Chapter 8

Conclusions and Future Scope

8.1 Summary and Conclusions

This work provides a comprehensive investigation into the appearance of **negative permittivity** in metal free real, processable/scalable materials. A variety of material systems including single-phase ceramics and functional ceramic-based composites were synthesized in this study. Some important conclusions drawn from the experimental results related to their structural, dielectric, and electromagnetic properties are summarized below.

1. The system, **$La_{2-x}Sr_xNiO_4$**, exhibited **negative permittivity near and above room temperature**, with a distinct **semiconductor-to-metal transition (SMT)** occurring around **450 °C**, consistent with prior studies. **Strontium (Sr) doping** was employed to enhance **electrical conductivity** and reduce the **synthesis temperature**. The Sr-doped samples followed similar dielectric trends but showed **lower SMT transition temperatures**, confirming a conductivity-dependent transition. This **semiconductor-to-metal transition** was found to significantly affect **EMI shielding**, with higher conductivity leading to **reflection-dominated shielding performance**.
2. In **perovskite oxide $LaNiO_3$**, similar behavior was observed. However, the **classical Drude model** was **inadequate** for fitting the dielectric response of this conductive ceramic. A **modified Drude model** was therefore introduced, yielding a more accurate match with experimental data.
3. All single-phase materials showing **negative permittivity** also exhibited **inductive behavior**, as confirmed by **positive imaginary impedance (Z″)** and **EIS (electrochemical impedance spectroscopy) fitting**.
4. Two different **composite systems** were proposed as **alternatives to metals** for electromagnetic shielding. In particular, **conductive ceramic $LaNiO_3$** was used as a filler, offering **multifunctional properties**. As the **$LaNiO_3$ filler content** increased,

permittivity shifted from **positive to negative**, once a threshold in **electrical conductivity and filler volume** reaches the threshold.

5. In **SnO_2/graphene (SGR) composites**, the minimum required **conductivity for negative permittivity** was identified as approximately **0.5 S/m**.

    - **SGR-2** displayed **epsilon-near-zero (ENZ)** behavior and was **absorption-dominated**.
    - **SGR-4** showed **extremely negative permittivity** with **very high conductivity**, indicating **reflection-dominated shielding**.

General Remark:

a) All materials with sufficiently high **electrical conductivity** tend to exhibit **negative permittivity**, and vice versa. This has been observed in cases such as La_2NiO_4 in both doped and undoped forms, as well as $LaNiO_3$, as demonstrated in **LED-based experiments**.

b) A strong **correlation** was consistently observed between **conductivity**, **negative permittivity**, microstructure and **shielding effectiveness (SE)**. Materials with greater conductivity exhibited **stronger negative permittivity** and more effective **reflection-based shielding**.

c) The **minimum DC conductivity required for epsilon-negative (ENG) behavior** was estimated to be approximately **0.2 to 0.5 S/m**, which serves as a benchmark for identifying or synthesizing materials exhibiting confirmed **negative permittivity**. This range aligns well with our experimental measurements and is broadly supported by values reported in the available literature. However, **more**

quantitative and systematic experimental studies are needed to further substantiate and refine these findings.

S.N.	Composition		Synthesis route	Synthesis Temperature T (°C)	σ_{total} (S.m^{-1})	ε_r	SE (dB)	Shielding Mode (8-12 GHZ)	Commercially Required Shielding (dB)
1	$La_{2-x}Sr_xNiO_4$	X=0	Solid state reaction method	1500	30	-4.5 x10^6	11	Reflection	*30-60 dB*
		X=0.1			66	-16 x10^6	-		
		X=0.3			79	-27 x10^6	47		
2	$LaNiO_3$	Pristine	Sol-gel	1000	130	-120 x10^6	33	Reflection	
3	SnO_2-$LaNiO_3$ composites	10% LaNiO3	Wet-grinding method	700	8.9x10^{-2}	+ 200	30	Absorption	
		20% LaNiO3			18	-8x10^2	43	Absorption	
		30% LaNiO3			45	-1.5x10^5	38	Reflection	
4	SnO_2-Graphene NPs	2% Graphene	Wet grinding method	200	1.2	-120	42	Absorption	
		4% Graphene			6400	-5x10^{10}	75	Reflection	

Figure 8.1 Summary of key experimental findings for the synthesized materials, including synthesis temperature, dielectric permittivity, electrical conductivity, and electromagnetic shielding effectiveness (SE). The data illustrate the relationship between processing conditions and the resulting functional properties, with SE values also compared against commercially required thresholds for effective electromagnetic shielding applications.

These findings provide **practical design guidelines** for the development of ENG and ENZ materials without the dependency on metals.

8.2 Future Outlook and Challenges

This thesis presents new insights into negative permittivity materials or ENG materials, and its applications in electromagnetic shielding and coil-less inductors, but several challenges remain. Materials with stronger negative permittivity exhibit higher electromagnetic reflectivity and very high inductance, controlled mainly by conductivity.

A key issue is the absence of a theoretical and experimental framework tailored to conducting ceramics. Existing models, such as the classical **Drude theory** developed primarily for **metals,** often fail to describe ceramic ENG systems accurately. For example, $LaNiO_3$ shows significant deviations from classical predictions at low frequencies. Although a modified Drude model is proposed here to better capture electron interactions, a more comprehensive theory that includes ceramic-specific mechanisms like polaronic transport, electron-electron interactions, and microstructural inhomogeneity is needed.

Experimentally, standard methods for measuring inductance and dielectric properties, adapted from semiconductor research, may not reflect the true electromagnetic behavior of conducting ceramics, potentially causing discrepancies between theory and experiment.

Scaling these materials while preserving negative permittivity and ensuring stability under environmental exposure and temperature variation presents additional challenges. The observed high inductance in ENG ceramics still puzzling and a matter of further research have underscored their potential for compact passive components and demands new integration strategies.

Materials exhibiting ENZ properties are very important for application like perfect absorber. Overall, the tunability of negative permittivity in ceramic-based ENG materials opens avenues for multifunctional metamaterials. **Researcher can further look into wearable and flexible ENG materials for medical and defence applications**. Overall, As **Sir John Pendry** stated, "*metamaterials are better than silver"*—a vision progressively realized through doping, composite engineering, and nanostructuring. These approaches enable optimization of ENG materials for applications ranging from cloaking and EMI shielding to advanced sensing and coil-less inductors.