Dielectric properties of composite films of $MgWO_4$, $FeWO_4$, $Zr(WO_4)_2$

K.H. Norian

Electrical and Computer Engineering Department, 19 Memorial Drive West, Lehigh University
Bethlehem, PA, USA 18015, Fax (610) 758-6279, Tel (610) 758-4082, E-mail **khn0@lehigh.edu**

Abstract

Resonance spectra were obtained to determine the dielectric properties of composite polymer-dielectric films, where the dielectric was $MgWO_4$, $FeWO_4$ or $Zr(WO_4)_2$. The resonant frequencies for the films were in the 383 to 473 MHz range. The relaxation times, the real and imaginary permittivities, and dissipation factor and conductivities for the three materials were determined.

Introduction

Dielectric composite materials made of epoxy resin as matrix, to which a ceramic filler powder is added to control the dielectric constant of the substrate on which electronic circuits are fabricated, are widely used in electrical engineering. Composites with ceramics fillers of alumina and boron nitride powders[1], strontium titanate[2], barium titanate[3], bismuth manganite and lead-titanate[4] have been fabricated and their complex dielectric constant has been studied.

In the present work resonance spectra[5] were experimentally obtained, to determine the complex relative permittivity ε_r^* as a function of frequency, of composite polymer-dielectric films, where the dielectric was $MgWO_4$, $FeWO_4$ or $Zr(WO_4)_2$. Such films have potential application in planar substrates where the electrical properties of circuits that are built on the substrates depend in part on the permittivity of the latter.

Theory

The current-voltage relationship of a parallel plate capacitor with air as the dielectric is

$$\underline{I} = j\omega C_o \underline{V} \qquad (1)$$

where ω is the angular frequency, C_o the capacitance, and $\underline{I}$ and $\underline{V}$ are the current and voltage phasors.

With a dielectric material other than air present between the capacitor electrodes, the complex relative dielectric permittivity ε_r^*, is introduced to account for the lag between the displacement and polarization in the dielectric. The current-voltage relationship can now be given as

$$\underline{I} = j\omega \epsilon_r^* C_o \underline{V} \qquad (2)$$

$$\underline{I} = j\omega(\epsilon_r' - j\epsilon_r'') C_o \underline{V} \qquad (3)$$

where ϵ_r^* has a real and imaginary component. The current I then has two components

$$\underline{I} = \underline{I_L} + \underline{I_D} \qquad (4)$$

where the quadrature, displacement current $\underline{I_D} = j\omega\varepsilon_r' C_o \underline{V}$, and the in phase, loss current $\underline{I_L} = \omega\epsilon_r'' C_o \underline{V}$ (see Fig. 1).

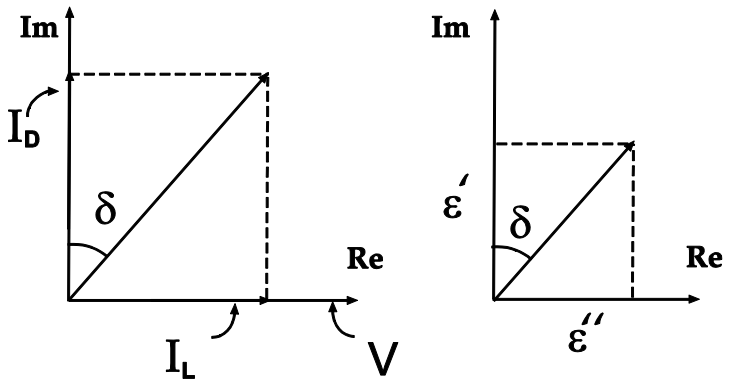

Fig. 1 Phasor representation of current, voltage and complex permittivity for dielectric film

I_L contributes to the true power dissipated in the dielectric, and I_D to the VAr , reactive power supplied to the component over part of a quarter cycle and then returned to the power source over the next quarter. Then δ in Fig. 1 is the phase angle between the total current I and the quadrature current I_D and the dielectric dissipation factor tan δ is given by

$$\tan\delta = \frac{\omega\varepsilon_r'' C_o V}{\omega\varepsilon_r' C_o V} = \frac{\varepsilon_r''}{\varepsilon_r'} \qquad (5)$$

The capacitor can also be represented by the series combination of a resistor, of resistance R, an inductor of inductance L, and a capacitor of capacitance C. Then for an AC voltage V across the capacitor the current through the capacitor is given by

$$\underline{I} = \frac{\left\{1 - j\left(\frac{\omega L}{R} - \frac{1}{\omega CR}\right)\right\}\underline{V}}{R\left\{1 + \left(\frac{\omega L}{R} - \frac{1}{\omega CR}\right)^2\right\}} \qquad (6)$$

Considering the RLC circuit, at and around the resonant frequency equations that apply are:

$$Q = \frac{\omega_o L}{R} \;; \quad Q = \frac{1}{\omega_o RC} \;; \quad Q = \frac{\omega_o}{BW} \qquad (7)$$

where ω_o is the resonant angular frequency, BW the bandwidth in frequency between the 3dB points in the resonance curve, and Q is quality factor of the circuit. Equs (6) and (7) give

$$\underline{I} = \frac{\left\{1 - jQ\left(\frac{f}{f_o} - \frac{f_o}{f}\right)\right\}\underline{V}}{R\left\{1 + Q^2\left(\frac{f}{f_o} - \frac{f_o}{f}\right)^2\right\}} \qquad (8)$$

where f is the cyclic frequency and f_o the resonant frequency.

Equating the real and imaginary parts of equs (3) and (8) gives

$$\epsilon_r' = \frac{Q\left(\frac{f_o}{f} - \frac{f}{f_o}\right)}{2\pi f C_o R\left\{1 + Q^2\left(\frac{f}{f_o} - \frac{f_o}{f}\right)^2\right\}} \qquad (9)$$

$$\epsilon_r'' = \frac{1}{2\pi f C_o R\left\{1 + Q^2\left(\frac{f}{f_o} - \frac{f_o}{f}\right)^2\right\}} \qquad (10)$$

Then substituting for ϵ_r'' and ϵ_r' in equ (5) gives the dissipation factor

$$\tan\delta = \frac{1}{Q\left(\frac{f_o}{f} - \frac{f}{f_o}\right)} \qquad (11)$$

Experimental

Capacitors of dielectric composite films of polymer-tungstate powder between copper plates were fabricated. The tungstates were 400 mesh powders of $MgWO_4$, $FeWO_4$ or $Zr(WO_4)_2$ in a polymer binder. The resulting composite dielectric film consisted of an epichlorohydrin-bisphenol A resin mixed with a telluride and cured with 2,4,6-Tris(dimethyl-amino-methyl)phenol. The dialectric films were about 0.18 mm thick. The impedance *Z*, resistance *R*, and reactance *X*, of the capacitor as a function of frequency were measured up to 2.5 GHz using a vector network analyzer. At the resonance frequency the reactance is zero and the magnitude of the impedance $|\underline{Z}|$ is equal to the pure resistance *R*.

Results and Discussion

Table 1 shows the experimentally determined values of resonant frequency and quality factor for the different films. Reproducibility of resonant frequency between films of the same material was less than 1%.

	f_o ±2 MHz	Q
$MgWO_4$	473	25.89
$FeWO_4$	448	13.18
$Zr(WO_4)_2$	383	9.34

Table 1 Resonant frequency vs quality factor for composite films

The results were used in Equs 9 and 10 to plot the variation of the real and imaginary permittivity for each material as a function of frequency (See Fig 2/Table 2 and Fig 3/Table3)

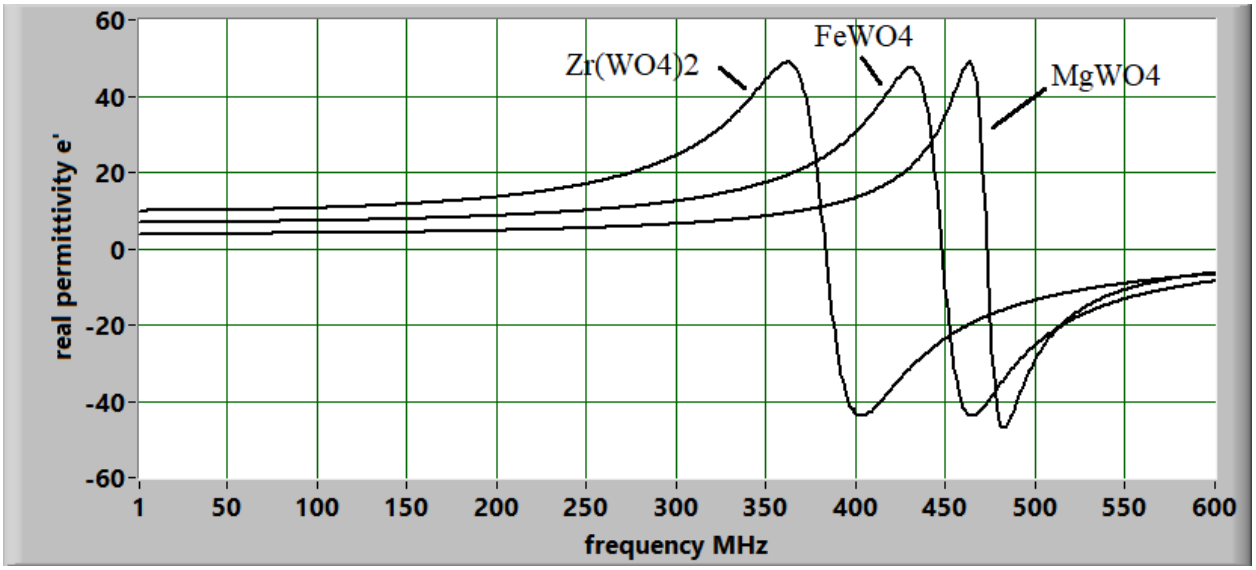

Fig 2 variation of ϵ_r' with frequency for films of polymer-tungstate composites

$Zr(WO_4)_2$				
f (MHz)	1	100	364	$f_{resonance}$ = 383
ϵ_r'	9.90	10.60	ε'_{max} = 48.57	0
$FeWO_4$				
f (MHz)	1	100	424	$f_{resonance}$ = 448
ϵ_r'	6.91	7.24	ε'_{max} = 44.97	0
$MgWO_4$				
f (MHz)	1	100	463	$f_{resonance}$ = 473
ϵ_r'	3.86	4.04	ε'_{max} = 49.02	0

Table 2 select data on ϵ' vs frequency derived from Fig 2

The results of Fig 2 show that the real permittivity is approximately constant between 1 and 100 MHz, for instance rising from 9.90 to 10.60 in this range for the $Zr(WO_4)_2$ film. After 100MHz, for $Zr(WO_4)_2$, ϵ_r' increases exponentially with frequency to a maximum of 48.57 at 364MHz before descending to zero at the resonance frequency of 383 MHz. values for ϵ_r' for $FeWO_4$ and $MgWO_4$ are given in Table 2 and within the 1 to100 MHz range, ϵ' is approximately 4, 7 and 10 for the $MgWO_4$, $FeWO_4$ and $Zr(WO_4)_2$ films, respectively.

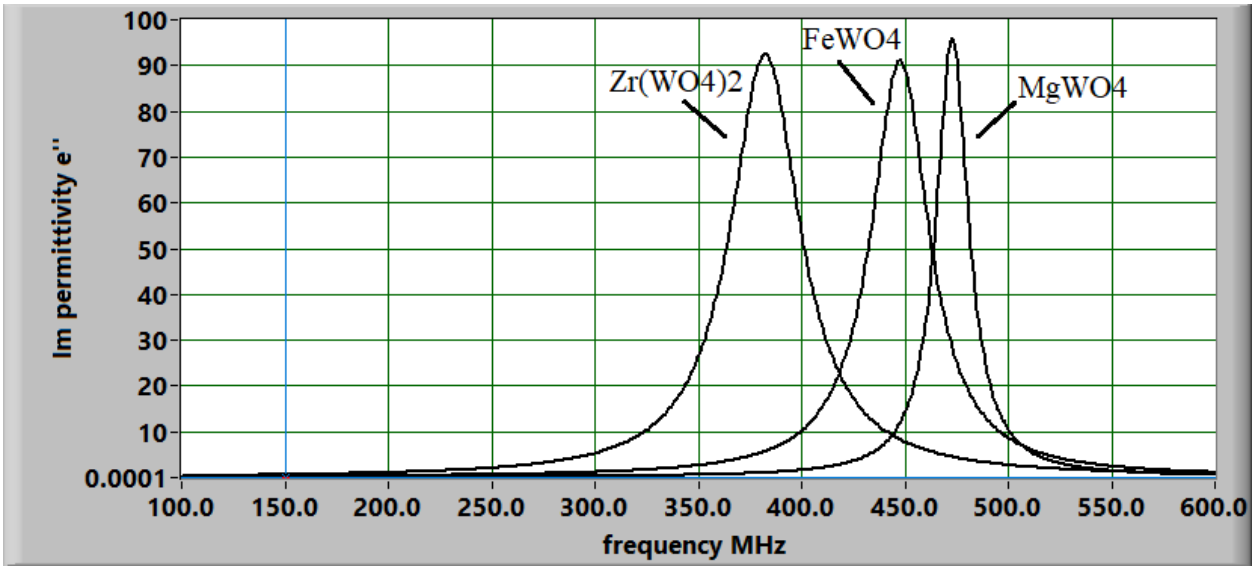

Fig 3 ϵ_r'' vs frequency for polymer-tungstate composite films

$Zr(WO_4)_2$				
f (MHz)	1	100	200	$f_{resonance}$ = 383
ϵ_r''	0.0028	0.32	1.04	92.42
$FeWO_4$				
f (MHz)	1	100	200	$f_{resonance}$ = 448
ϵ_r''	0.0012	0.13	0.36	91.11
$MgWO_4$				
f (MHz)	1	100	200	$f_{resonance}$ = 473
ϵ_r''	0.0003	0.036	0.10	96.14

Table 3 select data on ϵ'' vs frequency derived from Fig 3

At 1 MHz ϵ_r'' for $MgWO_4$ is 0.0003 while for $Zr(WO_4)_2$ ϵ_r'' is an order of magnitude larger at 0.0028. ϵ_r'' increases with frequency and at 200MHz it is still an order of magnitude larger for $Zr(WO_4)_2$ compared to $MgWO_4$. At resonance ϵ_r'' is of the same order of magnitude for all three materials. As the imaginary permittivity is proportional to losses the largest energy loss occurs in the $Zr(WO_4)_2$ based films.

The dissipation factor tan δ is given in Fig 4. It shows that the dissipation is highest at resonance but it is spread over a larger frequency band for the $Zr(WO_4)_2$ films and the smallest band for the $MgWO_4$ ones.

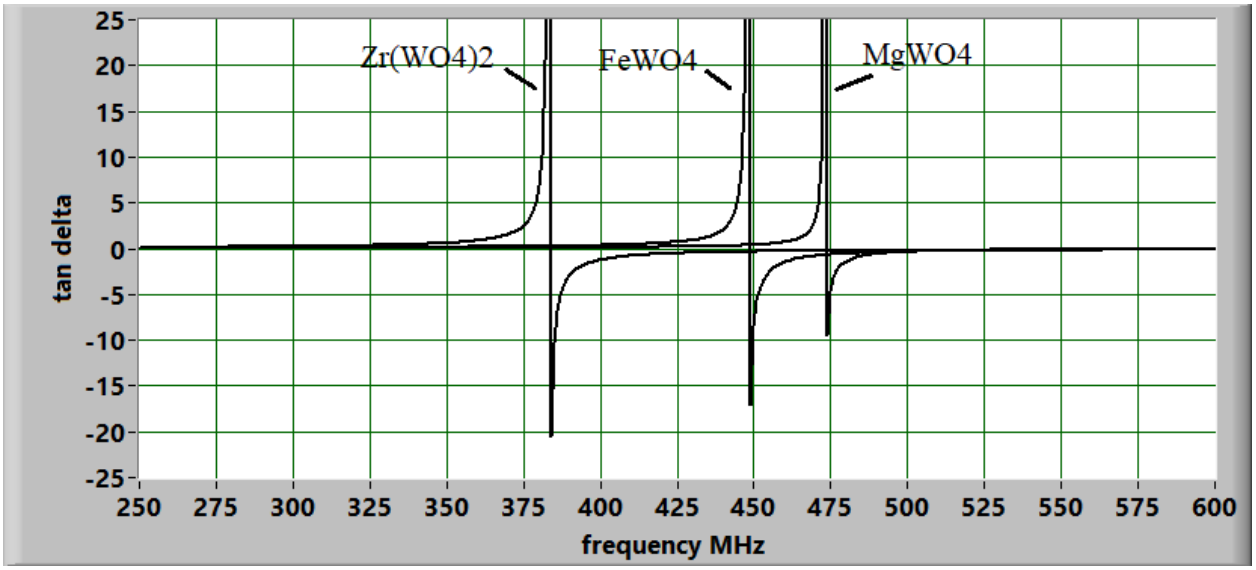

Fig 4 dissipation factor tan δ as a function of frequency

$Zr(WO_4)_2$											
σ (1/Ωm)	3.98 E-10	1.81 E-9	4.82 E-9	1.16 E-8	2.88 E-8	8.86 E-8	5.13 E-7	1.96 E-6	1.19 E-6		
f (MHz)	50	100	150	200	250	300	350	384 R	400		
$FeWO_4$											
σ (1/Ωm)	1.67 E-10	7.37 E-10	1.86 E-9	4.05 E-9	8.55 E-9	1.91 E-8	5.13 E-8	2.28 E-7	2.26 E-6	2.24 E-6	
f (MHz)	50	100	150	200	250	300	350	400	447 R	450	
$MgWO_4$											
σ (1/Ωm)	4.66 E-11	2.04 E-10	5.07 E-10	1.08 E-9	2.19 E-9	4.59 E-9	1.09 E-8	3.55 E-8	3.53 E-7	2.53 E-6	2.93 E-7
f (MHz)	50	100	150	200	250	300	350	400	450	473R	500

Table 4 conductivity vs frequency values derived from Fig 3 where R indicates the resonant frequency.

Since conductivity is given by $\sigma = 2\pi f \varepsilon_o \epsilon''$ where ε_o is the permittivity of free space, the data of Fig 3 were used to calculate the conductivity of the films as a function of frequency (see Table 4). These show that from 50 MHz up to just before resonance, conductivity is highest for $Zr(WO_4)_2$ and varies from the order of 10^{-10} to 10^{-7} Ω^{-1} m^{-1} for $Zr(WO_4)_2$ and $FeWO_4$ and from 10^{-11} to 10^{-7} Ω^{-1} m^{-1} for $MgWO_4$. The dielectric relaxation time $\tau = 1/\Delta f$ was obtained from Fig 3 where Δf is the frequency difference between the resonant frequency and the frequency where ϵ'' falls to half its maximum value. The relaxation time was 48, 56 and 111 nanoseconds for the zirconium, iron and magnesium tungstate composite films, respectively. As expected, the longer the relaxation time the sharper the resonance.

References

[1] The dielectric permittivity of ceramic powders used in composite polymers, C. Vanga Bouanga, S.Savoie, H. Couderc, M.F.Frechette, E. David.2011 Annual report conference on electrical insulation and dielectric phenomena, Cancun, Mexico,16-19, Oct. 2011, publisher IEEE, DOI:10.1109/CEIDP.2011.6232756.

[2] $SrTiO_3$-filled PTFE composite laminates for microwave substrate applications, S.Rajesh, K.P. Murali, K.V. Rajani, and R. Ratheesh, Int. J. Appl. Ceram. Technol. 6 [5] 553-561, 2009.

[3] Greatly enhanced permittivity in $BaTiO_3$-epoxy dielectric composites with improved connectivity of ceramic phase, M. Cao, L. Li, W.B. Hong, S.Y. Wu, X.M. Chen, Journal of Materiomics, 7,1-7, 2021.

[4] Influence of interfacial stresses on electrical properties of bismuth manganite-lead-titanate-epoxy composites, A.Z. Szeremeta, A. Nowok, M.Zubko, S. Pawlus, I. Gruszka, Janusz Koperski, A. Molak, Ceramics International, Available online 3 September 2021, https://doi.org/10.1016/j.ceramint.2021.08.376, publisher Elsevier 2021.

[5] Dielectrics, J.C. Anderson, Reinhold Publishing, New York, 1964.