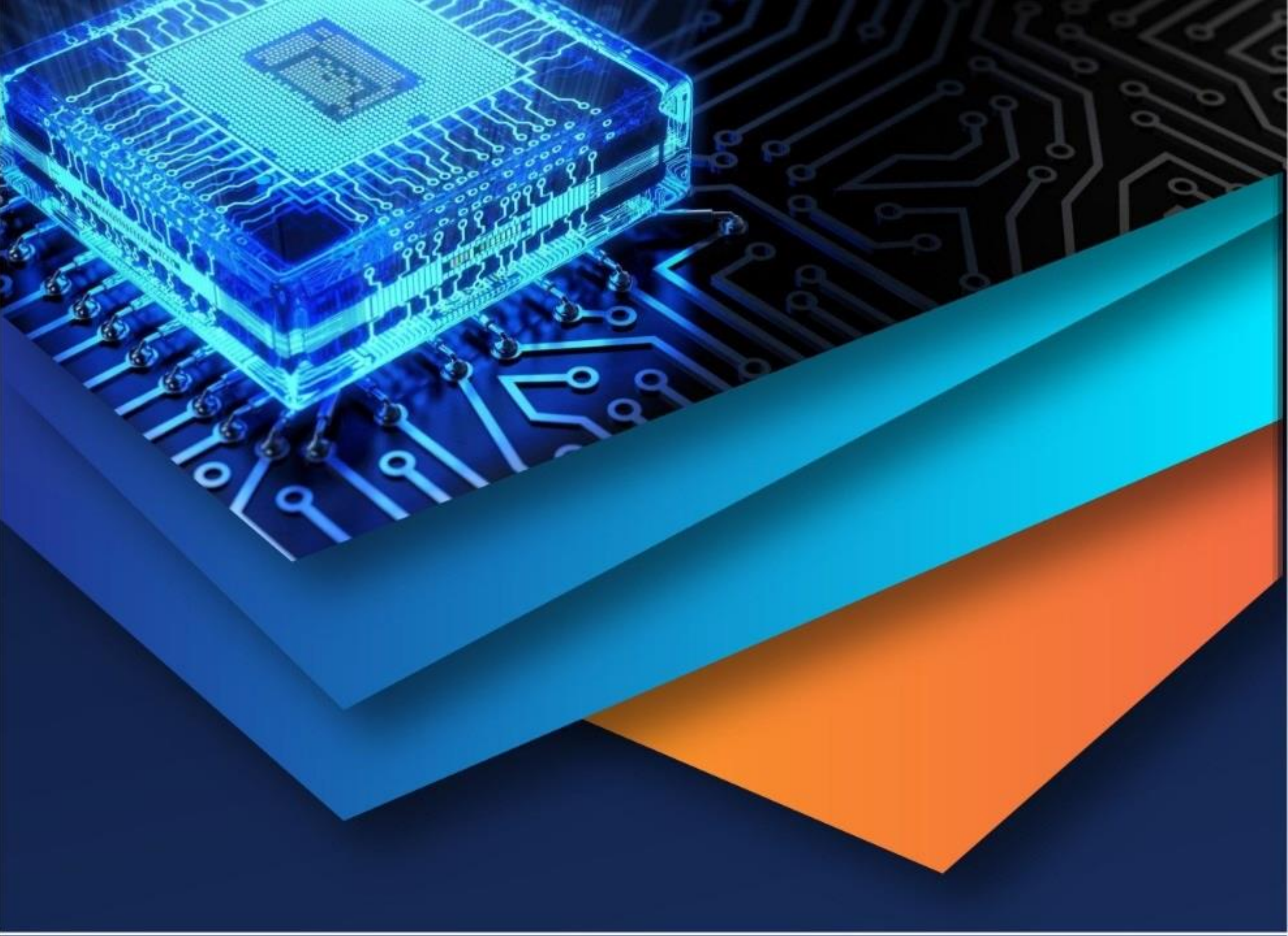

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