Dielectric Relaxation Dynamics in Silver Nanoparticle-Doped Chitosan Films

Can Koral[1*], Michela Marsico[2], Zahra Mazaheri[3,4], Gian Paolo Papari[3,4], Antonello Andreone[3,4], Roberto Teghil[2], Angela De Bonis[2]

[1]*Department of Health Sciences, University of Basilicata, 85100, Potenza, Italy;*
[2]*Department of Basic and Applied Sciences, University of Basilicata, 85100, Potenza, Italy;*
[3]*Department of Physics, University of Naples "Federico II", 80126, Napoli, Italy;*
[4]*National Institute for Nuclear Physics, Division of Naples, 80126, Napoli, Italy*

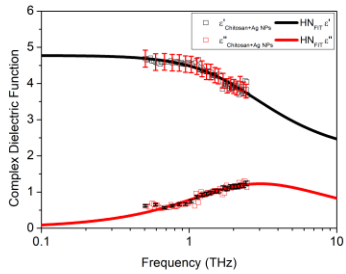
Abstract: This study explores Ag-NPs-doped chitosan's electro-optical properties using THz spectroscopy. Drude-Smith, Havriliak-Negami, and Cole-Cole analyses revealed increased conductivity and enhanced, broadened relaxation dynamics. These findings demonstrate that Ag-NPs incorporation fundamentally alters chitosan's dielectric mechanisms, suggesting potential for advanced terahertz applications.

Nanoparticle-embedded polymers have emerged as promising candidates, offering a versatile platform to engineer dielectric properties and manipulate THz wave interactions. Chitosan (CT) has garnered significant attention due to its exceptional properties. This study employs terahertz Time Domain Spectroscopy (THz-TDS) to investigate the enhanced relaxation dynamics in silver-doped chitosan nanocomposites, aiming to better understand the influence of the electronic properties and charge transport mechanisms[1].

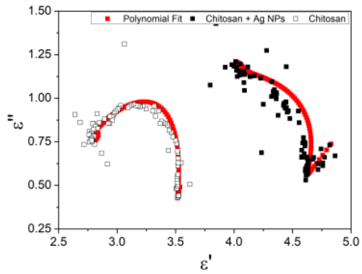
Ag nanoparticles were synthesized using pulsed laser ablation directly in the chitosan liquid matrix to prepare free standing films. Terahertz Time Domain Spectroscopy served as the primary experimental technique, enabling us to determine the complex dielectric function of our samples over a broad frequency range.

After extracting the dielectric function of the CT and the CT-Ag composite, we used the Drude-Smith (DS) equation to describe the frequency-dependent conductivity of the samples, providing insights into the dynamics of free and bound charge carriers. Moreover, we employed the Havriliak-Negami (HN) equation to model the complex dielectric function, accounting for the distribution of relaxation times(Fig 1,a). From the outcome of the initial analysis, we further investigated the complex impedance and dielectric modulus analysis to gain deeper insights into the electrical relaxation processes taking place in the composites. Finally, to complete and visualize the dielectric response, we utilized Cole-Cole plots, providing a clear graphical representation of the dielectric data and facilitating the analysis of relaxation processes and impedance characteristics(Fig 1,b).

The Drude-Smith model revealed a significant increase in high-frequency conductivity for the composite compared to pure chitosan, attributed to silver nanoparticles and structural defects, while confirming localized charge carriers for both films, which remained insulating. Havriliak-Negami analysis demonstrated enhanced relaxation dynamics in the composite, evidenced by a decreased relaxation time and a broadened distribution of relaxation times despite both materials exhibiting near-Debye behavior. Furthermore, Cole-Cole plots visually corroborated these findings, transitioning from a Debye-like semicircle in pure chitosan to a distinct quarter-circle with a linear region for the composite, indicating broader relaxation time distributions, interfacial polarization effects, and potential DC conductivity. Complementary complex impedance and modulus analyses further underscored significant departures from ideal Debye behavior in the composite, supporting the role of heterogeneity, interfacial effects, and altered local electric field distributions[2]. These results collectively provide strong evidence that silver nanoparticle incorporation fundamentally alters the dielectric properties and relaxation mechanisms of chitosan, showcasing their potential for advanced terahertz applications.



a)



b)

Fig.1 | a, Havriliak-Negami model fits to the complex dielectric function of the silver nanoparticle-doped chitosan composite film. b, Cole-Cole plot for the pure chitosan film and the silver nanoparticle-doped chitosan composite film.

Contacts:

Can Koral (can.koral@unibas.it)

References

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