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Exploration of Analyte Electrolyticity Using Multi-SRR-Hexagonal DNG Metamaterials and ZnO Thin Films

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ABSTRACT

Advanced engineered metamaterials (MTMs) significantly contribute to modern technological advancements, particularly through hybridization with semiconductor materials like zinc oxide (ZnO), which enhance sensor sensitivity and performance. This study aims to investigate the optical properties of hybrid MTMs and develop a novel sensor medium capable of detecting early electrolytic behaviors of analytes. Utilizing the finite-difference time-domain (FDTD) method, the sensor was designed, characterized, and integrated, featuring a hexagonal multi-cell split ring resonator (SRR) structure coated with a 200-nm ZnO thin film. The geometry of the SRR MTM was optimized using a modified Nicolson-Ross-Weir electromagnetic field function method. Results demonstrate that the MTM exhibits double-negative optical characteristics with a performance index reaching 10^2. Moreover, the sensor presents dual-band resonance frequencies for reflection and transmission attributed to the combination of the multi-SRR hexagonal design and ZnO coating, with an absorption peak at 8.71 GHz. Testing the sensor in varying electrolytic conditions, such as seawater, revealed a measurable reduction in resonance depth and increased sensitivity, characterized by a frequency shift of 5.25 MHz per 0.7 S/m increment in electrical conductivity. These findings highlight the MTM sensor's potential as an effective tool for enhancing spectrum readout accuracy and sensitivity in analyte detection applications.

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1. INTRODUCTION

In the contemporary digital era, researchers are increasingly expected to contribute significantly to the development and innovation of sensor technologies utilizing advanced renewable materials with superior performance characteristics. Sensor technology, crucial in various industrial sectors, has undergone notable advancements driven by the integration of renewable materials, particularly metamaterials (MTMs). Over the past decade, both academia and industry have shown growing interest in MTM sensor technologies, focusing primarily on enhancing sensor efficiency and effectiveness through the application of renewable methodologies and advanced material combinations [1-3].

Metamaterials are artificially engineered materials designed to exhibit unique electromagnetic properties, such as negative permittivity and permeability, which do not naturally occur in conventional materials. Although theoretical proposals of MTMs date back approximately 25 years, practical exploration and widespread experimentation have significantly intensified over the past decade [4]. MTMs have been successfully developed for diverse applications, including telecommunications systems like compact asymmetric square-shaped designs for 5G technology [5], military radar employing triangular leaf-shaped structures operating at 10 THz [6], medical sensors with rod-shaped configurations detecting blood sugar at frequencies around 1.99 GHz [7], and chemical sensors using double G-shaped designs at frequencies ranging between 8–12 GHz [8]. However, despite these notable advances, pure MTMs have yet to fully meet the stringent quality standards required by modern technological applications. This limitation underscores the necessity for comprehensive research aimed at optimizing MTMs into high-performance hybrid materials.

Recently, hybrid MTMs have emerged as promising solutions by combining various compound constituents to achieve sophisticated, renewable, and highly responsive materials [9, 10]. Specifically, the integration of nanoscale semiconductor materials like zinc oxide (ZnO) into MTM structures has attracted substantial attention due to their advantageous characteristics. ZnO demonstrates excellent potential for hybrid MTMs, particularly for sensing applications [11], owing to its chemical stability [12], thermal robustness [13], photocatalytic [14], and electrical conductivity properties [15]. Furthermore, the nanoscale properties of ZnO can be significantly enhanced through doping, thereby improving electron mobility [16], photocatalytic efficiency [17], catalytic stability and performance [18], and reducing charge carrier recombination [19].

Moreover, modern manufacturing technologies related to transmission lines and power cables provide new pathways for integrating MTM sensors into advanced infrastructure systems, emphasizing the potential of these hybrid structures in contemporary and future technological scenarios. Hence, this study aims to design and develop composite sensor materials by combining ZnO thin films with advanced MTMs. The resulting hybrid materials are anticipated to serve as highly sensitive semiconductive sensors, aligning with sustainability objectives and offering improved responsiveness essential for next-generation technological applications.

2. MTM-ZnO CONFIGURATION

The MTM is designed as a hexagonal pattern The structure consists of a 3×3 arrangement of SRR combined with a thin film of ZnO, as depicted in Figure 1.

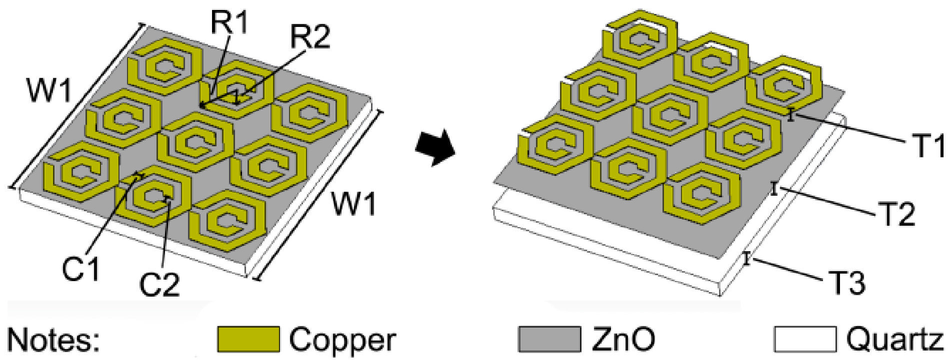

Figure 1. An integrated MTM with multi-SRR-hexagonal and ZnO thin film design and structure

The MTM structure is composed of two resonator rings constructed of pure copper (ε_r = 1), positioned on the upper surface of a quartz dielectric substrate (ε_r = 3.8). Concurrently, there exists a slender layer of ZnO positioned between the SRR layer and the substrate. The dimensions of the multi-SRR-hexagonal MTM structure are determined by the minimum wavelength of the operating frequency, which ranges from 0.009 – 9 GHz. The multi-SRR-hexagonal and ZnO thin film exhibit distinct differences in terms of their ring radii, widths, and thicknesses, as illustrated in Table 1.

Table 1. MTM geometry combined by multi-SRR-hexagonal and ZnO thin film

Parameter	Value (mm)	Parameter	Value (mm)
C1	0.61	T1	0.50
C2	0.43	T2	0.0002
R1	2.70	T3	1.00
R2	1.70	W1	16.28

The properties of MTMs are found by analysing spectral parameter data collected from simulations of electromagnetic wave propagation on samples of MTMs. The permittivity, permeability, and refractive index properties of the MTM are determined by processing the reflection (S11) and transmission (S12) spectrum data using the modified Nicolson-Ross-Weir equation as follows [20]:

$$\varepsilon_r = \frac{2}{jk_0 t_m} \times \frac{1-(S12+S11)}{1+(S12+S11)} \tag{1}$$

$$\mu_r = \frac{2}{jk_0 t_m} \times \frac{1-(S12-S11)}{1+(S12-S11)} \tag{2}$$

$$n = \sqrt{\varepsilon_r \mu_r} \tag{3}$$

where, k_0 is wavenumber, t_m is propagation length, ε_r is relative permittivity, μ_r is relative permeability, and n is refractive index.

3. DOUBLE-NEGATIVE INDEXED

The optical and electromagnetic characteristics of the double-negative (DNG) multi-SRR-hexagonal metamaterial (MTM) are significantly influenced by the application of a ZnO thin film layer. Quantitative analysis reveals an enhancement in the material’s relative permittivity, with a resonance depth reaching -198.93 at a frequency of 6.27 GHz (Figure 2a). This indicates a highly responsive interaction with electric fields, which is crucial for electromagnetic wave manipulation. Materials with elevated permittivity show stronger coupling with incident electric fields, improving energy localization and sensor sensitivity.

In terms of magnetic response, the hybrid MTM structure also demonstrates a significant improvement in relative permeability, with a resonance dip of -74.58 observed at 4.82 GHz (Figure 2b). This value reflects the MTM's strong interaction with magnetic field components of the incident wave, further supporting its classification as a double-negative metamaterial.

Additionally, the refractive index, which is derived from both relative permittivity and permeability, was found to be significantly negative. The hybrid MTM structure achieved a negative refractive index value of -7.62 at 1.91 GHz (Figure 2c). This high-magnitude negative index is crucial for applications such as superlensing and cloaking, where backward wave propagation and energy redirection are required.

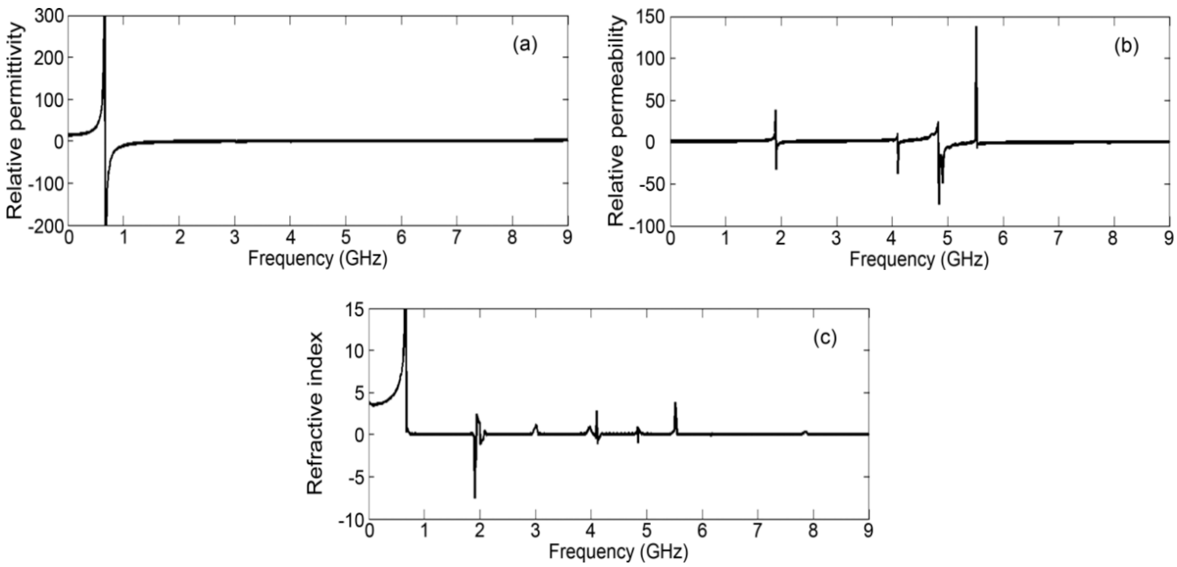

Figure 2. Integrated MTM resonant optical features: (a) permittivity, (b) permeability and (c) refractive index

The integration of ZnO thin films contributes not only to electrical and optical enhancements but also improves the magnetic resonance behavior of the MTM. These synergistic effects lead to increased performance in electromagnetic wave sensing, signal modulation, and field localization.

Table 2. Negative resonance in high DNG integrated MTMs

Properties	Frequency (GHz)	Value
Relative permittivity	6.27	-198.93
Relative permeability	4.82	-74.58
Refractive index	1.91	-7.62

Furthermore, Table 2 summarizes the peak resonance values of the permittivity, permeability, and refractive index across the investigated frequency spectrum, reinforcing the hybrid MTM's capability for high-sensitivity detection. The performance improvements demonstrate the suitability of ZnO-MTM composites for next-generation sensor technologies, particularly in applications requiring precise dielectric and magnetic field interactions.

4. SENSOR DESIGN

The sensor was designed by combining a multi-SRR-hexagonal MTM and a thin layer of ZnO. This was achieved by incorporating a circuit channel and incorporating various material components, as depicted in Figure 3.

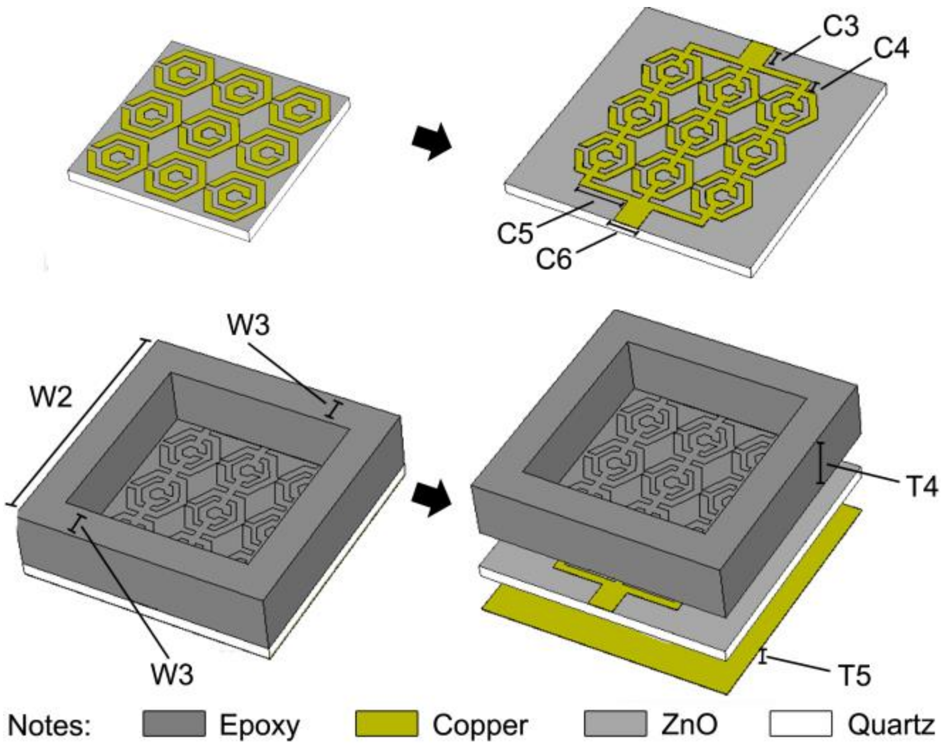

Figure 3. Integrating multi-SRR-hexagonal and ZnO thin film MTMs for analyte electrolyticity detection

Table 3. Complete MTM sensor structure parameters

Parameter	Value (mm)	Parameter	Value (mm)
C3	1.25	T4	3.87
C4	1.04	T5	0.50
C5	4.59	W2	20.0
C6	2.00	W3	1.86

The selection of circuit configuration and power supply interface is determined by the goal of optimising sensor performance to achieve maximum absorption of power transfer. The epoxy sample container, with a volume of 1 × 10^3 mm^3, has the capacity to hold just 1 ml of analyte sample. The sample should be created concentrically to the multi-SRR-hexagonal surface and covered with a thin film of ZnO. Furthermore, supplementary grounding elements are affixed to the rear side of the substrate. Table 3 provides specific information regarding the dimensions of the sensor structure.

5. DETECTION PERFORMANCE

Figure 4 presents the reflection, transmission, and absorption spectra of the ZnO-integrated MTM sensor in a sampleless measurement configuration. In the reflection spectrum, two prominent resonance dips are observed. The first occurs within 3.85 – 4.65 GHz, with a minimum reflected power of -30 dB at 4.22 GHz, indicating efficient energy coupling at this frequency (Figure 4a). A secondary reflection resonance appears at 8.15 GHz, where the reflection magnitude is reduced by 10 dB, demonstrating increased absorption or transmission at higher frequencies.

The transmission spectrum illustrates the energy passing through the sensor, with notable resonance dips at 2.25 GHz (-5.18 dB) and 6.36 GHz (-5.56 dB), indicating partially attenuated signal passage (Figure 4b). Anomalies at 8 – 9 GHz suggest structural dispersion effects in the MTM geometry.

The absorption spectrum (Figure 4c) complements the reflection and transmission findings. A strong absorption peak, reaching approximately 50% of incident energy, is observed at 8.71 GHz. This corresponds to reduced reflectivity and transmission, confirming that the sensor structure achieves optimal energy capture at this frequency.

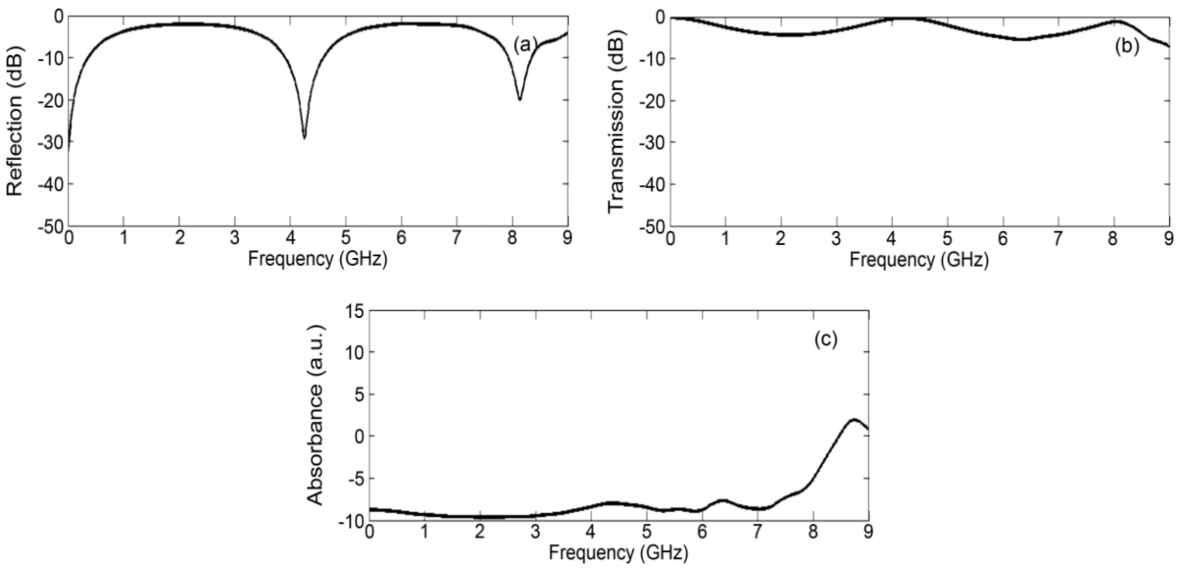

Figure 4. Performance of MTM sensor without sample: (a) reflectance, (b) transmission, and (c) absorbance

In Figure 5, sample-dependent spectral responses are analyzed for four materials: oil (0 S/m), distilled water (5.55 × 10^{-6} S/m), tap water (1.59 S/m), and seawater (3.53 S/m). These differences in electrical conductivity generate distinct shifts in resonance frequency. Notably, seawater and tap water—though similar in dielectric properties—exhibit slightly different resonance peaks in the 5.54 – 6.28 GHz range, reflecting variations in their electrolytic content.

The shift in resonance frequency mimics the behavior of an LC circuit, where changes in material properties affect system inductance and capacitance [23]. This principle explains why resonance shifts correlate with sample conductivity. The observed differentiation, particularly between tap water and seawater, highlights the sensor's sensitivity and resolution.

Figure 6 presents a detailed quantitative analysis of the relationship between resonance depth and electrical conductivity in seawater samples. Electrical conductivity values ranging from 3.53 to 7.03 S/m were tested, and a clear inverse correlation was observed as conductivity increases, the resonance depth consistently decreases. Specifically, the reflected resonance intensity dropped from -24.2 dB to -21.4 dB across the conductivity range, indicating greater power absorption by the sensor structure due to stronger induced electric fields in more conductive media [24].

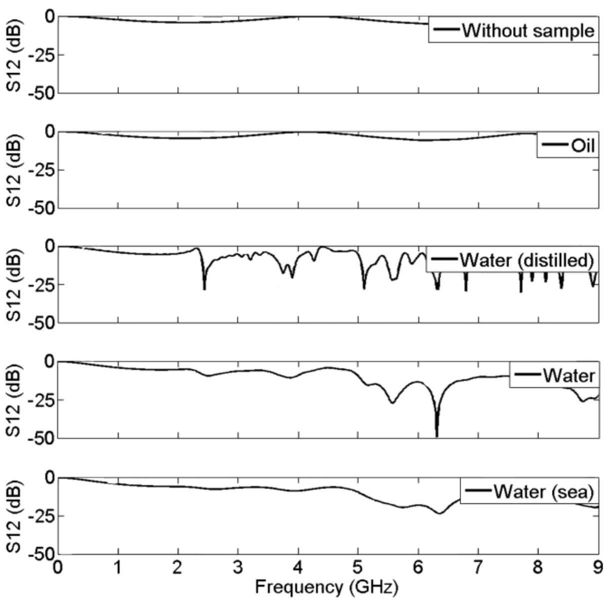

Figure 5. Transmission spectrum changes for some sample experiments

Additionally, a narrowing of the frequency separation between dual resonance peaks was recorded with increasing conductivity, suggesting increased field confinement and interaction efficiency at higher electrolyte levels. The sensor's sensitivity was quantified as 0.0525 GHz (or 52.5 MHz) per 0.7 S/m increase in conductivity. This corresponds to a resonance shift of 5.25 MHz for every 0.7 S/m increment.

This performance is notably improved compared to the findings of Saktioto et al. (2024) [9], where a square-shaped MTM SRR sensor achieved a shift of 3.13 MHz per 0.7 S/m. The current hexagonal MTM SRR integrated with ZnO demonstrates a 67.7% increase in sensitivity, equating to an enhanced detection capability of approximately 7.5 MHz per S/m.

These results validate the effectiveness of the ZnO-MTM hybrid configuration in enhancing sensing performance and demonstrate its strong potential for advanced dielectric and chemical sensing applications. In particular, the sensor exhibits high-resolution liquid discrimination based on electrical conductivity. Furthermore, these findings confirm that the integrated ZnO-MTM sensor can reliably distinguish between various liquids by tracking their conductivity characteristics. The device consistently shows a resonance frequency shift of approximately 5.25 MHz per 0.7 S/m change in conductivity, supporting its promising application in both chemical and biological sensing environments.

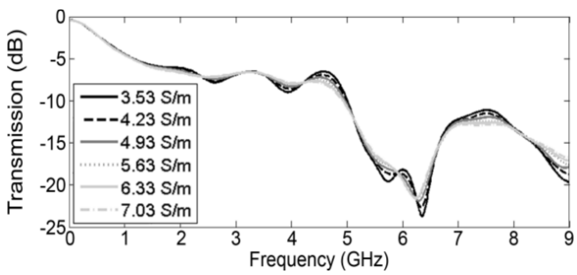

Figure 6. Electrolyticity or electrical conductivity determines seawater sample measurement outcomes

6. CONCLUSION

This study presents the successful design and simulation of a hybrid metamaterial (MTM) sensor integrating a multi-SRR-hexagonal structure with a ZnO thin film, resulting in enhanced double-negative electromagnetic properties with high permittivity (-198.93 at 6.27 GHz), permeability (-74.58 at 4.82 GHz), and a negative refractive index (-7.62 at 1.91 GHz). The sensor demonstrated strong dual-band resonance characteristics (3.85 – 4.65 GHz and 7.94 – 8.37 GHz) and a peak absorption of 50% around 8.71 GHz, confirming its efficiency in energy capture. Experimental validation showed that the sensor could detect conductivity-induced spectral shifts across a range of test liquids, achieving a consistent resonance shift of 5.25 MHz per 0.7 S/m change in conductivity—an improvement of 67.7% over previous designs. The ability to discriminate between similar liquids like tap water and seawater in the 6.21 – 6.35 GHz range further highlights its precision and resolution. These findings establish the ZnO-MTM sensor as a promising solution for high-sensitivity chemical, biological, and environmental sensing applications, offering a significant contribution to the advancement of hybrid metamaterial sensor technologies.

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