Universal Dielectric Response of Atmospheric Ice Using COMSOL Multiphysics®

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Abstract: Heterogeneous materials with different phases, are conductive and insulating (dielectric), and are physically present in different natural materials as e.g. atmospheric ice. Jonscher's proposed 'universal dielectric response' [1] is not sufficient for such materials, as it only reflects conductivity as a nonlinear function of frequency, whereas at lower temperatures dipolar vibrations are also very sensitive to temperature. In this paper an analytical equation of conductivity as a function of frequency and temperature is used but is shown implicitly which is numerically solved using Comsol. The results are then compared with the experimentally determined conductivity values of atmospheric ice.

1. Introduction

1.1 Dielectric Susceptibility

The complex dielectric susceptibility 'χ_r' is the polarizing ability indicator of any material due to an excitation field is related with complex dielectric constant by $\chi_r = 1 + \varepsilon_r$. By Kramers-Kronig relations[1], the relation between $\chi_r'(\omega)$ and $\chi_r''(\omega)$ can be defined as,

$$\chi_r'(\omega) = \frac{2}{\pi} \int_0^{+\infty} \frac{\omega \chi_r''(\omega)}{\omega^2 - \omega_r^2} d\omega$$

$$\chi_r''(\omega) = -\frac{2\omega_r}{\pi} \int_0^{+\infty} \frac{\chi_r'(\omega)}{\omega^2 - \omega_r^2} d\omega \qquad (1)$$

where 'ω_r' is the reference frequency and 'ω' is the frequency ranging from zero to infinity. Other than Kramers Kronig model, relations like Debye model and its some derivations such as Cole – Cole and Cole - Davidson models can also determine complex permittivity. These

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[1] A Hilbert integral transforms which is valid irrespective of any model or system under investigation because the conditions for its validity are a) causality, b) linearity of response with respect to applied signal amplitude.

models assume that real permittivity at low frequency 'ε_0' and at high frequency 'ε_∞' can be determined and hence they provide empirical relations to describe permittivity's dependency on conductivity. For ideal dielectric materials, the response function is independent of non-interacting polarizing entities, and polarizability is time independent given as $\chi_r''(\omega)/\chi_r'(\omega) = \omega\tau$ (Debye Response) that is loss increase in proportional to frequency (the property of viscous fluids, see Fig. 1) [2].

However in real dielectric materials there are different types of polarizing responses under a steady electric field, such as,

  i.   with only dipolar species, a finite amount of charge displacement takes place.
  ii. with hoping electronic or ionic charges, an indefinite amount of charge may be displaced leading to the rising dispersion at low frequencies.

The real dipolar response is generally broader than the Debye shape is given by two fractional power laws (two exponents 'm' and 'n' in Fig. 1) flanking the loss peak at ω_p. Here the frequency independent ratio of the imaginary to real component is given as [3]

$$\frac{\chi_r''(\omega)}{\chi_r'(\omega)} = \cot\left(\frac{n\pi}{2}\right) \qquad (2).$$

and in a low frequency region for dielectric decrements, it can be represented as [3]

$$\frac{\chi_r''(\omega)}{\delta\chi_r'(\omega)} = \tan\left(\frac{m\pi}{2}\right) \qquad (3).$$
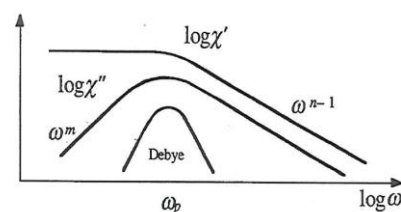

Figure 1. χ_r as a function of ω

1.2 Universal Dielectric Response (UDR)

Jonscher's proposed Universal Dielectric Response 'UDR' [4] in which he has defined conductivity as a function of excitation frequency with a power law characteristics. This power law characteristic is applicable to wide range of single crystals, polycrystalline and amorphous materials. Energy criterion model was further used to form a basis of power law characteristics for universal validity [2]. Furthermore, it is mentioned by Jonscher [3] that temperature is less important in Universal dipolar system 'UDS' than the Classical Dipolar System 'CDS' (Debye Systems) which may not be always true as in the case of dielectric mixture of atmospheric ice in the freezing domain temperature plays a dominant role. The strongly correlated system correspond to n → 1, and weakly correlated system corresponds to n→ 0, as proposed by Dissado et. al. [5].

1.1 Water Molecule As A Dielectric Material

A water molecule is a nonlinear polar molecule due to the electronegativity difference of around 1.2 between the constituent elements of H_2O. It has many dielectric mechanisms (atomic, electronic and dipolar) in different frequency domains associated with a cut off frequency in each domain which appears as a peak in $\varepsilon_r{''}=\varepsilon_r{''}(\omega)$ (likewise D=D(ω)) curve. It has strong dipolar effects at low frequencies particularly due to its orientation polarization which are also indicated in the experimental results of Fujino [6], Stiles et. al. [7], Kuroiwa [8] and Evans [9]. This dipolar orientation is generally associated with the relaxation[2] phenomenon, whereas the electronic and atomic polarizations are associated with the resonance phenomenon. In the frequency domain characteristics the relaxation frequency 'f_c is indicative of the relaxation time. For a detailed mathematical description of Debye Relations for atmospheric ice, see Mughal et. al. [10].

In ice relative to water, H-bonded networks are more extensive and as like water conductivity in ice molecule is due to proton hopping

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[2] Relaxation time 'τ_0' is a measure of the mobility of the dipoles that exist in the material to reorient themselves.

mechanism, which is termed as Grotthuss Mechanism. There are two stages of this mechanics, firstly the proton (H^+) 'hops' from the end of H-bonded chain to adjacent group till the next end and then in the second stage or the 'turn' stage, the molecule rotates and restores the original molecule before the first stage Markovitch et. al. [11].

The variation of conductivity for pure ice is given in Fig. 2, which clearly reflects that conductivity is dependent on frequency as well as temperature (a nonlinear dependency).
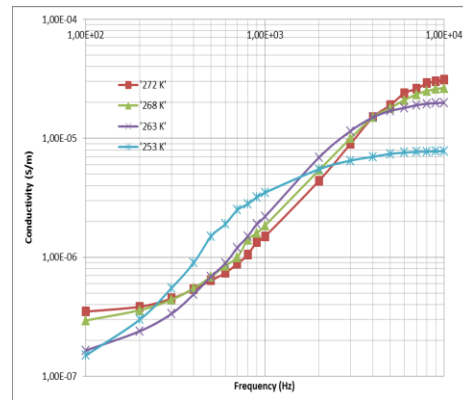

Figure 2. Conductivity variation with frequency and temperature for pure ice, Fujino [6].

2. Conductivity as a Function of Frequency

It has been known that all dielectric materials exhibit a frequency dependent electrical conductivity of the form Eq. (4) to some extent.

$$\sigma(\omega) = \sigma_0 + A(\omega^n) \qquad (4)$$

where 'σ_0' is the dc conductivity, 'A' is a linear constant and 'n' is same as for Eq. (2). Also it is explicitly shown by Panteny et. al. [12] through a resistor capacitor 'R-C' network (represent a microstructure that contains both dielectric and conductive regions as shown in Fig. 3 in which the disorder is on a small scale, whereby mobile oxygen vacancies are distributed in the insulating zirconia host) that the observed behavior is not equivalent to Debye behavior and the distribution of relaxation times For more details on R-C equivalent networks see [13, 14].
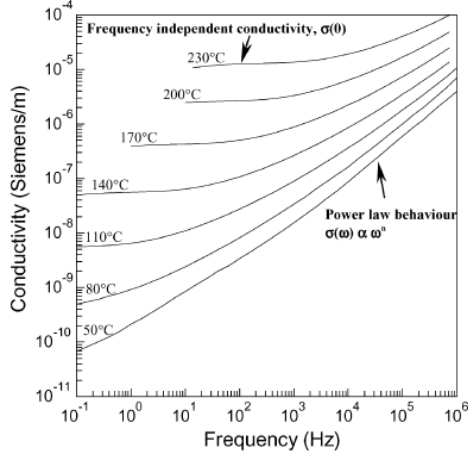
Figure 3. AC conductivity of doped zirconia at various temperatures

It is also observed from Fig. 3 that conductivity is just not the function of frequency but it also explicitly depends on temperature.

3. Conductivity as a Function of Frequency and Temperature:

Experiments of conductivity measurements reflect that pure ice has a static conductivity of order 10^{-7} S/m [15] which increases exponentially with rising temperature. Likewise semiconductors as like silicon or germanium, the conductivity of ice critically depends upon purity, e.g. ammonia or hydrogen fluoride largely changes its conductivity even as low as one part per million. Also ice is a 'protonic semi conductor' [16].

The conductivity of ice reaches a steady state value at frequencies higher than 10kHz. This high frequency conductivity 'σ_∞' varies exponentially with temperature [17],

$$\sigma_\infty = C_\infty e^{\left(\frac{-E_\infty}{kT}\right)}$$ (5)

where C_∞ is a constant, E_∞ is the activation energy at very high frequency and k is the Boltzmann Constant. For a more detailed overview of ice conductivity at high frequencies, see Paren [18].

Similarly the direct current conductivity or DC conductivity 'σ_s' is somewhat difficult to measure due to the inherent electrode polarization and surface conduction. However

surface conduction can be eliminated by protecting electrodes or probes with guard rings driven with unity gain amplifiers [19]. The bulk dc conductivity relation is given as,

$$\sigma_s = C_s e^{\left(\frac{-E_s}{kT}\right)}$$ (6)

where C_s is a constant & E_s is the activation energy at very small frequency or DC value. Few experimentally measured values for Eq. (5) & (6) are

Table 1. Experimental values of the parameters for conductivity relation

Parameter	Value	Reference
C_∞	1.6×10^6 S/m	[20]
E_∞	0.57 eV	
σ_∞ (@ 0^0C, 10kHz)	$(4.47\pm0.14) \times 10^{-5}$ S/m	[21]
E_∞ (@ 0^0C, 10kHz)	(0.56 ± 0.004) eV	
σ_s (@ 10^0C)	$(1.1\pm0.5) \times 10^{-8}$ S/m	[19]
E_s (@ T > 40^0C)	(0.34 ± 0.02) eV	

It is proposed that the σ_0 in Eq. (4) is not a constant but it is an exponential function of temperature, which in proton hoping case can be assumed of the form Eq. (6). Similarly the co-efficient A in Eq. (4) is also a function of the form Eq. (5) and Eq. (6). The frequency is non dimensionalized by ω_p (see Fig. 1) which in this case is equivalent to 10^4 Hz. The fractional power 'n' appeared from UDR and is a function of temperature which varies from zero to unity,

$$n = n(T) \qquad [0,1] \qquad (7)$$

4. Numerical Setup

The 'electrostatics' physics of COMSOL is used for this study as the frequency range to be dealt is relatively small $f \leq 10^6$ Hz. A numerical model (analytical model) (Fig. 4) equivalent with the experimental model (Fig. 5) was developed. A new material 'ice' with varying dielectric properties, dependent on temperature and relaxation time was introduced using the dielectric equations described in Mughal et. al. [22]. Second material used was 'air'. The relations for the variation in conductivity of atmospheric ice as a nonlinear function of frequency were used from the context of this paper (Eq. 4 to 7).
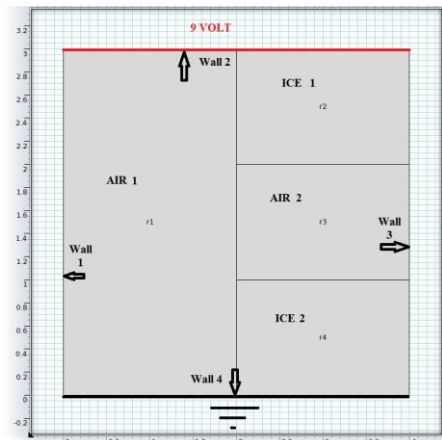
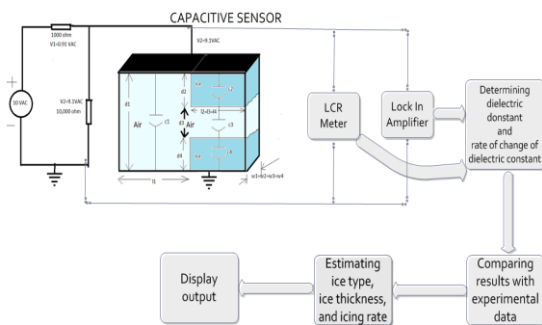Figure 4. Numerical Model of Capacitive Ice Sensor

Figure 5. Experimental Model of Capacitive Ice Sensor

The boundary conditions were defined such that Wall 1 is electrically insulated, Wall 2 is at 9 Volts, Wall 3 is electrically insulated and Wall 4 is at ground. Finer triangular meshing with total 1500 elements was used, Fig. 6. A frequency sweep from 1 Hz till 10^6 Hz, temperature sweep from -1^0C (272 K) till -25^0C (253 K) were used during the simulations.
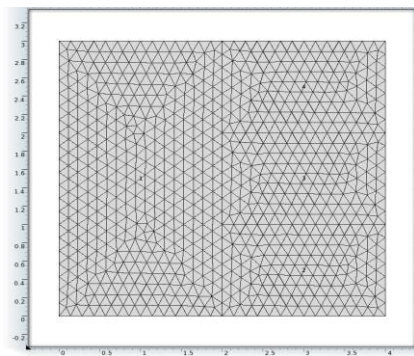

Figure 6. Meshing of the Model

4. Results

Using the Jonscher theory for Universal Dielectric Response and then utilizing Eq. 5 to Eq. 7 in it, the conductivity as a function of frequency and temperature is plotted in Fig. 8. The analytical results are then compared with experimental results of Fig. 2. From Fig. 9 to Fig. 12 the comparison between the two are shown explicitly. In Fig. 7 it is shown that ice 1 and ice 2 are holding potential of 9 Volts and 0 Volts. The conductivity values between analytical and experimental values at different frequency and temperatures are shown Tab. 2 'A'; where it is also shown that the variance between experimental and analytical model is quite low and similarly the R-squared values are very close to unity except at 253 K.

It is important to notice that in this paper an analytical model (proposed from the physics of ice itself) of conductivity is numerically solved using Comsol to compare with experimental results in order to validate the analytical model.
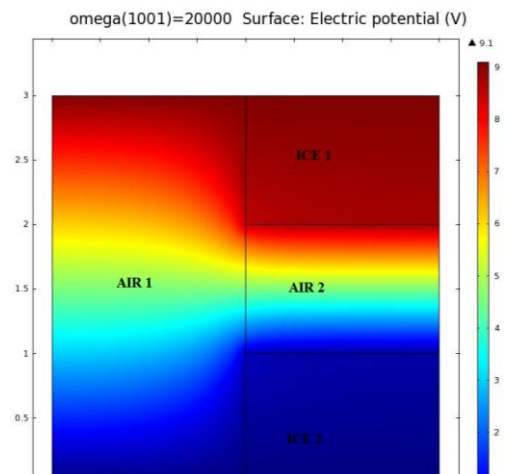

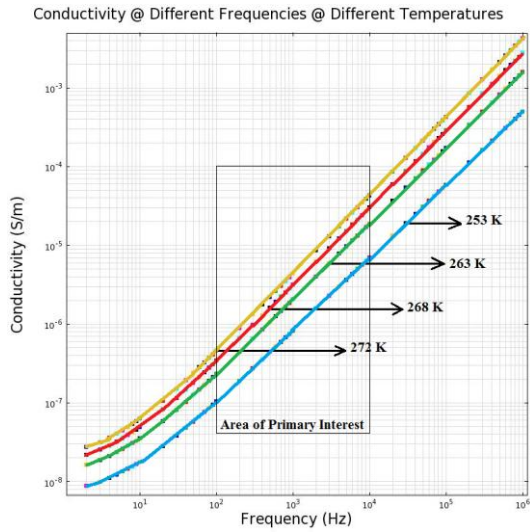Figure 7. Voltage Variation – Capacitive Icing Sensor

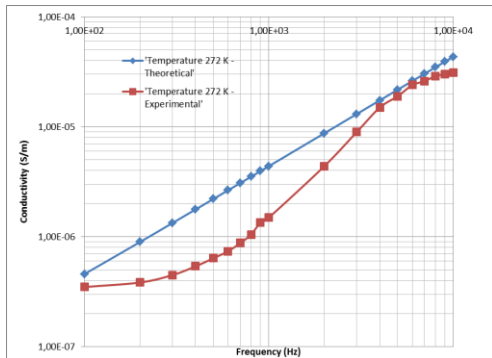Figure 8. Ice Conductivity as a function of frequency and temperature

Figure 9. Theoretical and Experimental Conductivities @ Temperature = 272 K
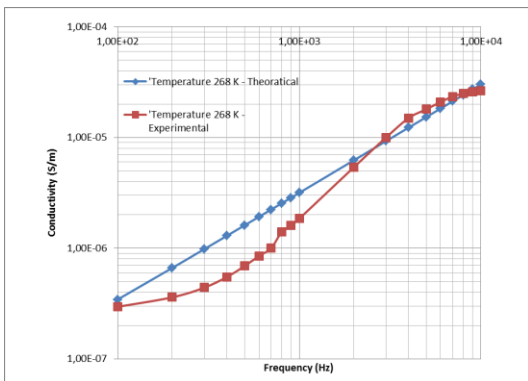

Figure 10. Theoretical and Experimental Conductivities @ Temperature = 268 K
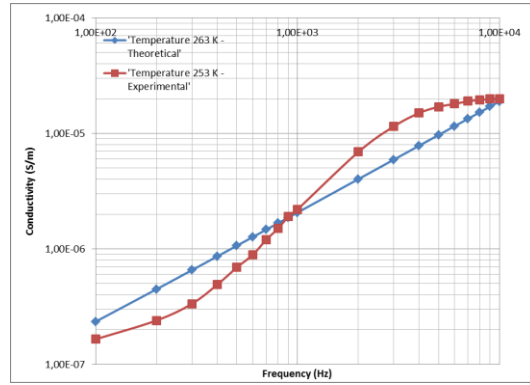

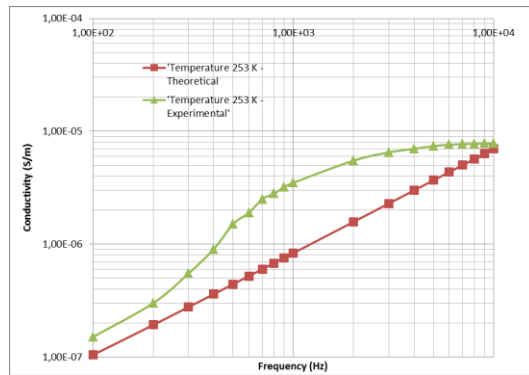Figure 11. Theoretical and Experimental Conductivities @ Temperature = 263 K

Figure 12. Theoretical and Experimental Conductivities @ Temperature = 253 K

Table 2. Conductivity Difference at Different Temperatures

ω	$\sigma_{Difference}$ ($\sigma_{Experimental} - \sigma_{Theoratical}$)			
	T = 272 K	**T = 268 K**	**T = 263 K**	**T = 253 K**
100	9,82E-06	9,10E-06	7,99E-06	4,23E-06
200	9,38E-06	8,78E-06	7,77E-06	4,14E-06
300	8,94E-06	8,46E-06	7,57E-06	4,06E-06
400	8,50E-06	8,15E-06	7,36E-06	3,97E-06
500	8,06E-06	7,83E-06	7,16E-06	3,89E-06
600	7,63E-06	7,52E-06	6,95E-06	3,81E-06
700	7,19E-06	7,21E-06	6,75E-06	3,73E-06
800	6,75E-06	6,90E-06	6,55E-06	3,65E-06
900	6,32E-06	6,59E-06	6,35E-06	3,58E-06
1000	5,88E-06	6,28E-06	6,16E-06	3,50E-06
2000	1,52E-06	3,21E-06	4,20E-06	2,75E-06
3000	2,82E-06	1,69E-07	2,29E-06	2,04E-06
4000	7,17E-06	2,85E-06	4,00E-07	1,34E-06
5000	1,15E-05	5,86E-06	1,47E-06	6,53E-07
6000	1,58E-05	8,86E-06	3,33E-06	2,28E-08
7000	2,02E-05	1,18E-05	5,18E-06	6,91E-07
8000	2,45E-05	1,48E-05	7,02E-06	1,35E-06
9000	2,88E-05	1,78E-05	8,85E-06	2,01E-06
10000	3,32E-05	2,08E-05	1,07E-05	2,66E-06
$Var(\sigma) = \frac{1}{n}\sum_{i=1}^{19}\left(\sigma_{i_{analytical}} - \sigma_{i_{experimental}}\right)^2$, $R^2 = 1 - \frac{\sum_{i=1}^{19}\left(\sigma_{i_{analytical}} - \sigma_{i_{experimental}}\right)^2}{\sum_{i=1}^{19}\left(\sigma_{i_{experimental}} - \sigma_{\mu_{experimental}}\right)^2}$				
Var(σ)	2,11E-10	9,67E-11	4,24E-11	9,29E-12
R^2	0,856	0,973	0,808	0,305

5. Conclusions and Discussions

UDR forms the basis of the conductivity variation with a fractional power of excitation frequency but it does not relate temperature with the power law. In this paper the Maxwell Boltzmann statistics is used for thermal excitation of proton jump for atmospheric ice and is used in the conductivity relation Eq. 4 which adequately supports the experimental results of Fujino [6]. Similarly the power exponent 'n' also varies from 0 to 1 and is also used as temperature dependent. At some temperatures the conductivity dependent on frequency and temperature shows more deviation e.g. 253 K (Fig. 12) which may be due to the nonlinear exponential interaction between the molecules but it's not clear. This study reflects that Universal Dielectric Response as proposed by Jonscher need some additional explanations of the assumed constants which in conductivity relation Eq. (4) are termed as 'σ_0', 'A' and 'n'. In this paper these all constants are found to be explicitly dependent on the temperature.

6. Future Work

Another relation for n is under consideration to improve the results however in this paper n is only treated as a linear function of temperature. Also the conductivity for snow is being considered to understand the heterogeneous interactions between ice, air and water.

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