

Characterizations and Environmental Applications of Various Synthesized Aluminum Oxide Nanoparticles Using Sol-Gel Technique: A Review

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Abstract: Nanotechnology is a broad new area of modern science and provides superior products with unique fundamental properties. Nanomaterials are also called nanoparticles (NPs) or nanopowders, are known as the fundamental components of nanotechnology. Aluminum oxide (Al_2O_3), generally known as alumina is proving to be one of the most commonly studied metal oxide NPs due to its higher chemical, thermal, and structural properties. The synthesis of alumina NPs with different phases has been recently developed. In this mini-review, the recent proposed sol-gel processes for alumina NPs, and their characterizations with various techniques are emphatically reported. It also aims to overview its environmental applications for the treatment of various chemical pollutants from wastewaters.

Keywords: Alumina Nanoparticles, Sol-Gel Method, Adsorbent, Pollutants Treatment

1. Introduction

Scientists have defined nanotechnology or nanoscience as dealing with tiny dimension materials, control, and understanding of natural and synthetic materials at the nanoscale (1-100 nm). Nanotechnology is also about a broad new area of science and giving wonderful products with unique chemical, physical, optical, thermal, biological and, mechanical properties (Kulkarni & Kulkarni, 2015; Roy, Ghosh, & Sarkar, 2017). Nanomaterials are also called nanoparticles (NPs) or nanopowders, are known as the fundamental components of nanotechnology (Hasan, 2015). NPs are also nano-objects with multi-dimensions in the mentioned scale and crystalline or amorphous (Kumar & Kumbhat, 2016).

Based on chemical structure, nanoparticles can be classified into various material forms, including metal or metal oxide nanoparticles, organic/polymeric particulate nanoparticles, and carbon-based nanoparticles (Hami, Abbas, Eltayef, & Mahdi, 2020). The synthesized nanoparticles from various elements oxide such as an Al_2O_3 , ZrO_2 , Fe_2O_3 , TiO_2 , ZnO , SiO_2 , and CeO_2 have been widely manufactured owing to their distinctive properties, and widespread applications (Kumar & Kumbhat, 2016).

Aluminum oxide (Al_2O_3), generally known as alumina, is an amphoteric oxide and inorganic ceramic material. It is also one of the most commonly studied metal oxide due to its higher chemical, thermal, and structural properties. Amorphous and crystalline are two available primary forms of alumina (Bhushan, 2017; Li et al., 2020).

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Several crystallographic forms of alumina nanoparticles, including α , δ , θ , and γ phase (Figure 1), are available with the following shifting material properties and synthesis method (Carstens & Enke, 2019). These crystalline structures have different properties and applications (Nordell, 2011; Singh, Srivastava, Mandal, & Mall, 2014).

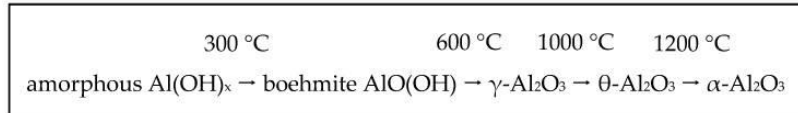


Figure 1: shows alumina phase transformation (Carstens & Enke, 2019).

In this mini-review paper, a sol-gel method, known as a chemical method, was focused on synthesizing alumina nanoparticles. This study also provides a brief discussion about the alumina nanomaterials application regarding environmental treatment from chemical pollutants.

2. Synthesis of Al₂O₃ NPs

Top-down and bottom-up approaches (Figure 2) can broadly applied to synthesize alumina NPs. Various methods are widely reported in the literature to synthesize Al₂O₃ NPs through top-down and bottom-up approaches (Behera, Sarkar, & Bhattacharyya, 2016; Ziva, Suryana, Kurniadianti, Nandiyanto, & Kurniawan, 2021).

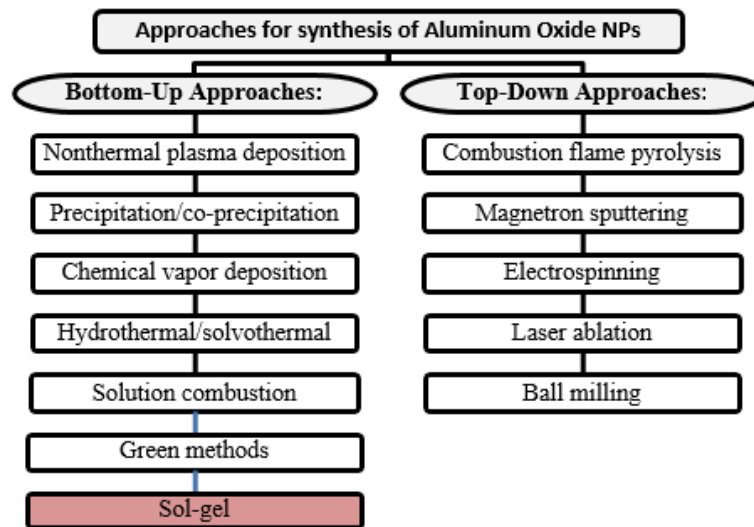


Figure 2: shows various technique for the synthesis of alumina NPs via top-down and bottom-up approaches

2.1 Sol-Gel Method

The sol-gel method is well adapted for synthesis metal oxide NPs and generally involved five main steps (Figure 3). The steps method include hydrolysis of precursors, polycondensation, aging, drying process by various routes, and thermal calcination/decomposition (Kulkarni & Kulkarni, 2015; Kumar & Kumbhat, 2016; Parashar, Shukla, & Singh, 2020) as follows:

2.1.1 Hydrolysis

Starting chemicals such as metal salts or alkoxides were used as a precursor to form a stable solution (sol) in water or alcohols by hydrolysis reaction. The precursors' hydrolysis can be rapidly enhanced and completed due to using acids (e.g., CH₃COOH, HF) or bases (e.g., NH₃, NaOH) as catalysts. MOR is used as a formula of metal alkoxides, where M and R means a cation metal and alkyl group (C_nH_{2n+1}). In this step, the –OR group is replaced with an –OH group (Equation 1).



2.1.2 Condensation

After the hydrolysis, the produced sol begins to form gel through condensation and adjacent molecules' polymerization (Equation 2). In this step, metal oxide linkages are formed due to eliminating water or alcohol molecules. Thus, it causes to grow new particles of polymeric networks to colloidal dimensions in the liquid state. During condensation, olation and oxolation are occurred and two main successive processes to form a hydroxyl (–OH–) bridge and an oxo (–O–) bridge between two metal centers (as metal–hydroxy-metal bonds and metal–oxometal bonds), respectively. A polycondensation reaction obtains gelation resulting due to an increase in the viscosity of the solutions. The used alkoxide precursor and pH of the medium are mainly responsible for forming different sizes and the cross-linking inside the colloidal particles (Parashar et al., 2020).



2.1.3 Aging

During aging process, the gel structure and property are continuous to modify due to further polycondensation reactions. The localized solution leads to re-precipitating the gel network, occurs phase transformation from the gel into a solid mass, expels the solvent from the gel pores, reduces porosity, and grows the thickness between colloidal particles.

2.1.4 Drying

The drying process is required to remove water molecules and other volatile organic compounds, which may disturb the formed gel network. Several drying processes, such as thermal/atmospheric drying, freeze-drying, and supercritical drying (Figure 3), can be individually used based on different gel structure implications (Parashar et al., 2020).

2.1.5 Thermal Calcination

A process of heating final product of the gel, which is called thermal treatment/calcination, under controlled temperature, is lastly carried out to drive the water molecules (dehydration) and residues from the desired sample. The calcination temperature is selected as a useful parameter to control the desired material's density and pore size (Parashar et al., 2020).

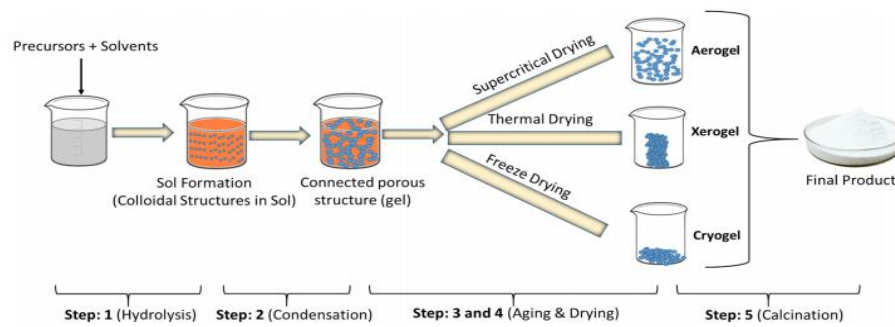


Figure 3: shows main five steps involved in the sol-gel process (Parashar et al., 2020)

2.2 Literature Review on Synthesis of Alumina NPs via Sol-Gel Method

In the last decade, several studies have documented the formation of Al_2O_3 NPs via the sol-gel method. Several researchers synthesized alumina powder as $\alpha\text{-Al}_2\text{O}_3$ phase at the nanometric scale due to calcination step at high temperature (Table 1). Mirjalili, Hasmaliza, and Abdullah (2010) synthesized $\alpha\text{-Al}_2\text{O}_3$ NPs by sol-gel method due to utilizing aqueous solutions of aluminum isopropoxide and nitrate (0.5 M). Obtained results verified that stirring time and types of surface-active agents are essential to control the degree of aggregation, particle size (20–30 nm), and the shape. Rogoan, Andronescu, Ghitulica, and Vasile (2011) also suggested the synthesis of nano alumina particles with superior properties due to using two different chemical structures of precursors, including AlCl_3 and aluminum triisopropylate ($\text{C}_3\text{H}_7\text{O}$)₃Al. Acetylacetone, as a relatively cheap precursor, was also proposed instead of the usual alkoxides. Results verified that the proposed procedure could provide α -alumina with a 5 nm scale (Ashrafi, Babanejad, & Ghasimi, 2015). In another work, a novel method was proposed to synthesis $\alpha\text{-Al}_2\text{O}_3$ NPs (28 nm at $>1000^\circ\text{C}$) by the non-surfactant templating (M Farahmandjou & Golabiyani, 2016). Dhawale, Khobragade, and Kulkarni (2018) also declared that aluminum oxide nanoparticles were well prepared via the sol-gel method. They stated that a good particle size (25 nm) with a rhombohedral structure was obtained.

Table 1: shows summarized studies on the synthesized of $\alpha\text{-Al}_2\text{O}_3$ NPs by sol-gel method

Precursors	Synthesis conditions	Properties	References
$\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, SDBS, Na(AOT))	Stirring: different periods. Evaporation: heating and stirring up to 60°C Drying: at 90°C Calcination: 1100 to 1200°C	The better dispersion, spherical shape, finer $\alpha\text{-Al}_2\text{O}_3$ NPs (20–30 nm) can be achieved (48 h) stirring with SDBS surfactant	(Mirjalili et al., 2010)

AlCl ₃ , (C ₃ H ₇ O) ₃ Al, NH ₃ , ethanol	Maturate for 30 hrs at room temperature Shaking: at 90 °C for 10 h. Drying: 100 °C, 24 h. Calcination: 1000 to 1200 °C (2 h)	Alumina powder at the nanometric scale (α -Al ₂ O ₃), which has superior properties as compared to the powders obtained in larger particle sizes, can be obtained	(Rogojan et al., 2011)
AlCl ₃ .6H ₂ O, acetylacetone, ethanol	Stirring: 2.5-3 hours Drying: 120 °C for about 48 h Calcination: at 1000 °C for 3 h	α -alumina (5 nm scale) was produced due to using cheaper chemicals and lower temperature compared to other methods.	(Ashrafi et al., 2015)
Al(NO ₃) ₃ .9H ₂ O ethanol	Stirring: at 80 °C Maintaining pH: 2-3 Evaporation: for 3 h & cooled Calcination: at 500 °C, then > 1000 °C 5 h	A novel (γ -Al ₂ O ₃ at 500 °C & α -Al ₂ O ₃ at >1000 °C) NPs (28 nm) were successfully obtained by the non- surfactant templating.	(M Farahmandjou & Golabiyani, 2016)
AlCl ₃ , NH ₃ , PVA	Stirring: continuously using a magnetic stirrer Drying: 100 °C (24 h) Calcination: 1200 °C, (4 h)	The average size of α -Al ₂ O ₃ NPs (25 nm) were obtained with having a rhombohedral structure	(Dhawale et al., 2018)

Na(AOT); sodium bis-2-Ethylhexyl sulfosuccinate, SDBS; 1,3-benzene disulfonic acid disodium salt, PVA; polyvinyl alcohol

Many works have also mentioned the preparation of δ -Al₂O₃ phase NPs (Table 2) through the sol-gel approach due to low calcination temperature. Poursani, Nilchi, Hassani, Shariat, and Nouri (2015) also proposed nano- γ -Al₂O₃ utilizing the sol-gel approach in organic solution (Figure 4).

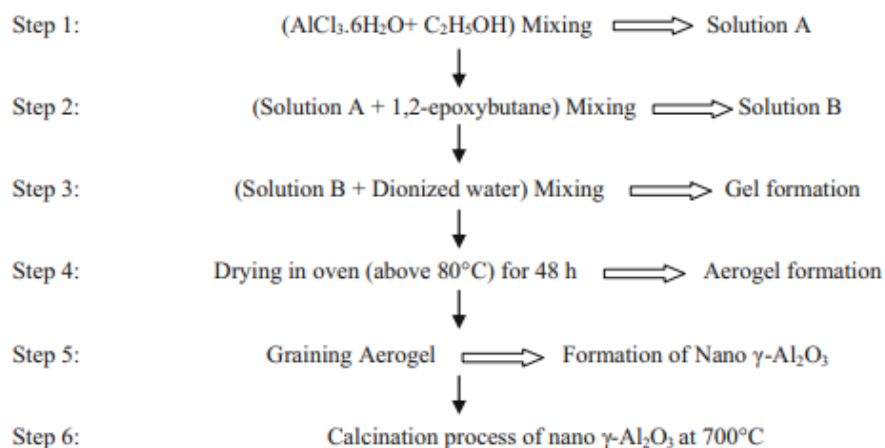


Figure 4: illustrates the synthesis process of γ -Al₂O₃ based on the sol-gel approach (Poursani et al., 2015).

According to suggested method by Thabet and Ismaiel (2016), γ -Al₂O₃ NPs can be easily obtained by using simple equipment set up at a lower temperature. The γ -alumina with a suitable pore size and relatively high specific surface area were also synthesized (Siahpoosh, Salahi, Hessari, & Mobasherpour, 2016). Tanna, Chaudhary, Gandhare, and Juneja (2016) also stated an efficient, alternate, and safer method for the preparation of γ -Al₂O₃ NPs in the existence of aluminum nitrate as a precursor, aqueous ammonia as the precipitating agent, and polyvinyl pyrrolidone. Sinkó (2017) was recently synthesized highly porous Al₂O₃ by various low energy consumption techniques. Various drying methods were applied, such as atmospheric pressure, under supercritical conditions, and vacuum. Siahpoosh, Salahi, Hessari, and Mobasherpour (2017) prepared porous γ -alumina NPs utilizing a facile sol-gel process. Compared with tested commercial γ -alumina NPs, the obtained results verified that the final active product has a specific surface area and high porosity due to applying various solvents. Esmailirad, Zabihi, Shayegan, and Khorasheh (2017) reported a modest method for preparation γ -alumina NPs with high thermal stability and high surface area.

Abdellah, Abdelfattah, Diab, and Saad (2018) prepared gamma aluminum oxide NPs from Al(NO₃)₃ and CH₃COOH through a citrate sol-gel method. They mentioned that changing the calcination temperature can cause to obtain several phases such as γ -Al₂O₃, α/γ -Al₂O₃, and amorphous NPs. A novel method for synthesizing mesoporous γ -Al₂O₃ was suggested using an uncommon precursor of Al polyoxohydroxide (Segal et al., 2018). In the existence of d-glucose, mesoporous alumina was achieved from Al oxidation in an alkaline solution mixed with ethanol. According to a recent study, alumina NPs were synthesized with porosities in the meso-range, same appearance, large masses, and good particle size distribution using Al cans (Fakhimi, Najafi, & Khalaj, 2020). In another study (Figure 4), the uniform Bead-Shaped Mesoporous Alumina was well synthesized with nano sizes and good mesoporosity (Kim et al., 2020).

Table 2: shows summarized studies on the synthesized of γ -Al₂O₃ NPs via the sol-gel approach

Precursors	Synthesis conditions	Properties of Al ₂ O ₃ NPs	References
AlCl ₃ .6H ₂ O, 1,2-epoxybutane, ethanol	Drying: in the oven (above 80 °C, 48 h) Calcination: 700 °C, 5 h	Formation of nano γ -Al ₂ O ₃ shows a good capability adsorption of heavy metals like Cr and Pb	(Poursani et al., 2015)
Aluminum oxide, Urea, NH ₄ OH Formaldehyde, ethylene glycol,	pH: 8 – 9 & Stirring for 2 h (70 °C) to achieve a gel Drying: overnight, calcination: (500 °C, 3 h)	γ -Al ₂ O ₃ NPs can be synthesized at a lower temp. with a simple instruments setup.	(Thabet & Ismaiel, 2016)
Aluminum isopropoxide, acetic acid, tert-butanol	Stirring: 150 rpm at room temperature for 3 h	γ -alumina NPs with narrow pore size distribution (1.09 cm ³ /g) and relatively high	(Siahpoosh et al., 2016)

	Drying: in the oven (120 °C) 6 h in the flow of air Calcination: at 600 °C (2 °C/min rate) for 6 h	active surface area (351 m ² /g) were obtained	
Aluminum nitrate, ammonia, PVP	Stirring: room temperature (12 h) Drying: at 120 °C Calcination: (500 °C, 2 h (rate 10 °C /min))	The green synthesis of aromatic organic compounds (dihydropyrimidinones derivative) in solvent-free condition can be contributed by γ -alumina NPs	(Tanna et al., 2016)
Aluminum isopropoxide, D.W., acetic acid, 1-butanol, tert-butanol, 2-propanol.	Organic solvents dissolving, Stirring for 3 h at 150 rpm Drying: oven (120 °C, 6 h in the flow of air Calcination: (600 °C (rate 2°C /min) 6 hours)	γ -alumina NPs achieved (<10 nm) sizes, high active surface area, suitable pore size distribution, large pore volume, which is desirable for the removal of metal ions	(Siahpoosh et al., 2017)
(Al(NO ₃) ₃ ·9H ₂ O), ethyl acetate, isopropanol, acetic acid, propanol, citric acid	Dissolving: the starting materials based on product Drying: varies based on the final products Calcination: at 500 °C to form the final structure	This method provides the ceramics with a mixture of crystalline and amorphous (γ -Al ₂ O ₃) characters with good adsorption ability and high specific surface area.	(Sinkó, 2017)
(C ₄ H ₉) ₃ AlO ₃ , ethanol, ammonia, methanol	Aging: room temperature, 24 h Drying: (100 °C, 24 h) Calcination: at 1000 °C under N ₂ flow for 2 h	The prepared γ -Alumina NPs have higher catalytic activity compared with the same catalyst supported on other γ -alumina supports.	(Esmacilrad et al., 2017)
Al(NO ₃) ₃ ·9H ₂ O, citric acid	Stirring: (60 °C, 10 min & 80 °C, 1 h) Drying: 200 °C, 2 h Calcination: at 500, 600, 800 & 1000 °C	γ -Al ₂ O ₃ NPs with a particle size (<8 nm) were prepared, can be effectively utilized for the adsorption of Cr(VI) from waters sample	(Abdellah et al., 2018)
Al powder, ethanol, water, d-glucose	Sonicated: in an ultrasonic bath (125 W/ 20 kHz) Drying: in an oven at 70 °C Calcination: at 300, 400, 700, and 900 °C for 2 h	Synthesis mesoporous γ -Al ₂ O ₃ (at 400 °C, lower temperature compared to other methods) with narrow pore size & high specific surface area (282 m ² /g)	(Segal et al., 2018)
(Al(NO ₃) ₃ ·9H ₂ O), NH ₄ OH, chitosan polymer, acetic acid	Dissolving: in acidic medium & Stirring: for 12 h Calcination: (600 °C, air flow, 2 h)	The uniform Bead-Shaped Mesoporous Alumina with organized & tunable particle size (1-20 nm) prepared	(Kim et al., 2020)

<p>$\text{Al}(\text{OC}_3\text{H}_7)_3$ (prepared from Al cans), CH_3COOH, isopropyl alcohol, NaOH</p>	<p>Stirring: at 80 °C Aging: (25, 60, 80°C and period of 12, 24 & 48 h) Calcination: the alumina phase is 500 °C for 1 h</p>	<p>Nanoparticles were created with porosities in the meso- range, same appearance, large masses, and the good (20-40 nm) particle size distribution.</p>	<p>(Fakhimi et al., 2020)</p>
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3. Characterizations of Al_2O_3 NPs

Many techniques have been used to characterize the synthesis of aluminum oxide NPs. Detailed information on the most commonly used techniques was documented in this review.

3.1 The Transmission Electron Microscopy (TEM)

Transmission electron microscopy (TEM) is utilized to investigate the particles actual size, morphologies of the particles, distribution of the crystallites, and their growth pattern (Saud, Majdi, & Saud, 2019). Figure 5 illustrates a TEM image of the produced sphere-like shape of alumina NPs with sizes (20-50 nm). (Majid Farahmandjou & Golabiyani, 2019).

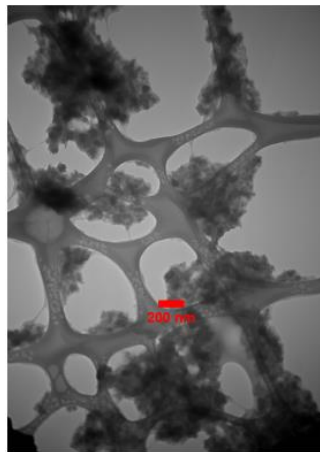


Figure 5: illustrates the TEM graph of the prepared alumina NPs (Majid Farahmandjou & Golabiyani, 2019).

3.2 Field Emission-Scanning Electron Microscopy (FE-SEM)

SEM analysis is also applied to investigate the materials morphological. This technique can provide clear information on any morphological changes in final products due to changing different parameters, such as calcination temperature (Majid Farahmandjou & Golabiyani, 2019). This technique can clearly explain the morphology of various phases of the desired NPs (including $\gamma\text{-Al}_2\text{O}_3$ and $\alpha\text{-Al}_2\text{O}_3$) at different temperatures. According to the obtained results in the literature, the calcination of pure Al_2O_3 powder (γ -alumina) at 800 °C and (α -alumina) 1200 °C can provide the individual products morphology which improved shapes from spherical to vermiform respectively (Saud et al., 2019).

Identification of different morphology of the synthesized γ and α -alumina NPs at different calcination temperatures are illustrated in the following SEM micrographs (Figure 6).

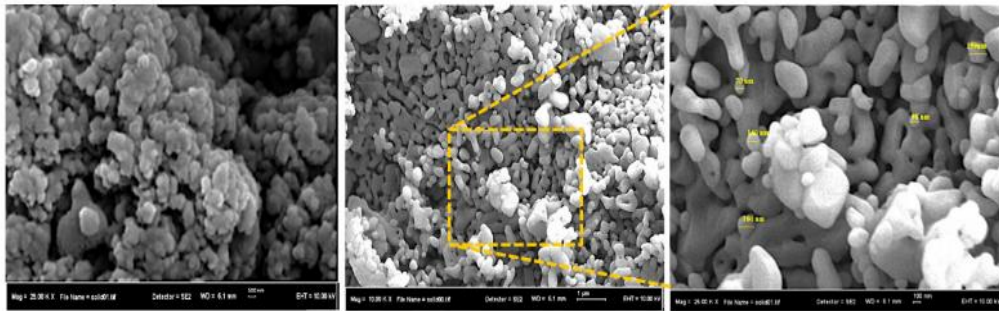


Figure 6: represents the SEM micrographs of Al_2O_3 powder calcined at various temperatures: a) 800 °C; and b,c) 1200 °C (Saud et al., 2019).

3.3 The X-ray Diffractometer (XRD)

The X-ray diffractometer (XRD) is widely utilized to identify the produced samples crystalline properties at room temperature. This technique was successfully preferred and applied to determine the Al_2O_3 composition during the starting material, intermediate changes, and final product. Besides, identifying the alumina phases can be easily determined due to applying this machine (Saud et al., 2019). At room temperature, the XRD machine equipped with a $\text{CuK}\alpha$ radiation anode ($\lambda = 0.15405$ nm) was applied to identify the phases of the Al_2O_3 powder. The measurement analysis was carried out under the following preferred conditions: a sweep of scanning range (10-80° at an angle 2θ) with scanning speed 5 °/min, and the used power source 40 kV/30 Ma (Saud et al., 2019; Toledo, Santoyo, Sánchez, & Rosales, 2018).

Majid Farahmandjou and Golabiyani (2019) recently used the XRD technique to specify the structure, size, and optical properties of the as-synthesis and annealed alumina NPs. Obtained results verified that this technique at 40Kv was successfully applied to detect the crystalline phase and size of the produced alumina. Figure 7 represents XRD patterns of the powder specification as-synthesis and annealed alumina NPs (before and after heat treatment at 1000 °C) during this assessment (Majid Farahmandjou & Golabiyani, 2019). Obtained results showed that the broad picks of γ - Al_2O_3 phase were presented with increasing temperature. A phase transformation from γ to α alumina occurs at a higher calcination temperature of more than 1000 °C. The α - Al_2O_3 phase is well known as the only produced phase exhibits for the powder calcined at elevated temperature (>1000 °C). The presented picks denoted the (012), (104), (110), (113), (024), (116), (018), (300) and (119) (Figure 7) of a rhombohedral structure of α - Al_2O_3 is detected utilizing the standard data (Majid Farahmandjou & Golabiyani, 2019).

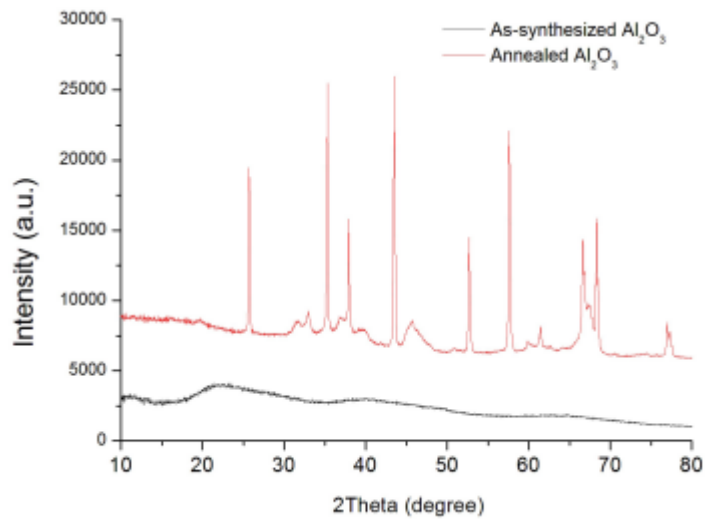


Figure 7: illustrates the XRD pattern of as-synthesized and annealed alumina at 1000 °C (Majid Farahmandjou & Golabiyani, 2019)

3.4 Fourier Transform Infrared (FTIR) Spectroscopy

FTIR technique equipped with KBr (high purity 99%) disk method in a frequency range of 4000 - 400 cm^{-1} is utilized to measure the vibrational spectroscopy of investigated materials at room temperature (Saud et al., 2019). Thus, the obtained FTIR spectra of alumina powder can assess the presented functional groups. The main absorption peaks are presented in Figure 8 and recorded in Table 3 (Saud et al., 2019).

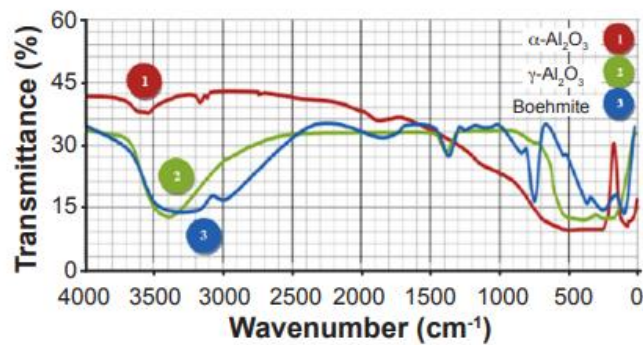


Figure 8: represents the recorded IR spectra of various forms of Al_2O_3 powder (1200 °C, 2hrs) (Saud et al., 2019).

Table 3: shows peak bond attributions of alumina powder.

Band	Peak position (cm^{-1})
O-Al bending vibration	~ 447, ~ 594
The stretching vibration (O-H) group related to Al-OH structure	~ 3457

Stretching vibration of (Al-O) group band related to α - Al_2O_3	640
Stretching vibration, for C-H band	2866, 2924
Physically adsorbed water molecules	1629

3.5 The Energy Dispersive Spectrometer (EDS)

The energy dispersive spectroscopy (EDS) instrument is also utilized to assess the purity of products and analyze the composition of the synthesized alumina NPs. The O and Al elemental analysis of the alumina sample can be carried out using this technique (Majid Farahmandjou & Golabiyani, 2019). The obtained EDS spectrum (15 Kv) at room temperature for Al_2O_3 NPs powder that was calcined at $1200\text{ }^\circ\text{C}$ is shown in Figure 9 (Saud et al., 2019). Obtained results (Figure 9) via EDS technique was also confirmed the chemical purity of the synthesized α - Al_2O_3 from any other components.

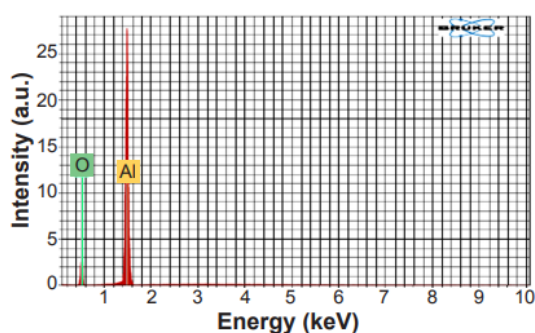


Figure 9: displays the EDS spectrum for Al_2O_3 NPs calcined ($1200\text{ }^\circ\text{C}$, 2 hrs) (Saud et al., 2019).

3.6 Thermal analysis TGA/DTGA-DTA

The thermal analysis approaches such as thermogravimetric analysis (TGA) and differential thermal analysis (DTGA-DTA) have been utilized to assess the thermal stability of produced samples (temp. 30 - $1000\text{ }^\circ\text{C}$, rate; $10\text{ }^\circ\text{C}/\text{min}$, airflow $100\text{ mL}/\text{min}$) (Toledo et al., 2018). The recorded data related to the instrument's analysis for chemicals such as Al_2O_3 -TG and Al_2O_3 -AR are exhibited in Figures 10a and 10b. The TG and AR denote the formed powders from Technical Grade and Analytical Reagent starting materials, respectively. Subsequent shifting of α , δ , θ , and γ phases thermal transformations cannot be explained by these techniques due to not occur weight loss. According to recorded data in the literature, the observed endothermic peak (Figure 10b) in the DTA curve ($950\text{ }^\circ\text{C}$) is attributed to θ - Al_2O_3 formation (Toledo et al., 2018).

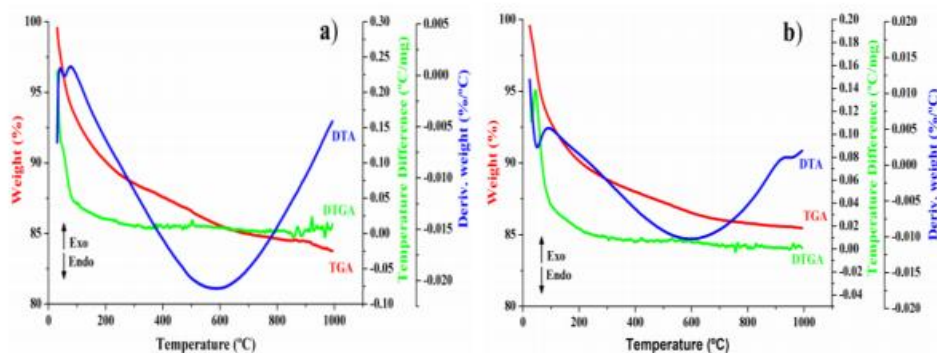


Figure 10: shows DTA and TGA/DTGA recorded curves of a) Al_2O_3 -TG material and b) Al_2O_3 -AR material (Toledo et al., 2018).

3.7 N_2 Physisorption

The N_2 adsorption/desorption isotherms process was also applied to determine products textural properties through appropriate isotherms. Brunauer–Emmett–Teller (BET) and The Barret–Joyner–Halenda (BJH) approaches can be applied to calculate specific/active surface area and accessible pore size/volume distribution, respectively. The N_2 adsorption/desorption isotherms process is applied to detect specific surface area, available pore-volume, and accessible pore size. Before examining the test at about 200 °C, the analyzed product was degassed for about 3 hours (Saud et al., 2019). The N_2 adsorption isotherms process was calculated at liquid nitrogen (77 K) and suitable pressures (under a vacuum instrument, 10⁻⁶-1.0 of P/P₀) to identify the available surface area (Saud et al., 2019; Toledo et al., 2018).

4. Applications of Al_2O_3 NPs for Environmental Issues

Alumina NPs have been commonly utilized as fillers, abrasive particles, precision optical constituents, coating materials, ceramic filtration membranes, flame retardant, and refractory materials. It is also used to prepare absorbent materials, crucibles, thermocouples, catalysts, chemical synthesis, and transparent ceramics/lamps (Kumar & Kumbhat, 2016; Nordell, 2011).

Metal oxides are the most commonly applied surfaces for removing chemical pollutants in the environment. Al_2O_3 is also one of the most common adsorbent NPs widely used to remove several contaminants in the environment.

4.1 Water Purification and Remediation

Several techniques, including adsorption, advanced oxidation processes, coagulation, filtration with coagulation, reverse osmosis, ion exchange, ozonation, and precipitation, have been commonly utilized to treat chemical pollutants from wastewater and polluted water (Rashed, 2013). The adsorption process has been selected to be superior and more potential than the other suggested

techniques among the possible techniques. This method is well-recognized as the potential separation process for water remediation owing to its designable, readily operatable, flexibility, low cost, and tactlessness to toxic chemicals/pollutants. Adsorption is a surface phenomenon with several mechanisms for removing various chemicals/pollutants (Table 4) in the environment.

4.1.1 Removal of Toxic Heavy Metal

Heavy metals (HMs) are the most dangerous pollutant in the environment. They can cause several problems to human health because of their toxicity, oxidation stress, non-biodegradable, accumulation, and persistent nature. Due to the continuing industrial growth and activities, the widespread of toxic HMs like arsenic (As), cadmium (Cd), mercury (Hg), lead (Pb), chromium (Cr), copper (Cu), nickel (Ni), and zinc (Zn) has effectively become significant wastewater pollutants and freshwater resources (Azimi, Azari, Rezakazemi, & Ansarpour, 2017; Khalid, Ali, & Almashhadany, 2020). In recent years, several studies were carried out to enhance the sorption capacities of Al_2O_3 NPs towards the verity of toxic HMs ions (Table 4) in water/wastewater (Ghosh, Prabhakar, & Samadder, 2019; Mahdavi, Jalali, & Afkhami, 2013; Poursani et al., 2015).

4.1.2 Removal of Organic Dyes

Most of the chemical dyes used in many industries like cosmetics, food coloring, textile, and paper industries are known as carcinogenic organic pollutants. The residues of chemical dyes in the environment can cause harm and have several dangerous side effects. Al_2O_3 NPs had been efficiently applied as an adsorbent in water/wastewater solution for the removal of residues of several dyes (Table 4), including Acid Orange-7 (Khosla, Kaur, & Dave, 2013), Congo Red (Zhang et al., 2019), Eosin Yellow (Thabet & Ismaiel, 2016), Eriochrom Black T (Abbas, Hami, Mahdi, & Waheb, 2020), Methylene Blue (Dhawale et al., 2018), Malachite Green (Aazza, Moussout, Marzouk, & Ahlafi, 2017), Orange G (Banerjee, Dubey, Gautam, Chattopadhyaya, & Sharma, 2019) and many others.

4.1.3 Removal of Pesticides

Pesticides are known as agrochemicals or mixtures of widely used substances in public health protection programs, urban green areas, and agricultural lands. They are used to kill undesirable organisms, protect humans from various vector-borne diseases, and plants from pests and diseases (Nicolopoulou-Stamati, Maipas, Kotampasi, Stamatis, & Hens, 2016). Based on target organisms, pesticides can be categorized into three main types: insecticides, herbicides, and fungicides. Their residues can cause many environmental effects, cause severe problems in dairy animals, and induce potentially adverse health effects on humans bodies (Rizzati, Briand, Guillou, & Gamet-Payraastre, 2016). According to a recent study by Mukherjee et al. (2019), the alumina NPs were

preferentially applied to remove many pesticides in aqueous solutions with high percentages (Table 4).

4.1.4 Removal of Different Hazardous Compound

Several studies for the removal of different hazardous compounds like cyanide (Iakovleva et al., 2018), phenols (Asmaly et al., 2016), formaldehyde (Afkhami, Bagheri, & Madrakian, 2011), polychlorinated biphenyls (Mobasser & Taha, 2015), oil spill (Franco, Cortés, & Nassar, 2014), nitrate (Manikandan et al., 2019), phosphates (Xie, Lin, Li, Wu, & Kong, 2015), and fluoride (Changmai, Priyesh, & Purkait, 2017) compounds in aqueous solution have been investigated and removed due to using Al_2O_3 nanoparticles as adsorbent (Table 4).

4.1.5 Removal of Pharmaceuticals

Al_2O_3 NPs are also used for the treatment of wastewater from the residue of several pharmaceutical products (Table 4). Tetracycline family, one of the common antibiotics, is usually utilized by a human to treat several organ infections. Tetracycline derivatives, including tetracycline hydrochloride, chlortetracycline hydrochloride, and oxytetracycline hydrochloride, were effectively removed from wastewater using alumina NPs adsorbent (Y. Chen et al., 2018).

4.2 Treatment of Airborne Pollutants

Airborne pollutants that contribute heavily to climate change can generally originate from sources of industrials, transportations, and power plants. Various methods can be used to remove harmful pollutants from the atmosphere such as adsorption by liquids/solids, catalysis, condensation, cryogenic distillation, membrane purification, and oxidation (Cramer & Cole, 2017). Based on the adsorption process (Table 4), alumina NPs have been applied for the treatment of gaseous pollutants/chemicals.

4.3 Biological Activities

The Al_2O_3 NPs have diverse biomedical applications due to their favorable low cost, optical, physicochemical, and structural features (Hassanpour et al., 2018). Thus, their particles were also examined for antimicrobial (Sikora, Augustyniak, Cendrowski, Nawrotek, & Mijowska, 2018), antibacterial (Manikandan et al., 2019), anticancer (Sun et al., 2010), antifungal (Jalal et al., 2016), anti-inflammatory, and antioxidant (El-Hussainy, Hussein, Abdel-Aziz, & El-Mehasseb, 2016) activities besides their numerous other applications in various aspects of the biological field (Sadiq, Chowdhury, Chandrasekaran, & Mukherjee, 2009).

The documented maximum capacities (mg/g) of the alumina NPs as adsorbents are shown in the Table 4 towards various chemical pollutants in the environment.

Table 4: clarifies the maximum adsorption capacity (mg/g) of the alumina as adsorbent towards the removal of various chemical pollutants in aqueous systems.

Groups	Chemical Pollutants (Adsorbates)	Maximum Adsorption Capacity (mg/g)	References
Toxic HMs	Ni(II)	35.9	(Mahdavi et al., 2013)
	Pb(II)	41.2	
	Cu(II)	47.9	
	Cd(II)	118.9	
	Cr(VI)	13.3	(Poursani et al., 2015)
	As(V)	1.00	(Ghosh et al., 2019)
	As(III)	0.769	
Organic dyes	Acid Orange-7	97.6	(Khosla et al., 2013)
	Congo Red	465.8	(Zhang et al., 2019)
	Eosin Yellow	47.78	(Thabet & Ismaiel, 2016)
	Eriochrom Black T	3.831	(Abbas et al., 2020)
	Methylene Blue	23.9	(Dhawale et al., 2018)
	Malachite Green	13.64	(Aazza et al., 2017)
	Orange G	93.3	(Banerjee et al., 2019)
Pesticides	Mean of 42 various pesticides	0.12	(Mukherjee et al., 2019)
The different hazardous compound in water	Cyanide	91.0	(Iakovleva et al., 2018)
	Phenols	2.105	(Asmaly et al., 2016)
	Formaldehyde	411.83	(Afkhami et al., 2011)
	Polychlorinated biphenyls	6.66	(Mobasser & Taha, 2015)
	Nitrate	94.0	(Manikandan et al., 2019)
	Phosphates	46.95	(Xie et al., 2015)
	fluoride	3.82	(Changmai et al., 2017)
	Acetaldehyde	395	(Sinkó, 2017)
	Acetone	613	
	Benzene	158	
	Methyl ethyl ketone	11	
Toluene	169		
Pharmaceuticals (Antibiotics)	Tetracycline hydrochloride	1.618	(Y. Chen et al., 2018)
	Chlortetracycline hydrochloride	1.814	
	Oxytetracycline hydrochloride	1.696	
Airborne pollutants	Ammonia	66.2	(Kim et al., 2020)
	Formaldehyde (as VOC)	16.71	(D. Chen, Qu, Sun, & Wang, 2014)
	Carbon monoxide	111.16	(Mozaffari, Mirzahosseini, Sari, & Aval, 2020)
	Sulfur dioxide	7200	(Z.-M. Wang, 2017)

	Hydrogen sulfide	52.0	(Tajizadegan, Rashidzadeh, Jafari, & Ebrahimi-Kahrizsangi, 2013)
	Nitrogen oxides	47.0	(Y. Wang et al., 2020)

VOC; Volatile organic compounds

5. Discussion

The sol-gel method is more preferable and utilized to synthesize high-quality metal oxide nanoparticles such as Al_2O_3 nanoparticles with a good texture, surface properties, different morphology, and sizes. Many advanced techniques have been used to characterize the synthesis of aluminum oxide NPs. Detailed information on the most commonly used techniques to characterize the alumina was mentioned in this review.

Recorded results from the literature showed that $\alpha\text{-Al}_2\text{O}_3$ NPs can be obtained with particle sizes more than 20nm due to applying high temperature. Controlling temperature, was selected as the main factor that selects nanoparticles to size through the sol-gel method. Thus, using high temperature for calcination in this method ($> 1000\text{ }^\circ\text{C}$) is suggested to obtain such a nano size particle more than 20 nm.

On the other hand, the obtained results from many recent studies verified that alumina NPs such as $\gamma\text{-Al}_2\text{O}_3$ phase could also be synthesized with smaller particle sizes due to controlling calcination at low temperatures. The controlling temperature at about $500\text{ }^\circ\text{C}$ selected as the effective parameters to rich this type of alumina phase through the sol-gel approach.

Results from published studies confirmed that the $\gamma\text{-Al}_2\text{O}_3$ included a larger surface area compared to the $\alpha\text{-Al}_2\text{O}_3$. Calcination temperature is responsible for controlling the surface area of the final products. High-temperature calcination is responsible for forming the agglomeration state of the formed $\alpha\text{-Al}_2\text{O}_3$ NPs. Thus, the agglomerated particles cannot allow the nitrogen gas molecules to move freely between the $\alpha\text{-Al}_2\text{O}_3$ particles.

Results verified that alumina nanopowder could be preferable, applied for the treatment of many environmental aspects and used as effective adsorbent due to its high surface area. Additionally, Al_2O_3 NPs have been applied to remove various chemical pollutants with high capacity and percentages, including toxic HMs, organic dyes, pesticides, pharmaceuticals (antibiotics), and other different hazardous compounds in water/wastewater samples. Airborne pollutants could also be removed due to applying alumina NPs as adsorbent.

6. Conclusions

Many researchers have recently studied the synthesis of alumina NPs via the use of the sol-gel approach. This approach can achieve Al₂O₃ NPs with good texture, sizes, and different morphology. The sol-gel process is preferred for synthesizing alumina NPs due to providing the desired products with a high surface area. Alumina NPs have been valuable as effective adsorbent to remove various chemical pollutants in a wide range of water/wastewater and airborne samples.

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