1. Introduction

Nanoscience has found extensive applications in diverse fields including optical switching, beam steering, image processing, and amplification [1]. In this area, aluminum-based nano adsorbents have been widely applied due to lower production cost and higher efficiency ratio than others [2]. Today, metallic oxide nanoparticles (NPs) significantly attracted attention because of their perfect physical and chemical aspects, which can be different from the bulk properties of related materials [3].

Material property of alumina has of interest because of valuable use as optical devices in high irradiation environment such as nuclear plants and space. It is known that it is superior in optical permeability and is radiation proof. Al₂O₃ is now used as infrared windows, phosphors, dosimeters, photoelectric material and devices, catalyst carriers and optical fiber materials for various applications [4]. It is noted that the Al₂O₃-based composite photocatalyst shows excellent photocatalytic activity for the degradation of organic dyes as Al₂O₃ provides a better active adsorption site [5].

According to the recorded reports, alumina with α, γ, η, κ, χ, δ, and θ crystalline phases have been detected [6]. The thermal treatment, the precursors, and their stabilizations can transform and change the phase of alumina [6]. Except for stable α-Al₂O₃, the other metastable transitional phases of Al₂O₃ can be employed as catalyst supports [7]. Due to the high stability of the γ-Al₂O₃ and having a large specific surface area, it has more interest use in the most catalytic reactions as a catalyst or catalyst substrate in the automotive and petroleum industries [8,9]. Furthermore, the γ-Al₂O₃ hybrid nanocomposite is very favorable for solar energy and solar collector heaters [10]. The introduction of γ-Al₂O₃ layer has successfully improved the detection performance of some components for solar-blind UV signals [11]. The γ-Al₂O₃ nanowires were applied for biodiesel production from cottonseed oil as well as from its waste [12].

The optical material applications are currently under development, so the production of new effective ingredients with improved properties is essential [13]. The refractive index (n) is a distinct and fundamental parameter for any optical material, and its measurement is considered as a powerful tool for exploring and selecting the materials used for optical applications [14].

Nowadays, the studies on nonlinear optical (NLO) materials with the ultrafast response, enhanced laser-induced damage threshold, larger two-photon absorption (TPA), and third-order nonlinearities have gained increasing interest due to their promising potentials in the next-generation photonic devices including optical switches [15–
properties [16,17,19–21], optical information processing [16,17,19–21], 3D optical data storage [17,19–22], optical power limiting [15,16,18–21], optical fiber communication [16,17,19–22], laser-predicated imaging, remote sensing [19–21], frequency conversion of coherent laser sources [17,22,23], optical computing [23], high-density data processing [23], nonlinear imaging [16], image manipulation [15], and color displays [17,23].

However, their features are not sufficiently efficient for these applications. The NLO materials have properties such as appropriate transparency cut-off wavelengths, good thermal stability and mechanical performance, high optical susceptibility (χ), and laser damage resistance. The exploration and design of new NLO materials led to significant research into understanding and improving the properties of such materials [16].

The interaction between light and substances is known as the optical aspects of the substances. The macroscopic and microscopic properties of the substances affected the optical properties. Moreover, optical properties are often used to probe other properties including reflection, refraction, transmission, and absorption. For an accurate explanation of the optical properties, the optical theory is important by means of some methodologies. One of the essential methodologies is obtaining the physical quantities, such as complex dielectric constant or complex refractive index of materials from the recorded spectra. Due to obtain these, least-squares refinement calculations, ellipsometry, and/or Kramers–Kronig (KK) analysis are benefit in the IR region [24]. Furthermore, the KK relationship expresses the temporal behavior of an excited material that can be seen as the result of a linear filter. KK method is faster and more reliable in a broad-spectrum range. Accurate determination of the complex refractive index of γ-Al2O3 NPs has significance for the wide variety of applications in optical sensor technology at the IR range. However, to our best knowledge, the optical constant and dielectric coefficient of γ-Al2O3 at the wavelength range of 2.5–22.0 μm have not been reported in the literature.

Herein, the linear/nonlinear optical properties of γ-Al2O3 NPs are investigated. Initially, the produced NPs are characterized. Then, the NLO coefficients of these NPs were calculated by the Z-scan method. Afterward, the Fourier-transform infrared (FTIR) spectrum of these NPs is taken. It is the relatively rapid and wide IR spectral range by the non-contact technique. Using FTIR spectrum and the KK method, the complex refractive index and dielectric coefficient are obtained.

2. Experimental details

2.1. Synthesis of γ-Al2O3 NPs

γ-Al2O3 NPs were prepared using a conventional sol–gel process. Sol–gel technology is a wet-chemical engineering method used to produce ceramic materials, glasses, films, and a wide range of materials [25]. First, aluminum nitrate nonahydrate (Al(NO3)3·9H2O) was dissolved in deionized water under magnetostirring. Then, triethanolamine (TEA, NC3H7O) and citric acid (C6H8O7) were added to the previous solution slowly and subsequently at 80°C for 1 h. By increasing the temperature up to 120°C for 1 h, the viscous gels are obtained. Finally, the dried gels were calcined at 800°C in the furnace and produced the γ-Al2O3 NPs [9]. The produced γ-Al2O3 NPs were characterized and used for the linear and nonlinear optical study.

2.2. Theory of Z-scan method

The interaction of laser light with the material can make the absorption and refraction phenomena [26]. Third-order materials play an important role in many applications of nonlinear optics. The third-order response leads to processes such as third harmonic generation and two-photon absorption, but more importantly leads to the intensity-dependent refractive index, which is the basis of most NLO switching devices. The intensity dependence of the refractive index is expressed by [27]:

\[ n(I) = n_0 + n_2 I \]  

(1)

The nonlinear absorption (NLA) is dependent according to the following equations:

\[ a(I) = a + \beta I \]  

(2)

In the above equation, \( a \) represents the linear absorption coefficient, while \( n_0 \) and \( n_2 \) show the linear and nonlinear refractive (NLR) indexes, respectively. \( \beta \) denotes the NLA coefficient and \( I \) is indicative of the laser beam intensity [28].

The sign and magnitude of the NLR index (and NLA coefficient) can be determined by Z-scan experimental setup at the far-field (and near field) with closed (open) aperture [21]. An important advantage of the Z-scan method is the possibility to separate processes related to NLA and NLR when both these processes proceed simultaneously in a sample [29].

Based on Z-scan data, the NLR index was dependent on self-focusing or self-defocusing; moreover, the NLA coefficient originated from the saturable absorption (SA) or reverse saturable absorption [30]. The experimental setup of the Z-scan apparatus is illustrated in Figure 1 [31].
A 532 nm wavelength of an Nd:YAG laser was utilized in the Z-scan tests. The sample solution was transferred to a quartz cuvette with 1 mm thickness. A lens with a 5-cm focal distance was employed to focus the incident laser beam on the quartz cuvette; the laser beam waist, \( \omega_0 \), at the focus was 32 \( \mu \)m. The peak laser intensity at the focal plane was \( I_0 = 3108.5 \) W/cm\(^2\). The obtained data were fitted to the following equation [32]:

\[
\Delta T_{p-v} = 0.406(1 - S)^{0.25} |\Delta \Phi_0| 
\]

In which \( \Delta T_{p-v} = T_p - T_v \) shows the normalized peak–valley transmittance difference, while \( \Delta \Phi_0 \) denotes the nonlinear phase of the sample at the focus. \( Z_0 \) represents the Rayleigh length and \( Z \) shows the sample position compared to the focus (\( Z = 0 \) mm). \( \lambda \) and \( P_0 \) indicate the excitation laser wavelength and the input power (50 mW), respectively, while \( S \) stands for the aperture transmittance in the absence of the sample (\( S = 0.28 \)).

The optical nonlinearity of Al\(_2\)O\(_3\) NPs was quantified by the Z-scan data [33]. The open-aperture Z-scan plot in Figure 5 can be fitted using the following equation [34]:

\[
T(z) = \ln(1 + q_0(z,t))/q_0(z,t) 
\]

In which \( q_0(z,t) = \beta_0 L_{eff} / (1 + z^2/L_{eff}^2) \), \( z_0 = k\omega_0^2 / 2 \) shows the diffraction length of the beam, and \( |z| \) denotes the normalized transmittance at \( Z \).

### 2.3. The KK theory

The optical constants of Al\(_2\)O\(_3\) NPs were determined using FTIR transmittance spectra findings combined with the KK method. The complex refractive index \( n(\sigma) = n(\sigma) + ik(\sigma) \) has the following components [35]:

\[
\begin{align*}
n(\sigma) &= \frac{1 - R(\sigma)}{1 + R(\sigma) - 2\sqrt{R(\sigma)\cos \phi(\sigma)}} \\
k(\sigma) &= \frac{-2\sqrt{R(\sigma)\sin \phi(\sigma)}}{1 + R(\sigma) - 2\sqrt{R(\sigma)\cos \phi(\sigma)}} \\
\phi(\sigma) &= -\frac{\sigma}{\pi} \int_0^1 \ln[R(\sigma) - R(\sigma')d\sigma'] \\
\end{align*}
\]

in which \( \sigma = \lambda^{-1} \).

The phase difference between the incident and reflected light is \( \phi(\sigma) \) obtained from the KK dispersion relation [36]. Several approaches have been reported for calculating \( \phi(\sigma) \) [37,38]. Using Maclaurin’s method, \( \phi(\sigma) \) is calculated more accurately by the following equation:

\[
\phi(\sigma_i) = \frac{4\sigma_i}{\pi} \Delta \sigma \times \sum \ln(\sqrt{R(\sigma)}) \left( \frac{\sigma^2 - \sigma_{i-1}^2}{\sigma_{i+1}^2 - \sigma_i^2} \right) \]

where \( \Delta \sigma = \sigma_{i+1} - \sigma_i \), and for odd (even) \( j \) values, \( i = 2, 4, \ldots, j - 1, j + 1 \), \( \sigma = 1, 3, 5, \ldots, j - 1, j + 1 \).

Moreover, the real and imaginary parts of the dielectric function can be determined by [36]:

\[
\begin{align*}
\varepsilon(\sigma) &= \varepsilon_1(\sigma) + i\varepsilon_2(\sigma) \\
\varepsilon_1(\sigma) &= n^2(\sigma) - k^2(\sigma) \\
\varepsilon_2(\sigma) &= 2n(\sigma)k(\sigma) \\
\end{align*}
\]

### 3. Results and discussion

#### 3.1. Structural properties of synthesized γ-Al\(_2\)O\(_3\) NPs

The XRD patterns are shown in Figure 2 which indicate that these NPs have a well-defined single crystal phase belonging to the cubic lattice of γ-Al\(_2\)O\(_3\) phase. The spectrum of Al\(_2\)O\(_3\) NPs exhibits peaks at 31.5°, 37.5°, 40.2°, 46.5°, 60.4°, 67.5°, and 85° which are referred to (220), (311), (222), (400), (511), (440), and (300) planes (JCPDS Card No: 00-029-0063), respectively [9]. The crystallite size has been estimated at about 8 nm by Debye Scherrer’s formula [39]:

\[
D = K\lambda / \beta \cos \theta 
\]

In the above equation, \( K \) represents the Scherrer constant (0.9), whereas \( \lambda \) is the X-ray wavelength.
Moreover, $\beta$ shows the full width at half-maximum of the reflections, and $\theta$ stands for the Bragg’s angle in radian [39].

The sample surface nature and morphology were assessed by the field emission scanning electron microscopy (FESEM) technique [21]. The FESEM micrographs of $\gamma$-Al$_2$O$_3$ NPs are shown in Figure 3. FESEM micrographs are indicating the formation of controlled size and regularly shaped NPs. The corresponding size distribution histograms of $\gamma$-Al$_2$O$_3$ NPs in Figure 4 show the presence of a monophasic homogeneous microstructure with an average particle size of 60 nm.

The optical transparency of NLO materials is a crucial asset when it is used for device applications. Ultraviolet-visible (UV-Vis) spectrum provides data about the electronic transition between molecules when it absorbs energy [23].

The UV-Vis absorption spectra of the aqueous solution of 0.1 mM concentration of $\gamma$-Al$_2$O$_3$ NPs are shown in Figure 5(a,b). The spectra show a sharp absorption edge at 280 nm. This is because of the photoexcitation of electrons from the valence band to the conduction band. The bandgap energy of the synthesized sample was 3.75 eV, which is in good agreement with other researches [40]. According to Figure 5(b), using Tauc’s method can estimate the bandgap energy according to Equation (6):

**Figure 2.** XRD pattern of γ-Al2O3 NPs.

**Figure 3.** FESEM image of γ-Al2O3 NPs.

**Figure 4.** Size distribution histogram of γ-Al2O3 NPs.

**Figure 5.** (a). UV-Vis absorption spectra of γ-Al2O3 NPs. (b) Optical energy bandgap of γ-Al2O3 NPs.
where $h$, $\nu$, $E_g$ and $\alpha$ are the Planck’s constant, the frequency, the energy of bandgap, and the absorbance. Furthermore, $n$ is related to electronic transmissions of direct allowed ($n = \frac{1}{2}$), indirect allowed ($n = 2$), direct forbidden transmission ($n = \frac{3}{2}$), and indirect forbidden transmission ($n = 3$) \cite{6}.

### 3.2. Optical properties of γ-Al₂O₃ NPs

#### 3.2.1. NLO properties of γ-Al₂O₃ NPs

The NLO properties of the γ-Al₂O₃ NPs suspended in Dimethylformamide (DMF) are calculated with the Z-scan technique. In this research, $\alpha$ is obtained by optical limiting method about 1.002 cm$^{-1}$ and $L_{\text{eff}}$ is calculated about 0.95 mm. The NLR index, $n_2$, of the γ-Al₂O₃ NPs aqueous solution was determined about $3.67 \times 10^{-8}$ (cm$^2$/W) from the closed-aperture-normalized transmittances presented in Figure 6 and utilizing Equations 3 and 4.

The valley-to-peak configuration refers to the positive sign of NLR index $n_2$ and the self-focusing effects \cite{20} under continuous wave (CW) excitation. This origin of nonlinearity response is photothermal and points toward the potential optical limiting application of these NPs in the CW regime \cite{18}.

The Z-scan measurements of open aperture were performed to obtain the NLA coefficients. The normalized transmission curves as a function of the focal distance for open-aperture setup are shown in Figure 7 for γ-Al₂O₃ NPs.

The typical peak symmetric around the focal point showed by open-aperture Z-scan profile reveals that the NLA coefficient has a positive sign, which indicates the existence of nonlinear SA. In SA, the absorption in the sample decreases with an increase in the intensity \cite{30}. The values of the NLA coefficient, $\beta$, are calculated from the open-aperture normalized transmittances given in Figure 7 and using Equation 5 that are calculated about $6.6 \times 10^{-4}$ cm/W. The results demonstrate that γ-Al₂O₃ NPs have excellent third-order NLO response (Table 1).

#### 3.2.2. FTIR analysis of γ-Al₂O₃ NPs

FTIR measurement of γ-Al₂O₃ NPs was carried out in the absorbance mode to find out the optical and dielectric properties. The structures of the obtained γ-Al₂O₃ powders were further investigated by the infrared absorption spectra recorded in the range of 400–4000 cm$^{-1}$ and illustrated in Figure 8.

There are two broad peaks at 3450 and 1630 cm$^{-1}$ and are corresponded to the O-H stretching and...
bending modes of water molecules absorbed by the sample from the environment because of the presence of the KBr pellet [15,41,42]. The peaks below 1000 cm\(^{-1}\) can be attributed to metal oxide arising from interatomic vibrations [43]. There are some characteristic peaks of the amorphous or nanocrystalline nature of alumina powder in 400–1500 cm\(^{-1}\) (finger-print region) [44,45]. The peaks were observed around 1174, 657, and 609 cm\(^{-1}\) related to stretching and bending vibration of Al–OH or Al–O bonds [6,46]. Furthermore, the intense and weak peaks around 470 and 445 cm\(^{-1}\) correspond to AlO\(_6\) and AlO\(_4\) bonds, respectively [41,47].

3.2.2.1. The calculated optical constants. The real and imaginary components of the complex refractive index of γ-Al\(_2\)O\(_3\) NPs are depicted in Figure 9. The value of the static refractive index (n\(_0\)) is calculated by n\(_0\) = \(\sqrt{\varepsilon_\infty} = |(\varepsilon_1 + i\varepsilon_2)_{\infty}|\) [48]. The value of n\(_0\) is found to be 4.089 and it was in correspondence with the other reports [49,50]. The high-frequency dielectric constant |ε\(_\infty\) = (ε\(_1\) + iε\(_2\))\(_\infty\)| is 16.52.

Both parts of the dielectric function (ε\(_1\), ε\(_2\)) of γ-Al\(_2\)O\(_3\) samples are presented in Figure 10. As can be seen, both these parameters varied in the range of 430–490 cm\(^{-1}\), and its trend was plaque in the beyond ranges.

Concerning the IR spectrum, the optical interactions with the lattice could be described by the optical phonon modes. These interactions can be attributed to transverse and longitudinal optical phonon modes (TO and LO). The intersecting points in the n and k graphs can be related to the LO and TO optical modes emerging at 486 and 467 cm\(^{-1}\), respectively (Table 2) [38].

The lower wavenumber (TO) and the higher wavenumber (LO) are shown in Figures 9 and 10.

![Figure 9](image9.png)

**Figure 9.** The refractive index (solid line) and extinction coefficient (dash line) for γ-Al2O3 NPs.

![Figure 10](image10.png)

**Figure 10.** The real (solid line) and imaginary (dash line) parts of the dielectric function for γ-Al2O3 NPs.

**Table 2.** Longitudinal and transverse optical phonon modes (LO and TO) of γ-Al\(_2\)O\(_3\) NPs.

| Optical phonons          | Wavenumber (cm\(^{-1}\)) |
|--------------------------|--------------------------|
| Transvers optical phonon (TO) | 467                      |
| Longitudinal optical phonon (LO) | 486                      |

Furthermore, when the sign of the real dielectric function changes from positive to negative, intermediate point of these two domains is TO frequency mode. In a similar situation, the intermediate point of the negative to positive data of real dielectric function is LO frequency mode [51]. In some literature, the TO and LO modes are the maximum position of ε\(_1\) and Im\((-1/\varepsilon)\) plots, respectively [39]. The LO and TO modes are confirmed by two methods since the LO mode can be observed in Figures 10 and 11. Our results are in good agreement with other researches [49,50].

![Figure 11](image11.png)

**Figure 11.** The Im\((-1/\varepsilon)\)of γ-Al2O3 NPs.
4. Conclusions

Analysis of structural and optical properties of the synthesized γ-Al2O3 NPs by sol–gel technique shows that

(1) The XRD pattern confirms the formation of the cubic structure of γ-Al2O3 NPs. The average crystallite size is obtained at about 8 nm.
(2) The average particles size is obtained at about 60 nm by the FESEM image.
(3) The NLR index is in order of 10⁻⁸ (cm²/W) with a positive sign and a self-focusing effect.
(4) The β is in order of 10⁻⁴ (cm/W) with a positive sign and SA phenomena.
(5) The third-order nonlinearity of γ-Al2O3 NPs measured from Z-scan studies conducted in CW excitation regime confirms its substantial optical nonlinearity and potential optical limiting applications.
(6) The LO and TO modes are the two intersection points in the n and k graphs.

Disclosure statement

No potential conflict of interest was reported by the authors.

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