Synthesis and investigation of dielectric ceramic nanoparticles for microstrip patch antenna applications

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Zinc aluminate (ZnAl2O4) is a well-recognized ceramic demanded in several microwave applications. Further, the addition of dielectric materials in ZnAl2O4 improved its dielectric properties, which is promising for the realization of a microstrip patch antenna. This article reports the investigation of ZnAl2O4TiO2 (ZAT) dielectric ceramic nanoparticles synthesized by the sol–gel process. The X-ray diffraction analysis revealed the crystalline nature of the prepared nanoparticles, with a tetragonal structure of anatase-, and rutile-TiO2 phases coexisting with the cubic phase of ZnAl2O4. The estimated crystallite size of the dielectric ceramic is 13.3 nm. Transmission electron microscopy (TEM) micrographs demonstrated the spherical grains with their mean diameter of 14.75 nm, whereas the selected-area electron diffraction (SAED) pattern endorsed the crystallinity of the sample. Raman measurement revealed the vibrational modes in accordance with the TiO2 and ZnAl2O4 compounds. The dielectric properties of the ZAT sample showed the dielectric permittivity in the range of 22.12–21.63, with its minimum loss from 0.056 to 0.041. Finally, a prototype microstrip antenna was fabricated using the prepared nanoparticles, which demonstrated a return loss of −30.72 dB at the resonant frequency of 4.85 GHz with its bandwidth of 830 MHz.

Dielectric ceramic microwave (MDC) is extensively used in ultra-fast wireless networks and smart transmission systems within the millimeter wave range. The dielectric/ceramic used in millimeter wave technology must have an excellent quality factor, suitable dielectric constant, and a small resonant frequency at near-zero temperature coefficient1,2. In the past several decades, the microwave-based wireless communication sectors have been revolutionized by employing dielectric/ceramic materials in miniaturizing antennas with low-cost fabrication. The tailoring of the dielectric permittivity of the materials yielded their unique electrical characteristics, and therefore, found promising in miniaturizing the antenna.

Due to recent advancements in wireless communication, simple, durable, cost-effective, lightweight, low-profile patch antennas have been demanded3. However, due to the relatively low impedance of microstrip patch antennas, their usage in electronic equipment is limited. Numerous techniques are claimed to increase the bandwidth of microstrip patch antennas, including thick substrates, parasitic patches, and so on4,5. Microwave-controlled devices such as microstrip patch antennas are critical for automated high-speed cars. In recent years, considerable interest has been noticed in a novel type of microstrip patch antennas6. These low profile antennas are inexpensive, lightweight, and simple in processing, and they are extensively demanded in various applications, including defence and consumer products7. Additionally, these can operate in various wireless communication bands, including wireless local area networks, wireless fidelity, and worldwide interoperability for microwave access8. Further, extensive research has been focused on designing the various shapes of the microstrip patch antenna and their impact on the antenna dimension, materials used in antenna fabrication, and their corresponding performance9. A portable designed device based on 22 array of cylindrical dielectric antennas demonstrated the increased gain and lower-polarization. The integrated differential feeding array components were responsible for the reduced cross-polarization characteristic and enhanced antenna gain. The designed antenna exhibited −10 dB impedance bandwidth in the frequency range from 5.78 to 5.9 GHz. The obtained antenna

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Because of its unique properties, zinc aluminate (ZnAl₂O₄) spinel-type ceramic has become increasingly important in modern technology. This material is beneficial in various applications due to its synergetic properties, including excellent durability, moderate heat treatment, good thermal resistance, better mechanical robustness, and broad bandgap (3.8 eV)¹¹–¹³. Additionally, this has been employed as a transparent conductor for ultraviolet radiation, detector, dielectric, and optical substances¹⁴–¹⁷. ZnAl₂O₄ has also been demanded in the telecommunications industry for resonating, filtering, and oscillating wireless fax, mobile phones, GPS, military radar systems, smart transmission systems, and satellites. The dielectric properties of ZnAl₂O₄ as microwave dielectric ceramic by adding TiO₂ are studied¹⁸–²¹. They reported the potential applications of ZnAl₂O₄:TiO₂ in future microwave substrates and antennas. The addition of a small amount of TiO₂ to the ZnAl₂O₄ resulted in an increased dielectric permittivity. Recently, the properties of ZnAl₂O₄ ceramic based on either composite or doped with TiO₂, Mg, TiO₂ – xSrTiO₃, Co, TiO₂, Mg, TiO₂ etc. are being investigated. The ZnAl₂O₄ based ceramic is a well-known patch material for GPS or microwave substrates. Abdullah et al. fabricated and studied the performance of the patch antenna using the nanoparticles of (1 – x)ZnAl₂O₄ – xSiO₂. The crystallite size of this compound was estimated to be in the range of 39.79 to 44.34 nm, along with the dielectric permittivity of 8.57. This patch antenna showed its return loss of ~ 14.25 dB at the resonant frequency of 3.46 GHz with its bandwidth of 60 MHz. Kim et al. studied the various dielectric ceramics having a large dielectric permittivity (εr) and positive temperature coefficient of resonant frequency (τf) values². It implies that low-dielectric ceramics (< 20) are critical for frequency stability across a range of temperatures. According to Narang and Shalini et al., it is possible to improve the properties of dielectric ceramics by incorporating an appropriate material and modifying the synthesis approach²². A study conducted by Kumar et al. revealed that ceramic materials containing metal (conductive) oxides are important in enhancing the structural properties of ZnAl₂O₄. Likewise, various researchers reported the use of doped ZnAl₂O₄ in a variety of optical and catalytic applications, including Zn(1 – x)MnₓAl₂O₄, Sr(II):ZnAl₂O₄, ZnAl₂O₄:Eu²⁺, and ZnAl₂O₄:TR (TR = Eu²⁺, Tb³⁺)²³–²⁶. Thirumana-athan et al. reported the study of the formation of bismuth titanate nanoparticles by the combustion process²⁷. They performed the dielectric properties of prepared nanoparticles and fabricated the patch antenna. They observed the dielectric permittivity value of 450, the dielectric loss of 0.98, and antenna’s return loss of ~ 4.95 dB at the resonant frequency of 2.45 GHz. In another work, Rahman et al. studied the properties of patch antenna using the sol–gel derived garnate (ZnAl₂O₄) nanoparticles²⁸. The obtained dielectric constant, optical bandgap, and quality factor were 8.7, 4.08 eV, and 4592, respectively. The prepared microstrip antenna demonstrated the antenna’s return loss of ~ 25.4 dB at the resonant frequency of 12.78 GHz with its bandwidth of 760 MHz and 8.1 GHz in the low- and high-frequency bands, respectively. Abdullah et al. examined the performance of a patch antenna based on magnesium doped ZnAl₂O₄ ceramic nanoparticles. By varying the concentration of magnesium doping, they reported the crystallite size, lattice parameter, dielectric permittivity, return loss, and bandwidth in their range from 19.2 to 12.9 nm, 8.082 to 6.048, –16.34 to –21.38 dB, and 90 to 225 MHz, respectively. They fabricated the GPS patch antenna and noticed its resonant frequency at 1.570 GHz²⁹. This paper reports the synthesis and investigation of ZnAl₂O₄:TiO₂ dielectric ceramic nanoparticles prepared by an inexpensive and straightforward sol–gel route. Further, we employed these nanoparticles to fabricate a prototype microstrip patch antenna, which demonstrated its return loss of ~ 30.72 dB at a resonant frequency of 4.85 GHz. To the best of our knowledge, no similar work of fabricating a microstrip patch antenna using ZnAl₂O₄:TiO₂ has been reported for the C-band applications. “Materials and methods” section presents the materials and methods of synthesizing ZnAl₂O₄:TiO₂ composite nanoparticles. The characteristics of ZnAl₂O₄:TiO₂ nanoparticles, dielectric properties, and microstrip patch antenna performance are discussed in “Results and discussion” section. Lastly, “Conclusions” section summarizes the paper.

**Materials and methods**

For the sol–gel synthesis of ZnAl₂O₄:TiO₂ (ZAT) dielectric ceramic nanoparticles, titanium tetra isopropoxide (TTIP, Sigma Aldrich), zinc acetate (CH₃COO)₂Zn₂H₂O (Lobychem), aluminum nitrate nonahydrate (Al₃(NO₃)₂·9H₂O, Sigma Aldrich), ethanol (C₂H₅OH, Sigma Aldrich), ethylene glycol (EG, SdFine) and nitric acid (HNO₃, Lobychem) were procured and used without any further purification.

Figure 1 illustrates the step-by-step process followed in synthesizing the composite ZnAl₂O₄:TiO₂ nanoparticles and the pellet preparation for the measurement of dielectric properties. Initially, 5 ml distilled water was added to 75 ml ethanol and stirred. Then 4.5 ml TTIP solution was added and stirred for about 4 h at approximately 85 °C. Followed by this, a white powder was obtained, calcined at 700 °C and ground. The prepared nanoparticles were employed to prepare a pellet using the silver paste by doctor blade process.

During this process, 0.24 ml nitric acid (HNO₃) was dropped into the solution for preparing the homogeneous solution and stirred at temperature 75 °C for another 1 h till the formation of a clear solution. Doing so, (1 – x)ZnAl₂O₄ – xTiO₂ powder was obtained while x is the concentration of the TiO₂ (i.e., x = 0.1). The sample was dried in an oven for 30 min at a temperature of 180 °C. Lastly, the sample was calcined at a temperature of 700 °C for 1 h and then ground. The prepared nanoparticles were employed to prepare a pellet using the pellet press machine. The prepared pellet was 1.12 mm thick and 10 mm in diameter. Prior to dielectric measurement, the as-prepared pellet was thermally treated at temperature 700 °C for 1 h and then it’s both sides were coated using the silver paste by doctor blade process.
The prepared sample named as ZA was characterized by using an X-ray diffractometer (XRD, X-Pert Pro, UK), Raman spectroscopy (BWTEK, Japan), Transmission electron microscopy (TEM, TALOS F200S G2, USA), energy dispersive X-ray spectroscopy (EDS), and LCR meter (PSM1735 N4L, Newtons4th Ltd, UK). The fabricated prototype microstrip patch antenna based on composite nanoparticles was tested using the Vector Network Analyzer (VNA, R&S®ZVL, Germany).

Results and discussion
The X-ray diffraction (XRD) investigation is advantageous for determining the crystalline phases of the nanomaterials. Figure 2 depicts the XRD pattern of ZAT nanocomposite dielectric ceramic material. It represents the various peaks of ZnAl2O4 crystal structure corresponding to the typical face-centered cubic morphology and was found consistent with the reported literature. One can also notice the formation of the crystalline structure of titanium dioxide (TiO2) with its anatase and rutile phases and the wurtzite structure of ZnO. Compared to the pristine ZnAl2O4 sample, which showed the various peaks of ZnAl2O4, some peaks of ZnO can also be noticed. Further, in the composite sample of ZnAl2O4TiO2 the additional peaks of TiO2 were noticed. In addition, the peak locations were found slightly shifted with the increased value of TiO2 concentration. In other words, the unit cell dimension was noticed to be decreased with the enhanced crystallinity. In our case, the crystallite size was estimated to be 13.3 nm using the Scherrer formula (d = 0.94λ/(βcosθ), where β is the X-ray wavelength, and β is the full-width at half-maximum intensity of the diffraction line).

Figure 3 shows the Raman spectra of ZnAl2O4TiO2 nanoparticles sintered at a temperature of 700 °C. We noticed various peaks from the prepared sample that originated due to ZnAl2O4, ZnO, and TiO2 contents. We can observe two Raman peaks originated at 395 cm⁻¹ and 519 cm⁻¹ assigned to B1g and A1g/B1g modes, respectively, showing the impression of anatase-TiO2. Further, a peak 439 cm⁻¹ known as E₂ high vibration mode was associated with oxygen atoms and assigned to ZnTiO3 nanocrystals. A broad peak at 618 cm⁻¹ represents the thermodynamically stable rutile-TiO2 relates the space group D₄h assuming the site symmetries for the Ti and O atoms within the unit cell. In literature, this peak is attributed to the Raman-active “lattice vibration” designated as A₁g.

TEM measurement was carried out to know the morphology of ZAT dielectric ceramic nanoparticles. Figure 4a depicts the good yield of ZnAl2O4TiO2 nanoparticles as recorded at the scale of 200 nm. We can observe almost well-dispersed nanoparticles with very low agglomeration in the TEM micrograph as shown in Fig. 4b, which was recorded at the scale of 50 nm. Figure 4c depicts the high-resolution TEM micrograph of ZAT nanoparticles. The calculated inter-spacing (d) values were 0.130, 0.145, and 0.167 nm, corresponding to the reflection...
of the planes of (201), (103), and (110). The selected-area electron diffraction (SAED) pattern depicted in Fig. 4d
depicts the appearance of solid rings representing the polycrystalline nature of the ZAT nanoparticles. The result
coincides with the XRD pattern as discussed in Fig. 2.

Figure 5a illustrates the histogram of ZnAl$_2$O$_4$TiO$_2$ nanoparticles by measuring the diameters of about 40
grains to estimate the average size. As can be seen, the dielectric ceramic nanoparticles have an average diameter
of 14.75 nm.

The EDS spectrum of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles is shown in Fig. 5b. The EDS spectrum
revealed the elemental peaks of O, Zn, Al, and Ti at energy values 0.52, 1.11, 1.48, and 4.5 keV, respectively.

The dielectric permittivity value represents the material's ability to store electric energy when an electric
field is applied, and it is related to the capacitance associated with the dipole orientation of charge carriers. After
obtaining the parallel capacitance values, we have calculated the dielectric permittivity by using an expression,
$$\varepsilon_r = \frac{C d}{\varepsilon_0 A},$$
where C is the capacitor's capacitance, d is the pellet's thickness, \(\varepsilon_0\) is the permittivity of free space,
and A is the cross-section area of the pellet. The dielectric characteristic of ZnAl$_2$O$_4$TiO$_2$ nanoparticles was
studied using the LCR meter with its frequency range from 100 Hz to 1 MHz at room temperature.

Figure 6 shows the dielectric measurement setup used in this study. The LCR meter is connected to the com-
puter, while the front panel (bottom-left) of the LCR meter is loaded with a pellet under the test. One can also
do the dielectric measurement by varying the temperature through a separate unit, as shown here. However, the
dielectric measurement was carried out at room temperature in this case.

Figure 7 depicts the variation of dielectric permittivity in accordance with the frequency. With the increased
frequency, one can observe the decreased permittivity. The dielectric permittivity was varied from 22.12 to 21.63

Figure 2. XRD pattern of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles prepared by the sol–gel method.

Figure 3. Raman spectra of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles.
Figure 4. TEM micrographs at scale 200 nm (a) at scale 50 nm (b), HRTEM image (c) and SAED pattern (d) of ZnAl₂O₄TiO₂ dielectric ceramic nanoparticles.

Figure 5. Histogram (a) and EDS spectrum (b) of ZnAl₂O₄TiO₂ dielectric ceramic nanoparticles.
with an increased frequency range from 100 kHz to 1 MHz. Roshini et al. also reported the ZnAl$_2$O$_4$TiO$_2$ material with its dielectric constant of 9.6. Similarly, Abdullah et al. studied the dielectric permittivity of ZnAl$_2$O$_4$-SiO$_2$ nanoparticles and reported its value of 8.57. An abrupt decrease in dielectric permittivity in the lower frequency range was noticed, which was found constant in the higher frequency region. This typical characteristic of such materials can be attributed to the reduced polarization.

Dielectric loss (tanδ) is an important parameter representing the energy dissipation, and therefore, it needs to be studied. Dielectric loss is also regarded the microstructure faults, e.g. microstructural defects, porosity, micro-crashes, the spontaneous orientation of crystallite, etc. Figure 8 depicts the dielectric loss of ZnAl$_2$O$_4$TiO$_2$ nanocomposite ceramic sample as a function of frequency. The dielectric loss was noticed to be decreased from 0.056 to 0.041 with an increased frequency range from 100 kHz to 1 MHz. In general, one can observe the reduced dielectric loss with the increased frequency. This nature is because the hopping ions lag behind the applied electric field. At the lower frequency range, one can notice the increased dielectric loss value, which further decreases in the higher frequency region. In the first case, this relates to the high resistivity resulting from the associated effect of grain boundaries.

We have calculated the real ($Z'$) and imaginary ($Z''$) impedance values with respect to the frequency, which are plotted in Figs. 9 and 10, respectively. As shown in Fig. 9, we can notice the decreased real-impedance from 3.28 kΩ to 467 Ω with the rise in frequency. Similarly, Fig. 10 depicts the same trend of increased imaginary impedance values from −59 to −10 kΩ with the increased frequency from 100 kHz to 1 MHz. The $Z'$ value fluctuates with temperature and joins together in the higher frequency region (not shown here). This happens due to the
Figure 8. Dielectric loss of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles.

Figure 9. Impedance variation (real part) of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles.

Figure 10. Impedance variation (imaginary part) of ZnAl$_2$O$_4$TiO$_2$ dielectric ceramic nanoparticles.
Figure 11. Conductivity of ZnAl₂O₄TiO₂ dielectric ceramic nanoparticles.

Figure 12. Digital images of the prototype microstrip patch antenna.

liberation of charge carriers and semiconducting characteristics at high temperatures. However, the Z" value rises with temperature, and the larger value at a higher frequency regime indicates the increase in tangent loss.

Figure 11 illustrates the frequency-dependent ac conductivity at room temperature. The ac conductivity was estimated using the relation $\sigma_{ac} = \omega \varepsilon \varepsilon_0 \tan \delta$, where $\varepsilon_0$ is the free space dielectric permittivity, $\varepsilon$ is dielectric permittivity, $\omega$ is the angular frequency, and $\tan \delta$ is the tangent loss. The investigation of conductivity as a function of frequency relates to the process of charge transport. We can notice the enhancement in conductivity from $2.2 \times 10^{-5}$ to $9.8 \times 10^{-5}$ with the increased frequency. This increasing trend of conductivity in the lower frequency range (not shown here) can be attributed to space charges scattering cations across adjacent sites. The conductivity curve coincides at high-frequency band, representing that the conductivity curves obey Jonscher's power law and therefore exhibit low-frequency dispersion phenomena.

The prepared ceramic dielectric ZnAl₂O₄TiO₂ nanoparticles were employed for preparing a patch antenna. Initially, ZAT paste was prepared, which was cast on the FTO substrate and then it was silver coated on both sides for metal contacts. Finally, the SMA connector was connected to it, and antenna performance was evaluated using a vector network analyzer. Figure 12 depicts the patch antenna's top view which illustrates its dimension and shape. The fabricated prototype microstrip patch antenna has its length and width of 25 mm and 15 mm, respectively, as illustrated in Fig. 12a.

Figure 13 depicts the measured return loss of ZnAl₂O₄TiO₂ microstrip patch antenna measured in the range from 4 to 6 GHz. The fabricated antenna covered the minimum required value of return loss, i.e. ~ 10 dB. The prototype microstrip antenna demonstrated a resonant frequency of 4.85 GHz and the return loss of -30.72 dB with its bandwidth of 830 MHz.

Conclusions

Dielectric ceramic ZnAl₂O₄TiO₂ nanoparticles prepared using the low-cost and easy technique have been studied. The synthesized nanoparticles were crystalline with their crystallite size of 13.3 nm. The Raman study evidenced the corresponding Raman shift of the constituent elements presented in the composite nanoparticles. The morphological investigation of the nanoparticles endorsed the formation of spherical grains with their mean diameter of 14.75 nm. The crystallinity of the prepared sample studied by the SAED pattern was consistent with the XRD result. The LCR meter measurement showed the decreased dielectric permittivity and loss as a function of
applied frequency. The performance of the prototype microstrip patch antenna based on the dielectric ceramic nanoparticles was also studied. The microstrip patch antenna exhibited its return loss of −30.72 dB at the resonant frequency of 4.85 GHz with its bandwidth of 830 MHz. In summary, a microstrip patch antenna demonstrated its resonant frequency in the C-band range, which lies from 4 to 8 GHz, and therefore, this material would be suitable for satellite communications, weather radar systems, terrestrial microwave links, and 802.11 versions of Wifi devices.

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Competing interests
The authors declare no competing interests.

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