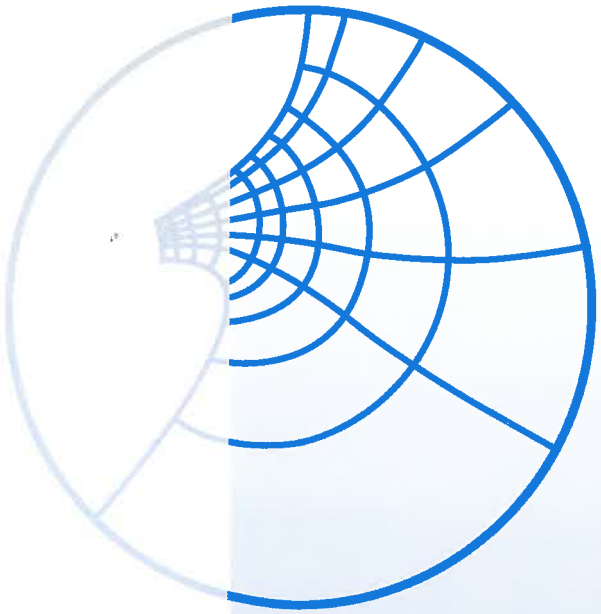


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The Numerical Relationship between Permittivity and Loss Tangent

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The Numerical Relationship between Permittivity and Loss Tangent

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1. Introduction: Checking our Dielectric Measurements

How can we have confidence that our dielectric measurements are delivering substantially correct results? There are at least two avenues open to us to check measurements: (i) measure a 'known' reference material to ascertain that our measurement equipment is operating correctly, (ii) intercompare results obtained by a number of different measurement methods. There is, however, a third method, which can be used to check that measurements are reasonably *self*-consistent whenever we measure permittivity and loss over a significant frequency range, say a decade of frequency or more. The formula given below relates the change in the real part of the relative permittivity, ε', over a range of frequencies to the mean loss tangent, $\tan \delta = \varepsilon''/\varepsilon'$, over that range, where ε'' is the imaginary part of the relative permittivity. The formula was given significant publicity as long ago as 1971 [Lynch, 1971], but it is still perhaps not as widely known as it should be.

2. The Kramers-Kronig Relations

Assume that we have a homogeneous dielectric specimen which is linear in its electrical response. In the *frequency domain* its response to an applied electrical signal is characterised by *two* parameters: $\varepsilon'(f)$ and $\varepsilon''(f)$, where f is the frequency in hertz. These are the two parts of the complex permittivity discussed in Section 1. Viewed in the *time* domain, however, the relationship between the voltage, V, applied across a capacitor formed from this dielectric and the current which can be obtained from it, I, will be given by a *single* function of time. The fact that both $\varepsilon'(f)$ and $\varepsilon''(f)$ are derived from a *single* real function of a *single* real parameter (time) implies that their values must be closely related to each other. They are in fact related by the well-known Kramers-Kronig relations [Daniel, 1967, James and Andrasic, 1990], see also equations (1) and (2) of Lynch [1971] and Section 6 below. The significance of these relations is that if $\varepsilon'(f)$ is known over a sufficiently wide frequency range, $\varepsilon''(f)$ can be computed directly from it. Likewise, if $\varepsilon''(f)$ is known over a sufficiently wide frequency range, along with ε_∞, the high frequency real-permittivity limit, $\varepsilon'(f)$ can be computed directly from it. The quantities $(\varepsilon'(f) - \varepsilon_\infty)$ and $\varepsilon''(f)$, in fact, form a *'Hilbert Transform Pair'* [James and Andrasic, 1990]. So, if we have measured the values of both $\varepsilon'(f)$ and $\varepsilon''(f)$, these quantities can be used to check each other: that is to say that the internal *consistency* of our measurements can be checked. The mathematically inclined can use the full Kramers-Kronig relationships in exactly this way and this can be recommended when we have a considerable body of data available to us over a wide frequency range, see for example Jonscher [1983], James and Andrasic [1990]. But for a cost-effective check upon measurements over a limited

frequency band, the full Kramers-Kronig relationships are rather inconvenient to use because they are normally expressed as integral equations over the frequency range 0 Hz to ∞ Hz (or from time $\tau = 0$ to time $\tau = \infty$ in the time domain). In short, they are unnecessarily complicated to apply as a simple check of consistency.

3. The simplified equation.

In 1971 Arnold Lynch derived and published in an accessible form [Lynch, 1971] a simplified equation for relating changes in ε', $\Delta\varepsilon'$, to tan δ when the response of the material is characterised by dielectric relaxation behaviour. The formula is:

$$\Delta\varepsilon' / \varepsilon' = m \tan\delta \log_{10}(f_2 / f_1) \qquad \ldots (1)$$

Here $\Delta\varepsilon' = (\varepsilon'_1 - \varepsilon'_2)$, where the two permittivities are measured at frequencies f_1 and f_2 respectively. If we make $f_1 < f_2$, we will have $\varepsilon'_1 > \varepsilon'_2$, and with the definition just given, $\Delta\varepsilon'$ will be a positive quantity representing the *fall* in ε' between f_1 and f_2. Note that dielectric relaxation always results in a fall in ε' with frequency. The value of the numerical parameter m in Lynch's derivation depends upon assumptions made about the nature and frequency spread of the relaxation processes which are giving rise to the loss, tan δ, and to the changes in ε'. Typically, $1.0 \leq m \leq 2.3$, and as Lynch noted, setting $m \approx 1.5$ provides an equation which holds within a factor of 1.5 for most practical cases.

Expressions of this general sort were known to the dielectrics community long before 1971, see Garton [1946] and Whitehead [1946]. In 1946 formulas similar to the following were being discussed:

$$\Delta\varepsilon' / \varepsilon' = (2/\pi) \tan\delta \ln(f_2 / f_1) \qquad \ldots (2)$$

where conversion of the natural logarithm to base10 again gives equation (1) with $m \approx 1.5$. This formula would, in fact, be exact for materials in which tan δ was independent of frequency. Lynch's contribution in 1971 was to suggest that the equation could be used as an approximation even when tan δ was not constant and he used existing data to show how far this was possible.

4. How to use the simplified equation for checking dielectric measurements.

The approximate equation applies to homogeneous linear dielectrics over frequency ranges in which tan δ can reasonably be construed to have a meaningful 'average' value. Reference to Lynch's paper [1971] should be made for an idea of what 'reasonable' might mean in this context. Changes by a factor of 10 in tan δ from f_1 to f_2 will clearly not give a well-defined 'average' from which an accurate check of $\Delta\varepsilon'$ can be derived. However, tan δ usually changes very slowly with frequency in typical dielectric relaxations and a change by a factor of 2 from f_1 to f_2 will generally give agreement for $\Delta\varepsilon'$ within Lynch's allowance factor of 1.5, provided tan δ is taken at a median 'centre' frequency between f_1 and f_2 measured on a logarithmic scale of frequency.

Example 1: This example is taken from a set of measurements in an NPL admittance cell. The measurements were upon a medium loss polymer disc specimen. Here are the results:

Frequency	Measured Real Permittivity ε'	Measured Loss Tangent $\tan\delta$
10 kHz	2.85 ± 0.03	0.076 ± 0.08
300 kHz	2.48 ± 0.03	0.054 ± 0.05
10 MHz	2.25 ± 0.03	0.031 ± 0.03

Between 10 kHz and 10 MHz, 300 kHz is the 'central' frequency on a logarithmic scale. Using equation (1) with $m = 1.5$, $f_1 = 10$ kHz, $f_2 = 10$ MHz, we have $\log_{10}(f_2/f_1) = 3$. The 'central' value of ε' is 2.48 and that of tan δ is 0.054 and so we calculate that the fall in ε' from 10 kHz to 10 MHz should be:

$$\Delta\varepsilon' \approx 1.5 \times 3 \times 0.054 \times 2.48 = 0.60$$

In fact the *measured* change in ε' is also 0.60. This level of agreement must be regarded as somewhat fortuitous when we consider that tan δ changes by a factor of 0.076/0.031 = 2.45 over our measured frequency range. As stated above, an agreement to within a factor of 1.5 may be regarded as acceptable.

For the check to be valid, the uncertainties in $\Delta\varepsilon'$ have to be small compared to the value of $\Delta\varepsilon'$ itself. In the above example, it would not be possible to perform the check if the uncertainty in $\Delta\varepsilon'$ was over ± 0.2, say. In fact, the uncertainty in $\Delta\varepsilon'$ was certainly less than ± 0.05 (coverage factor 2), so the check is a valid one. In the circumstances, had we measured $\Delta\varepsilon'$ to be much larger, eg. over 1.0, or much smaller, eg. less than 0.3, say, we could have had good reason to doubt our measurements. Likewise if, with $\Delta\varepsilon' = 0.60$ (as measured), we had measured tan δ values as low as 0.02 or as large as 0.1 we should be equally suspicious of our results: they would not be consistent.

The formula clearly cannot be used if the loss tangent, tan δ, lies below the resolution of the measurement equipment but it may provide a useful means of checking just what the resolution is in cases where it is not previously known. Here is another example which uses a lower loss material:

Example 2:

Frequency	Measured Real Permittivity ε'	Measured Loss Tangent $\tan\delta$
10 kHz	3.085 ± 0.04	0.0074 ± 0.0008
300 kHz	3.025 ± 0.04	0.0097 ± 0.0010
10 MHz	2.965 ± 0.04	0.0075 ± 0.0008

This material exhibits a broad relaxation which, in fact, reaches its loss peak near 300 kHz. With $m = 1.5$, $f_1 = 10$ kHz, $f_2 = 10$ MHz and $\log_{10}(f_2/f_1) = 3$ we have from equation (1):

$$\Delta\varepsilon' \approx 1.5 \times 3 \times 0.0097 \times 3.025 = 0.132$$

As we can see, the actual value of $\Delta\varepsilon'$ is 0.120, so the agreement is well within Lynch's region of tolerance. Indeed one could argue that the 'mean' value of tan δ between 10 kHz and 10 MHz is going to be somewhat less than 0.0097, which is close to its peak value. Using a slightly lower value for tan δ could give an even better agreement. On the other hand, it might be thought that as the uncertainty in ε' is $\pm$ 0.04, then the uncertainty in $\Delta\varepsilon'$ must be approx. $\pm$ 0.07, so the test should be regarded as only just sensitive enough for this material. However, this need not be the case: $\pm$ 0.04 was the *total* estimated uncertainty for each ε' value. This total uncertainty takes account of errors of measurement (like the effects of fringing fields) which can give wrong absolute values for ε' but which are common at all measurement frequencies. As all of the data at all frequencies were obtained in one measurement run without removing the specimen from the cell, the estimated uncertainty in $\Delta\varepsilon'$ is actually much smaller than the absolute uncertainty in ε'. In fact, in this case we estimate an uncertainty of as low as $\pm$ 0.005 for the measured $\Delta\varepsilon'$ and we can again conclude that the above test is valid. It is not uncommon in dielectric measurements for uncertainties relating to *changes* in ε' to be much less than uncertainties in ε' itself: ie. measurement *precision* is commonly much better than *accuracy* in these measurements. This is a consequence of the fact that the same measurement instrument is being used throughout and its systematic errors change slowly with frequency.

5. When the formula cannot be applied.

As discussed above, Lynch showed that equation (1) is reasonably accurate where the dielectric response is dominated by relaxation behaviour, ie. where tan δ is varying slowly with frequency. One should not expect it to hold at much higher frequencies (eg. infra-red) where sharp dielectric resonances, rather than relaxations, occur. Care in its application must also be taken with *conducting* materials: materials with free charge carriers. Consider such a material with conductivity σ, whose behaviour, for all practical purposes in the frequency range of interest, can be described by a equivalent circuit consisting of a fixed resistor in parallel with a fixed lossless capacitor. We assume that we do not know what the properties of this material are (that is why we are measuring it!). Working at radio or microwave frequencies we will not in the first instance (without additional information) be able to distinguish between *true* dielectric loss, caused by a relaxation (which, in fact, is absent in this case) and the *apparent* dielectric loss caused by the effects of conduction. We will obtain an *equivalent* loss tangent, $\tan \delta_e = \sigma / 2\pi f \varepsilon' \varepsilon_0$, where ε_0 is the permittivity of free space, falling with frequency. So if equation (1) is applied, there is an apparently *finite* value for tan δ to insert into equation (1), giving rise to a *finite* predicted $\Delta\varepsilon'$. But in fact $\Delta\varepsilon' = 0$ for such a material. Thus, unless additional information is available which allows one to distinguish conductivity losses from relaxation losses, or unless it is known that the conductivity loss, though finite, is small compared to the relaxation losses, equation (1) cannot be expected to hold in materials known to have free charge carriers.

Care should also be taken with anisotropic specimens. If the specimen is measured with the electric field in different directions at f_1 and f_2, a spurious change in ε' may be attributed to the specimen. It may be thought that this is caused by the change in frequency, but in fact it would have been caused by the change in the field orientation in the specimen. Use of the formula with ferroelectrics should also, in general, be ruled out (see comments in the next section).

6. Derivation of the formula.

This section is included for the mathematically inclined: all of the practical aspects of the application of equation (1) have already been dealt with in sections (1) - (5). The paper by James and Andrasic [1990] covers similar considerations from a more general viewpoint.

For derivation of equation (1), the form of the Kramers-Kronig relations which we require is written

$$\varepsilon'(f) - \varepsilon_\infty = \frac{2}{\pi} \int_{u=0}^{f_m} \frac{u\,\varepsilon''(u)}{u^2 - f^2}\, du \qquad \ldots (3)$$

where u is a dummy variable for frequency, to be integrated up to f_m which is the frequency at which ε_∞ is specified. It can be seen that the integrand is significant only for values of u of the same order as f, the frequency at which ε' is to be derived. When $u << f$ the factor multiplying $\varepsilon'' \rightarrow u/f^2$, and when $u >> f$ the factor $\rightarrow 1/u$. The approximation we now make is that, in the vicinity of frequency f, ε'' can be replaced by its mean value $\overline{\varepsilon''}$ over this range. It can then be taken outside the integral

$$\varepsilon'(f) - \varepsilon_\infty = \frac{2\,\overline{\varepsilon''}}{\pi} \int_{u=0}^{f_m} \frac{u}{u^2 - f^2}\, du \qquad \ldots (4)$$

which can be integrated directly, since $2u\,du = du^2$,

$$\varepsilon'(f) - \varepsilon_\infty = \frac{\overline{\varepsilon''}}{\pi} \left[\ln\left(u^2 - f^2\right) \right]_{u=0}^{f_m}$$

$$= \frac{\overline{\varepsilon''}}{\pi} \left[\ln\left(f_m{}^2 - f^2\right) - \ln f^2 - i\pi \right]$$

$$\varepsilon'(f_1) - \varepsilon'(f_2) \approx \frac{2\,\overline{\varepsilon''}}{\pi} \ln\left(\frac{f_2}{f_1}\right) \qquad \ldots (5)$$

This is the formula required, so we should recap the conditions and approximations required to reach it.

(i) The Kramers-Kronig (K-K) relations relate to the response of a linear causal system, when expressed in the frequency domain. Causality need never trouble us, but linearity must be considered. In the case of dielectrics we have assumed that the polarisation (the system response) is directly proportional to the applied electric field (the input). Thus ferroelectrics are ruled out. Many more materials are ruled out if the K-K relations are applied to the magnetic parameters.

(ii) It may be noted that $(\varepsilon - 1)$, not ε, is the true transfer response since ε contains a component of the applied field. However this is taken care of in eq. (3) by writing $(\varepsilon - \varepsilon_\infty)$.

(iii) Consideration of the transfer response also shows why the effect of free charge carriers, giving rise to dc conductivity, must be ruled out: because the application of a steady E-field does not lead to a steady polarization; charge movement continues as $t \rightarrow \infty$. A little more algebraic manipulation would show that the effect on $\varepsilon'' = \sigma/\omega\varepsilon_o$ is such as to make no contribution to the integral in eq (3).

(iv) Having accepted the conditions for K-K, the main approximation required is that ε'' should be slowly varying in the vicinity of the frequency at which ε' is to be found. A factor of 10 either way in frequency would be very generous and the practical examples given earlier should give some feel for this.

(v) The approximation involved in eq 3. is readily quantified. We have neglected the term $\ln\left(f_m^{\,2} - f_1^{\,2}\right)\Big/\left(f_m^{\,2} - f_2^{\,2}\right)$ so we require $f_i^{\,2} << f_m^{\,2}$ which is easily met.

7. References

V V Daniel, 1967, *'Dielectric Relaxation'*, (Academic Press, London), pp 75-76.

C G Garton, 1946, *'The distribution of relaxation times in dielectrics', Trans. Faraday Soc.* **XLIIA**, pp 56-60; also C G Garton, 1946, *'The charactersitics and errors of capacitors used for measurement purposes', J IEE*, **93**, Pt. II, pp 398-408.

J R James and G Andrasic, 1990, *'Assessing the accuracy of wideband electrical data using Hilbert transforms'*, *Proc IEE*, **137**, Pt. H, pp 184 - 188.

A K Jonscher, 1983, *'Dielectric Relaxation in Solids'*, (Chelsea Dielectrics Press, London), pp 47 - 61.

A C Lynch, 1971, *'Relationship between permittivity and loss tangent', Proc. IEE*, **118**, pp 244 - 246.

S Whitehead, 1946, *'A note on the analysis of dielectric measurements, Trans. Faraday Soc.* **XLIIA**, pp 66 - 75; see also Garton reference above and the General Discussion, *Trans. Faraday Soc.* **XLIIA**, pp 75 - 78. This comes close to equation (2) but fails to take the final step.