

Which Parameters Control Maxwell–Wagner–Sillars Relaxation?

A Finite-Element Study of Carbon-Coated Silica / Epoxy Nanocomposites

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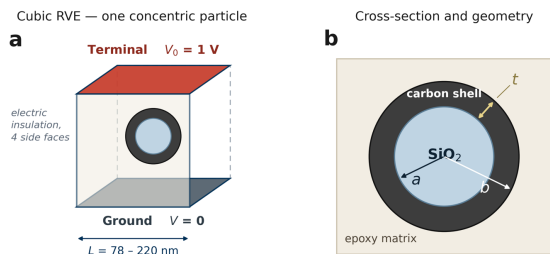


Filler loading sets HOW MUCH interfacial polarization there is.
The core-to-particle ratio $\beta = (a/b)^3$ sets HOW FAST it responds.
Neither brings the relaxation within 700× of the measurement band.

1 The puzzle

Core–shell nanocomposites are designed around Maxwell–Wagner–Sillars polarization — yet measured spectra show no loss peak and no step in ϵ' . Absent, or just out of reach?

2 The model



Electric Currents, frequency domain, COMSOL Multiphysics® 6.2. One coated sphere per cubic cell; L set by ϕ , so loading is exact.

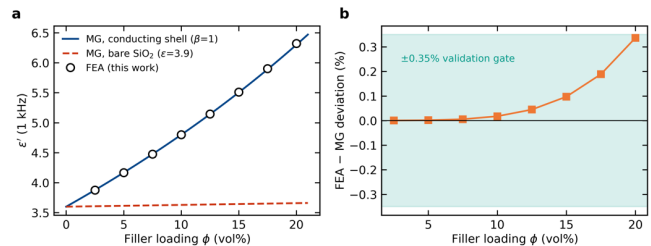
17 RVEs · 1 mHz – 1 GHz · 1337 converged points, validated against the analytic coated-sphere solution to four significant figures in ϵ' .

Phase	ϵ_r	σ (S/m)
SiO ₂ core	3.9	1×10^{-14}
Carbon shell	12	1×10^2
DGEBA–DETA epoxy	3.6	1×10^{-8}

Sweep	Levels	Held fixed
ϕ	2.5–20 vol% (8)	d = 70, t = 5 nm
d core	35, 50, 70, 100 nm	ϕ = 10%, t = 5 nm
t shell	5, 10, 15, 20, 25 nm	ϕ = 10%, d = 70 nm

3 The gain is real — and the shell does all of it

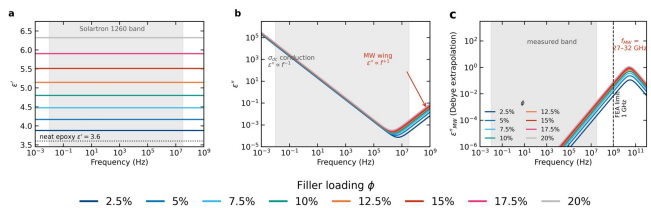
ϵ' rises 3.60 → 6.32 (+75.6%) at 20 vol%, matching Maxwell–Garnett for a perfectly conducting inclusion to 0.34% with no free parameters. Bare silica gives only +1.6%.



ϵ' at 1 kHz vs loading, against the two analytic limits.

4 ... but the relaxation is off-scale

ϵ' is flat to 0.05% over twelve decades. In ϵ'' , DC conduction (slope –1) meets a rising +1 flank at 2.5 MHz — the wing of a Debye peak whose apex sits at 23–70 GHz.



$\epsilon'(f)$ and $\epsilon''(f)$ for all eight loadings; peaks reconstructed from the +1 flank.

5 Geometry sets the timescale — through one dimensionless ratio

Absolute size is irrelevant: f_{MW} is a function of β alone.

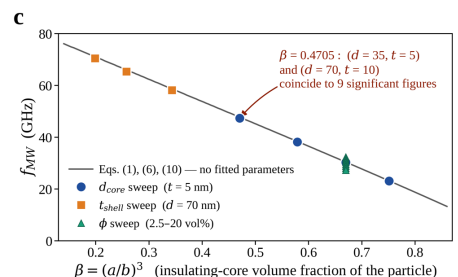
Two independently built cells — d = 35 / t = 5 nm and d = 70 / t = 10 nm — share $\beta = 0.4705$ but differ twofold in size. Both relax at 47.3812 GHz, agreeing to better than 0.01 ppm.

ϵ' itself does not move: 2.1 ppm across a 2.9× core-diameter and 5× shell-thickness range.

Why: interfacial charging is the RC time of the particle itself, not a transport time across a fixed distance.

Conclusions

- Loading sets the magnitude; geometry does not.
- $\beta = (a/b)^3$ sets the timescale; absolute size is irrelevant.
- $f_{MW} = 23.1\text{--}70.5$ GHz — 700× above the band: saturated, not absent.



Next: SiO₂@C@SiO₂

An outer silica shell should break the ϵ' –leakage lock. Analytic prediction, not yet FEA: σ_{dc} falls to 8.6×10^{-9} S/m and the impedance corner drops from 49.9 Hz to 32–37 Hz — a corner immovable across all 17 cases.

[1] K. W. Wagner, Arch. Elektrotech. 2, 371 (1914). [2] R. W. Sillars, J. Inst. Electr. Eng. 80, 378 (1937). [3] A. Sihvola, Electromagnetic Mixing Formulas and Applications, IEE (1999). [4] R. A. Gerhardt, J. Phys. Chem. Solids 55, 1491 (1994). [5] G. Patil and R. A. Gerhardt, ACerS Annual Spring Meeting, Seattle WA (2026). Modelled in COMSOL Multiphysics® 6.2. The authors thank the Gerhardt Research Group at Georgia Tech for useful discussions.