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# Extreme DNG metamaterial integrated by multi-SRR-square and ZnO thin film for early detection of analyte electrolyticity

***Abstract**. Advanced engineering in metamaterials can realize the potential for better sensor performance in combination with zinc oxide (ZnO) semiconductor materials. This research aims to investigate the optical properties and develop a sensor media invention based on hybrid metamaterials. The finite-difference time-domain method was carried out in this research for the design, characterization, and integration of a metamaterial sensor by a multi-cell split ring resonator (SRR) with a square pattern and a thin film of ZnO (200 nm). Geometry characterization of the SRR metamaterial was carried out using a modified Nicolson-Ross-Weir electromagnetic field function approach. The application of this type of sensor is used to detect early electrolyticity of analytes resulting from transmission spectra based on the electrical conductivity of several samples. Analysis of metamaterial characteristics identified double-negative (DNG) optical properties that increased drastically to the extreme point on the scale of $10^4$. The performance of the metamaterial sensor integrated by multi-SRR-square and ZnO thin film provides reflection and transmission resonance frequencies in a triple bandwidth. In addition, 73% of the absorption spectrum is at a frequency of ~8 GHz. Seawater electrolyticity sensor testing provides better spectrum readings with a reduction in resonance depth and frequency shift sensitivity of 3.13 MHz for every increase in electrical conductivity of 0.07 S/m.*

***Streszczenie.** Zaawansowana inżynieria metamateriałów może wykorzystać potencjał lepszej wydajności czujnika w połączeniu z materiałami półprzewodnikowymi z tlenku cynku (ZnO). Celem tych badań jest zbadanie właściwości optycznych i opracowanie wynalazku nośnika czujnikowego opartego na metamateriałach hybrydowych. W tych badaniach przeprowadzono metodę różnic skończonych w dziedzinie czasu w celu zaprojektowania, scharakteryzowania i integracji czujnika metamateriału za pomocą wielokomórkowego rezonatora z dzielonym pierścieniem (SRR) o wzorze kwadratowym i cienkiej warstwy ZnO (200 nm). Charakterystykę geometrii metamateriału SRR przeprowadzono przy użyciu zmodyfikowanego podejścia opartego na funkcji pola elektromagnetycznego Nicolsona-Rossa-Weira. Zastosowanie tego typu czujnika służy do wykrywania wczesnej elektrolitowości analitów na podstawie widm transmisyjnych na podstawie przewodności elektrycznej kilku próbek. Analiza właściwości metamateriału pozwoliła zidentyfikować właściwości optyczne podwójnie ujemne (DNG), które drastycznie wzrosły do skrajnego punktu w skali 104. Wydajność czujnika metamateriału zintegrowanego z cienką warstwą multi-SRR i ZnO zapewnia odbicie i częstotliwości rezonansowe transmisji w potrójną szerokość pasma. Ponadto 73% widma absorpcji przypada na częstotliwość ~8 GHz. Testowanie czujnika elektrolitowości wody morskiej zapewnia lepsze odczyty widma przy zmniejszeniu głębokości rezonansu i czułości przesunięcia częstotliwości o 3,13 MHz przy każdym wzroście przewodności elektrycznej o 0,07 S/m. **(Ekstremalny metamateriał DNG zintegrowany z wieloma kwadratami SRR i cienką warstwą ZnO do wczesnego wykrywania elektrolitowości analitu)***

**Keywords:** double-negative, electrolyticity, metamaterial, split ring resonator, ZnO.
**Słowa kluczowe:** podwójnie ujemny, elektrolityczność, metamateriał, rezonator z dzielonym pierścieniem, ZnO.

## Introduction

Industrial developments in the modern digital era now require researchers to play a greater role in developing and creating sensor technology from renewable materials with high-performance quality. The technology currently being developed is a sensor device with quality achievements in the industrial sector [1]. This technology continues to develop due to the contribution made by the nature of renewable materials, namely metamaterials. Metamaterial sensor technology has proven to have attracted interest from researchers and industry in the last ten years with efforts to improve the quality of sensor performance through renewable techniques and materials that are more effective and efficient [2, 3].

Metamaterials are artificial materials with unique characteristics and high sensitivity. Theoretically, metamaterial discoveries existed around 25 years ago and were experimented with in the world in the next 10 years, but their development is still relatively new. In addition, metamaterial technology breakthroughs are very high and widespread because of their renewable materials and structures [4]. However, in the development of science and technology, pure metamaterials in their current applications are still below the requirements of modern technology with high quality. This is a problem that needs to be researched to improvise metamaterials into superior materials. In addition, the high engineering properties of metamaterials can create hybrid materials from a mixture of other compound elements as advanced renewable materials [5]. This has the potential to increase the sensitivity of metamaterials to become more active and responsive with superior characteristics.

One-dimensional nanoscale zinc oxide (ZnO) semiconductor material has attracted much attention from researchers because it has great potential in aspects of study and application. Potentially from a fundamental study aspect, ZnO is the right candidate to be used as a mixture of hybrid metamaterials. This is because ZnO has good chemical and thermal stability properties to be used as an analyte sensor [6, 7]. ZnO semiconductive transparency can provide good electrical conductivity and optical properties to increase sensor sensitivity. In addition, ZnO doping can improve nanoscale optical physical properties such as increasing electron mobility, modifying photocatalytic activity, faster, more stable catalyst space, and reducing charge carrier recombination [8, 9]. The positive influence based on the potential use of metamaterials for ZnO provides an opportunity for this research to realize technology based on advanced, renewable materials. Therefore, this research aims to develop a hybrid sensor media based on ZnO thin films and renewable metamaterials. This design is expected to provide opportunities as a semiconductive sensor with high sensitivity from the characteristics of renewable materials.

## Hybrid metamaterial configuration

The metamaterial is designed as a square pattern split ring resonator (SRR) structure with a 3×3 arrangement integrated with a thin film of ZnO as shown in Figure 1. The designed metamaterial structure consists of two resonator rings made of pure copper ($\varepsilon_r$ = 1) which are placed on the top surface of a quartz dielectric substrate ($\varepsilon_r$ = 3.8). Meanwhile, a thin film of ZnO is between the SRR layer and the substrate. The dimensions of the multi-SRR-square metamaterial structure are designed based on the smallest wavelength of the operating frequency of 0.05 – 9 GHz. Multi-SRR-square and ZnO thin film have different ring radii, widths, and thicknesses as shown in Table 1.

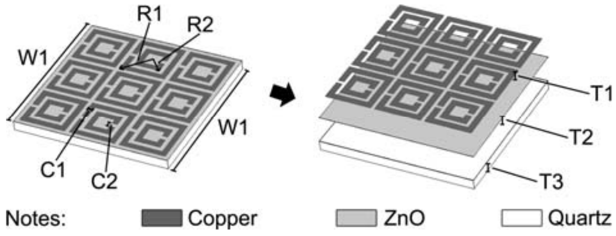

Fig.1. Design and structural details of an integrated metamaterial by multi-SRR-square and ZnO thin film ZnO

Table 1. Metamaterial structure geometry integrated by multi-SRR-square and ZnO thin film

| Parameter | Value (mm) | Parameter | Value (mm) |
|---|---|---|---|
| C1 | 0.61 | T1 | 0.50 |
| C2 | 0.43 | T2 | 0.0002 |
| R1 | 3.43 | T3 | 1.00 |
| R2 | 2.57 | W1 | 16.28 |

Metamaterial characteristics are determined based on spectral parameter data obtained from simulations of EM wave radiation on metamaterial samples. Reflection (S11) and transmission (S12) spectrum data are processed to obtain the permittivity, permeability, and refractive index properties of the metamaterial using the modified Nicolson-Ross-Weir equation as follows [10]:

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

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

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

where: $k_0$ – wavenumber, $t_m$ – propagation length, $\varepsilon_r$ – relative permittivity, $\mu_r$ – relative permeability, dan $n$ – refractive index.

## Extreme double-negative indexed

The ZnO thin layer used has a great influence on the optical properties parameters of the double-negative (DNG) multi-SRR-square metamaterial. The use of this material provides an extreme increase in relative permittivity up to a resonance depth of -1.3453 × $10^4$ at a frequency of 0.82 GHz (see Figure 2a). Materials with higher permittivity have a stronger response to electric fields. Apart from that, the integration of this hybrid metamaterial also affects the relative permeability properties with a quite extreme resonance depth of -0.1214 × $10^4$ at a frequency of 1.83 GHz (see Figure 2b). This permeability characterizes the material as responding strongly to the induction of a given magnetic field. The interaction between relative permeability and relative permittivity determines the increasingly optimal refractive index of the metamaterial. As the DNG parameters of permittivity and permeability of a material increase, the negative refractive index also increases. It can be seen that the refractive index of the metamaterial has a fairly high negative value of -0.0193 × $10^4$ at a frequency of 1.82 GHz (see Figure 2c). This proves that ZnO thin films offer advantages from their electronic, optical, and magnetic properties that enable metamaterial properties to respond constructively [11].

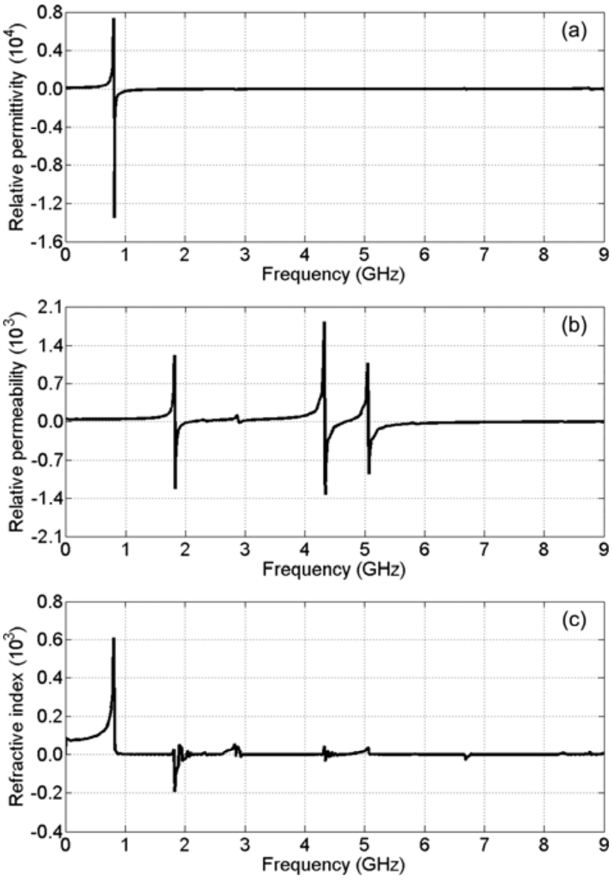

Fig.2. Resonance optical properties of integrated metamaterials: (a) permittivity, (b) permeability, and (c) refractive index

Table 2. Negative resonance in extreme DNG metamaterials

| Properties | Frequency (GHz) | Value ($10^4$) |
|---|---|---|
| Relative permittivity | 0.82 | -1.3453 |
| Relative permeability | 1.83 | -0.1214 |
| Refractive index | 1.82 | -0.0193 |

## Metamaterial-ZnO integrated sensor

The sensor design was carried out by integrating a multi-SRR-square metamaterial and a thin film of ZnO with a circuit path and the addition of several material components as shown in Figure 3. The choice of circuit shape and power supply port is based on optimizing sensor performance to maximize power transmission absorption. The epoxy sample container (1 × $10^3$ $mm^3$) can only accommodate 1 ml of analyte sample formed concentrically to the multi-SRR-square surface and a thin film of ZnO. In addition, additional grounding elements are applied to the back of the substrate. Details of the size of the sensor structure can be seen in Table 3.

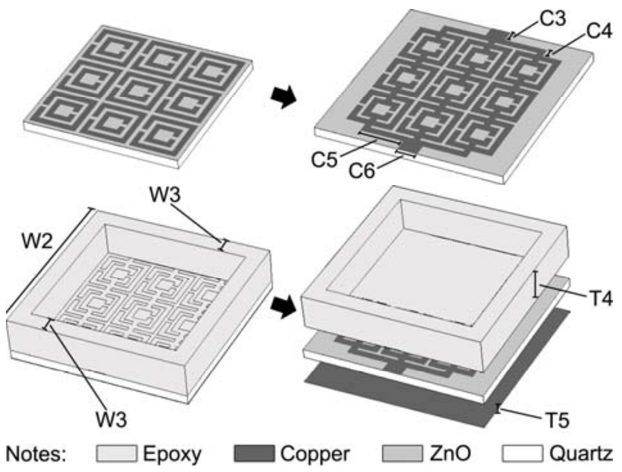

Fig.3. Design of integrated metamaterial structure by multi-SRR-square and ZnO thin film for analyte electrolyticity detection

Table 3. Complete parameters of the metamaterial sensor structure

| 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 |

## Guaranteed initial performance

Figure 4 shows the reflection, transmission, and absorption spectra of the integrated metamaterial-ZnO sensor design for sampleless measurements.

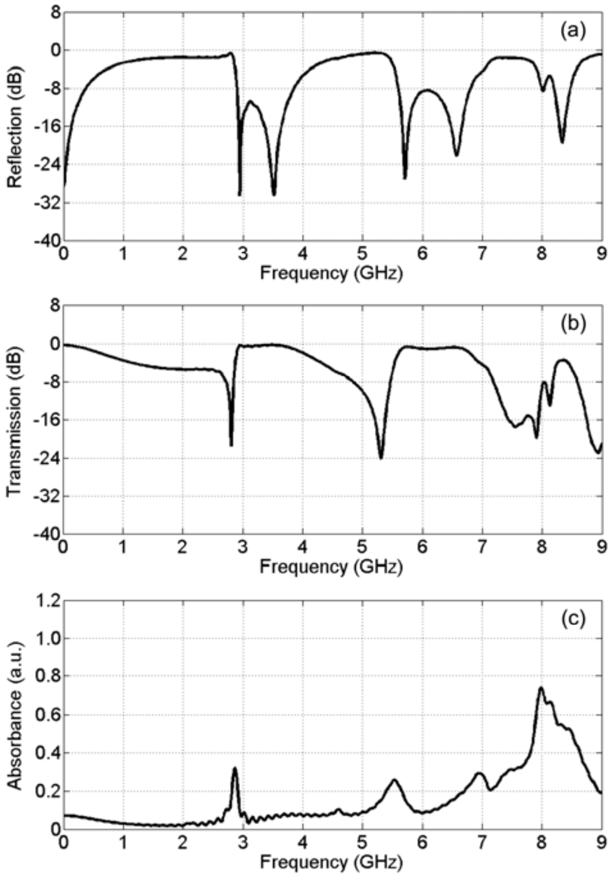

Fig.4. Performance of integrated metamaterial sensor for without samples: (a) reflection; (b) transmission; and (c) absorbance

The resulting reflection spectrum describes the power that is reflected or not absorbed through the sensor structure. Based on the reflection spectrum, the sensor structure has two resonance depths at the same bandwidth. The highest reflected power is at the resonance frequency 2.95 – 3.55 GHz of -30 dB (see Figure 4a). Meanwhile, at the high frequency of 8.45 GHz, the reflected power decreased by 4.5 dB. This indicates that more power distribution is reflected in the sensor structure with a larger wavelength. Apart from that, the transmission spectrum also describes the distribution of power that is transmitted through the sensor structure without being absorbed or reflected [12]. Based on the transmission spectrum, the resonance depth of -21.8 dB at the low frequency of 2.85 GHz tends to increase towards the higher frequency of 5.36 GHz by -24.6 dB (see Figure 4b). However, at high frequencies of 8 - 9 GHz the transmission spectrum has many defective resonances with relatively decreased depth. This is proven by the presence of a peak in the absorption spectrum at high frequencies, where the power distribution is absorbed 73% higher at a high frequency of ~8 GHz by the sensor structure at a small wavelength (see Figure 4c).

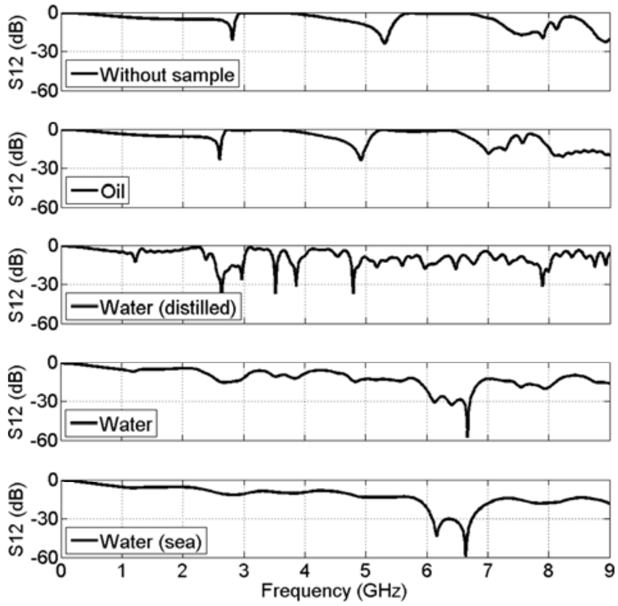

Fig.5. Changes in the transmission spectrum for some sample experiments

There appears to be a clear difference in resonance frequency and depth due to changes in electrolyticity or electrical conductivity of samples in the form of oil (0 S/m), distilled water (5.55 × $10^{-6}$ S/m), water (1.59 S/m), and seawater (3.53 S/m) (see Figure 5). In general, the shift in resonance frequency and transmission spectrum intensity occurs due to differences in the electromagnetic properties of materials, such as permittivity and material permeability. The resonant frequency shift in this sensor is analogous to an LC circuit where the frequency response will change with the inductance and capacitance [13]. The resonant frequencies of the four types of test materials are relatively more significant, except for water and seawater (6.18 – 6.62 GHz). However, this is quite interesting because sensor integrated have been proven to be able to identify these two types of water. There is no difference in the permittivity and permeability values between the two. So it can be concluded that the cause of the slight difference in the resonance frequency of the two types of water is the difference in electrical conductivity of the water and seawater samples.

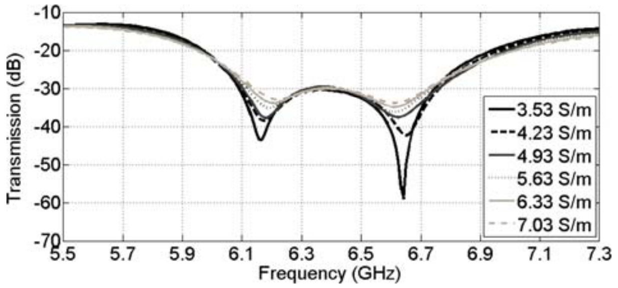

Fig.6. Seawater sample measurement test results are based on electrolyticity or variations in electrical conductivity

Figure 6 shows a fairly good relationship between resonance frequency depth and electrical conductivity. Very significant changes in the depth of the resonance frequency occurred in sensor tests on seawater samples with different electrical conductivities from 3.53 – 7.03 S/m. The resonance depth decreases as the electrical conductivity increases. This is because the power distribution transmitted through the sensor structure experiences power absorption in the form of an induced electric field by samples that have greater electrical conductivity [14]. In addition, the frequency shift of the two resonance depths also becomes narrower as the electrical conductivity increases. The detection sensitivity of seawater samples reached 0.0313 GHz, this shows that a resonance frequency shift of 3.13 MHz occurs for every 0.07 S/m increase in electrical conductivity.

## Conclusion

The metamaterial structure integrated by multi-SRR-square and ZnO thin film has been successfully designed and simulated. The extreme DNG metamaterial characteristics are obtained with a relative permittivity value of $-1.3453 \times 10^4$ at 0.82 GHz, a relative permeability of $-0.1214 \times 10^4$ at 1.83 GHz, and a refractive index of $-0.0193 \times 10^4$ at 1.82 GHz. The performance of the metamaterial-ZnO hybrid sensor media provides resonance bandwidth in the frequency ranges of 2.89 – 3.52 GHz, 5.28 – 6.54 GHz, and 7.57 – 8.46 GHz. Meanwhile, the highest absorption spectrum 73% was obtained at a frequency of ~8 GHz. Sensor testing on oil samples experienced a transmission spectrum shift from results without samples. Distilled water experiences fluctuations due to the sensor's response to small electrical conductivity. Water and seawater have relatively the same resonance at a frequency of 6.18 – 6.62 GHz. Testing the electrolyticity of analytes on seawater samples experienced a reduction in resonance depth and a shift in resonance frequency of 3.13 MHz.

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