

A PERCOLATION MODEL FOR ELECTRICAL CONDUCTION IN WOOD WITH IMPLICATIONS FOR WOOD–WATER RELATIONS

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(Received May 2008)

Abstract. The first models used to describe electrical conduction in cellulosic materials involved conduction pathways through free water. These models were abandoned in the middle of the 20th century. This article re-evaluates the theory of conduction in wood by using a percolation model that describes electrical conduction in terms of overlapping paths of loosely bound or capillary water (Type II water). The model contains two physical parameters: w_c , the critical moisture fraction, which is the amount of water required to form a continuous path of Type II water in wood; and σ_c , the conductivity of the aqueous pathways. The model gives a good fit to previously published data of the DC conductivity of wood when w_c is equal to 16% moisture content and σ_c is equal to 0.88 S m^{-1} . This analysis indicates that electrical conduction in wood can be explained by percolation theory and that there exists a continuous path of Type II water in wood at w_c , which is below the traditional fiber saturation point.

Keywords: Ionic conduction, conductivity, percolation theory, wood–water relations.

PREVIOUS MODELS OF CONDUCTION IN WOOD

Modern theories of electrical conduction in wood can be traced to Hearle's (1953) work on the electrical properties of textiles. Hearle reviewed two models that related the conductivity of textiles to their moisture content. Both models assumed that textiles were ionic conductors whose conductivity, σ (S m^{-1}), could be calculated by the traditional formula:

$$\sigma = nz\mu \quad (1)$$

where n is the number density of charge carriers (m^{-3}), z their charge (C), and μ their mobility ($\text{m}^2 \text{ s}^{-1} \text{ V}^{-1}$).

The first model of conduction in cellulosic materials states that conduction occurs along water pathways interrupted by breaks. This model was first proposed by Murphy and Walker (1928) and later O'Sullivan (1948). In this model, only the mobility changes with moisture content. The mobility was calculated from the velocity of ions in water paths, the number of breaks per path, and the average time that ions stop at breaks. At higher moisture contents, there are fewer breaks in water paths and therefore the conductivity is higher. This model has a clear physical picture that is similar to the classical (Drude 1900) model for electrical conduction in conducting solids in which conductivity is a function of the number of electrons and their mean free path. However, because the paths of water cannot be

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measured directly, implementing this model to fit conductivity data requires many assumptions. Additionally, Hearle found that this model did not fit the data as well as other models. Because of these disadvantages, Hearle rejected the Murphy, Walker, and O'Sullivan model in favor of a second model involving the number of charge carriers.

This second model describes the increase in conductivity with moisture content in terms of an increase in the number density of charge carriers (n). The increase in n results from more ions dissociating as moisture content is increased. Using an Arrhenius relation, Hearle related the degree of dissociation to an activation energy, which is inversely related to the dielectric constant at a given frequency, $\epsilon'(\omega)$. In general, $\epsilon'(\omega)$ increases with increasing moisture content, which results in a lower activation energy, more ions dissociated, more charge carriers, and therefore higher conductivity.

The resistivity ($\rho \equiv 1/\sigma$) is given by:

$$\log(\rho) = \alpha + \beta/\epsilon'(\omega) \quad (2)$$

where α and β are fit parameters related to the mobility and activation energy, respectively. Using Brown's (1962) relationship for the dielectric constant of wood at 2 MHz as a function of percentage moisture content (MC; see Eq 6),

$$\epsilon'(2 \text{ MHz}) = 3.94(10^{0.0242 \cdot \text{MC}}) \quad (3)$$

we can fit Hearle's model to Stamm's (1929, 1964) conductivity data of slash pine (*Pinus elliotii*) as a function of moisture content. The model clearly fits the data well (Fig 1). Stamm's data were chosen to test these models because they are the most complete conductivity data on any species in the literature with the most data points and the broadest range of moisture content. Recent measurements have confirmed the accuracy of these data (Zelinka et al in press).

Although Hearle's model fits the data extremely well, the predictive power of the model comes from the relationship between $\epsilon'(\omega)$ and moisture content. Traditionally, the relationship between $\epsilon'(\omega)$ and moisture content has come from

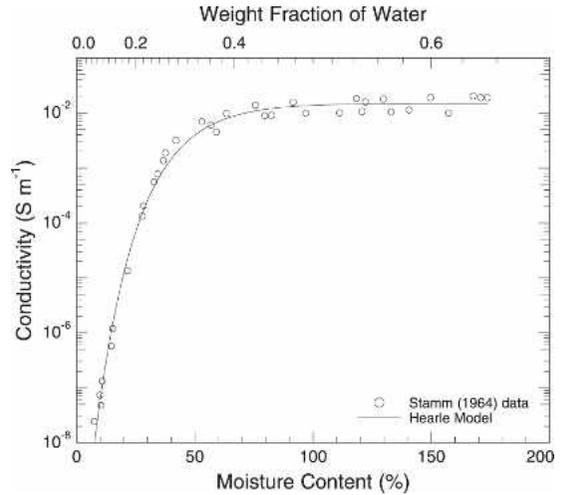


Figure 1. Hearle model fit of Stamm's data of the conductivity of slash pine as a function of moisture content.

empirical data (Hearle 1953; Brown 1962; Vermaas 1974). Because empirical data were used to fit the resistivity data, it is not possible to describe the changes in resistivity with moisture content in terms of material parameters. More importantly, the Hearle model is based on a relationship between $\epsilon'(\omega)$ and ρ derived from Debye-Hückel theory. However, this relationship is in fact circular because $\epsilon'(\omega)$ and ρ are not independent (see Appendix A).

In short, although Hearle's model fits the data, this fit is the result of the purely mathematical relationship between $\epsilon'(\omega)$ and ρ . This model lacks a physical mechanism of ionic conduction and a description of wood-water relations.

In this article, we propose a new model for electrical conduction in wood based on percolation theory. The advantages of this model are summarized in Table 1. The key feature of this model is a percolation threshold, the minimum moisture content needed for ionic conduction in solid wood. We then relate the percolation threshold to recent experiments, which suggest that two distinct types of water exist below fiber saturation, as well as physical properties of wood that exhibit threshold-like or discontinuous behavior with moisture content in wood such as corrosion (Dennis et al 1995), longitu-

Table 1. Comparison of models for electrical conduction.

	Murphy and Walker (1928), O'Sullivan (1948)	Hearle (1953)	Zelinka et al (this work)
Good fit to data		✓	✓
Requires $\epsilon'(\omega)$ data		✓	
Physical picture consistent with wood–water relations	✓		✓
Threshold for ionic conduction			✓
Relation to Type II water			✓

dinal shrinkage (Ying et al 1994; Kretschmann and Cramer 2007), and mold growth (Viitanen 1996; Viitanen and Ojanen 2007).

PERCOLATION MODEL FOR CONDUCTION IN WOOD

Percolation theory is a branch of mathematics that deals with the connectivity of randomly distributed elements. Although percolation models are used to describe widely different phenomena, these models all contain a percolation threshold, that is, a fraction of sites that need to be occupied before an infinite, continuous network exists. Percolation models have been used to describe electrical conduction in two-phase composites in which one phase is conducting and the other is insulating.

If a binary composite contains a volume fraction p of a conductive component (with a purely real conductivity σ_c) and the remainder of a perfectly insulating component ($\text{Re}(\sigma_i) = 0$),[‡] then it has been shown (Clerc et al 1990; Stauffer and Aharony 1992; Nan 1993) that the conductivity of the composite, σ , near the percolation threshold can be described by:

$$\sigma = \begin{cases} 0 & p < p_c \\ \sigma_o(p - p_c)^t & p \geq p_c \end{cases} \quad (4)$$

where p_c is the percolation threshold, that is, the volume fraction at which there is a continuous path of the conducting phase through the composite. The constant of proportionality, σ_o , can be related to the conductivity of the conducting

phase, σ_c , through (McLachlan and Heaney 1999):

$$\sigma_c = \sigma_o(1 - p_c)^t. \quad (5)$$

Finally, t is called the critical exponent and is related to the fractal dimension of the conducting path.

In the literature, percolation models of conduction have used volume fraction to describe the percolation threshold because there is a clear connection between the critical volume fraction and the geometry of a continuous conducting path. In fact, it is possible to calculate p_c from computer simulations if the size and aspect ratio of the particles in the conducting phase are known. However, these models were developed for composites in which the randomly distributed phases do not change volume when mixed. Wood, in contrast, interacts with water in a complicated manner; sorption of water within the cell walls causes volumetric expansion. Also, water is not randomly distributed throughout the entire wood volume because of lumina in the cell wall structure and crystalline regions in the cell wall itself. Therefore, we present a percolation model based on weight partitioning of the phases.

Conductivity data have traditionally been plotted in terms of percentage moisture content given by:

$$\text{MC} = \left(\frac{m_{\text{water}}}{m_{\text{wood}}} \right) \cdot 100\% \quad (6)$$

where m_{water} is the mass of water in the sample at a given moisture content and m_{wood} is the mass of the oven-dry wood. This conventional partitioning into two components is suggestive of a percolation model, in which conduction occurs through the “water phase” but not through

[‡] This model can also be used if $\text{Re}(\sigma_i) \neq 0$ so long as $\text{Re}(\sigma_c) \gg \text{Re}(\sigma_i)$ (McLachlan and Heaney 1999).

the insulating “wood phase.” However, moisture content is not the ideal partitioning variable because the percolation model requires this variable range from 0 to 1 and moisture contents over 100% are possible when $m_{water} > m_{wood}$. For the percolation model, it is necessary to describe the amount of water in wood as the weight fraction of water, w :

$$w = \left(\frac{m_{water}}{m_{wood} + m_{water}} \right) \tag{7}$$

which can be calculated from moisture content by:

$$w = \frac{MC}{MC + 100}. \tag{8}$$

Because the percolation threshold in wood cannot be calculated from first principles for reasons discussed previously, it must be experimentally determined. The threshold for ionic conduction in wood was measured as long as 40 yr ago by Lin (1965), although he did not analyze the data in terms of a percolation model. Lin measured the response of radioactive sodium and iodide ions to an applied electric field in western white pine (*Pinus monticola*) and found that below 16% MC ($w = 0.14$), there was no net movement of the ions, but above 16% MC, the ions moved in response to the electric field. The threshold remained at 16% MC even when the magnitude of the electric field was increased. Furthermore, using neutron activation analysis, Langwig and Meyer (1973) confirmed long-range migration of sodium, potassium, chloride, and bromide between 15.7% and 18.0% MC in three tropical wood species under an applied electrical field. Using impedance spectroscopy, Zelinka et al (in press) found that at 20% MC, the spectra of southern pine exhibited Warburg ($\omega^{-0.5}$) behavior indicative of long-range migration of charge carriers, which was not observed at 12% MC (Zelinka et al 2007). These data collectively suggest that 16% MC is the percolation threshold for ionic conduction in wood.

We can now fit the historical conductivity (Stamm 1964) data to the modified percolation model:

$$\sigma = \begin{cases} 0 & w < w_c \\ \sigma'_o(w - w_c)^z & w \geq w_c \end{cases} \tag{9}$$

where volume fraction is replaced with weight fraction, σ_c can be calculated from

$$\sigma_c = \sigma'_o(1 - w_c)^z \tag{10}$$

and z is still a critical exponent that describes the same physical phenomenon as t but has a different numeric value. We took w_c as 0.14 from Lin’s data (1965). From Stamm’s data (Fig 1), it appears that there is a nonzero electrical conductivity below the percolation threshold. We believe this is the result of a mechanism other than ionic conduction with different charge carriers such as protons (Christie et al 2004). This mechanism may continue to contribute to the conductivity at higher moisture contents but is quickly overshadowed by the rapid increase in conductivity after the percolation threshold is reached.

Figure 2 shows the linear least squares fit of the percolation model to the historical (Stamm 1964) conductivity data, in which the axes are transformed to make the data linear. The percolation model describes the behavior near the percolation threshold. It does not describe data far above the threshold, in which the conductivity remains constant with increasing moisture con-

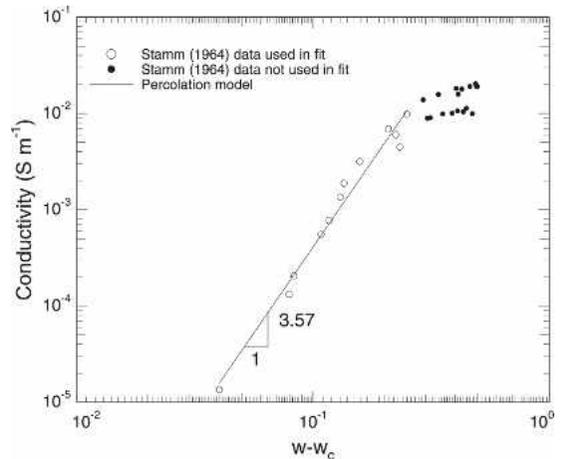


Figure 2. Results of the percolation model fit: $w_c = 0.14$, $\sigma'_o = 1.52 \text{ S m}^{-1}$, and $z = 3.57$.

tent, and therefore data above 63% MC were not included in this fit but are still shown in Fig 2 for reference.

Both the percolation model and the Hearle model appear to fit the data well as shown in Fig 3. To quantify the goodness of fit, the R^2 statistic (Kvålseth 1985) was calculated by:

$$R^2 = 1 - \frac{\sum_i (y_i - \hat{y}_i)^2}{\sum_i (y_i - \bar{y})^2} \quad (11)$$

where we took y_i as the logarithm of the measured conductivity, \hat{y}_i as the logarithm of the predicted conductivity, and \bar{y} as the arithmetic mean of all y_i between the percolation threshold and 63% MC. The R^2 from the Hearle model fit was 0.964 and the R^2 from the percolation model fit was 0.975. Both models fit the data well.

Further support for the hypothesis that conduction in wood is a percolation phenomenon comes from impedance spectroscopy measurements above and below the percolation threshold (Zelinka et al 2007, in press). The impedance spectra exhibited a time constant whose frequency dependence was ω^{-n} in which $0 < n < 1$. Zelinka et al fit this ω^{-n} behavior with a constant phase element, which has a distribution of relax-

ation times with the exponent n being related to the width of the distribution. In percolating systems, there is a distribution of relaxation times as a result of the stochastic nature of the conducting path. It has been shown that the AC behavior of percolating systems also exhibits ω^{-n} ($0 < n < 1$) behavior (Raistrick 1987; McLachlan and Heaney 1999; McLachlan et al 2005), and AC percolation data can be fit with a constant phase element (Liu 1985; Tomkiewicz and Aurian-Blajeni 1988). Christie et al (2004) observed ω^{-n} behavior in cellophane and suggested this behavior may be the result of a percolation cluster phenomena but did not go so far as to fit the data to a percolation model. Therefore, we believe this observed ω^{-n} behavior in southern pine (Zelinka et al 2007, in press) is also suggestive of a percolation mechanism for conduction in wood.

IMPLICATIONS OF THE PERCOLATION MODEL

Physical Interpretation of σ_c and z

From the linear least squares fit, the value of σ'_o was 1.52 S m^{-1} , and with w_c equal to 0.14, the linear least squares fit (Fig 2) results in a σ_c of 0.88 S m^{-1} . Because σ_c is the conductivity of the conducting phase, which in our model is water with ions, we should be able to relate σ_c to the concentration of mineral ions in wood using a relationship between conductivity and salinity. McMillin (1970) measured the concentration of mineral ions in the sapwood and heartwood of loblolly pine (*Pinus taeda*) and found the average concentration of mineral ions in oven-dry wood to be 1.8 parts per thousand by mass (ppt). Using the method provided in *Standard Methods for the Examination of Water and Wastewater* (Anon 1999), the conductivity of water with a salinity of 1.8 ppt is 0.28 S m^{-1} , lower than our measured σ_c of 0.88 S m^{-1} but within an order of magnitude.

It is interesting to note that our value of σ_c , 0.88 S m^{-1} , is greater than the asymptotic limit of the conductivity with moisture content, which from Fig 1 is approximately 0.02 S m^{-1} . This is not surprising because the maximum moisture con-

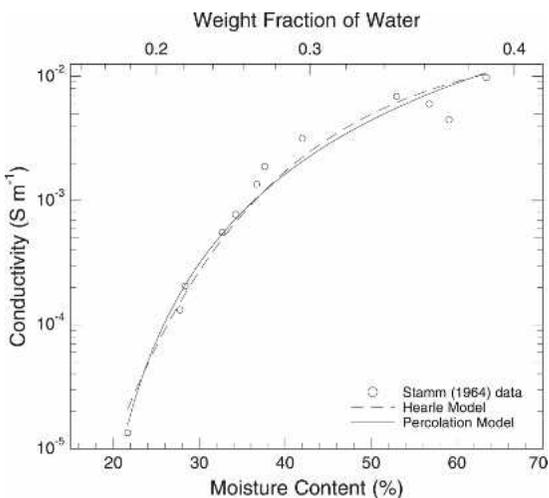


Figure 3. Comparison of the Hearle model fit and the percolation model fit.

tent of southern pines is achieved at weight fraction of only approximately 2/3 water (Forest Products Laboratory 1999). It is likely that this asymptote represents the conductivity limit of water in wood and not the conductivity of the water phase by itself.

Implicit in this model is the assumption that σ_c is independent of moisture content. However, it is reasonable that σ_c may change with moisture content because the concentration of ions and the diffusivity of sorbed water depend on moisture content. Assuming 100% dissociation above the percolation threshold, the ion concentration depends on moisture content. Using the bounds of the percolation model, at 16% MC, the effective salinity in the water phase is 11 ppt, whereas at 63% MC, it would be 3 ppt, which results in conductivities of 1.5 S m^{-1} and 0.4 S m^{-1} , respectively. These conductivities bracket our measured σ_c of 0.88 S m^{-1} . The mobility of sorbed water, as reflected in the diffusion coefficient, is a weak function of moisture content increasing less than a factor of ten between 16% and 28% MC (Stamm 1959). These two processes have opposite effects on σ_c and some cancellation of those effects can be expected.

For 3-D percolation models that use a volume fraction, the critical exponent t is approximately 2, and this value has been confirmed both theoretically and experimentally (Clerc et al 1990; Stauffer and Aharony 1992; Nan 1993; Garboczi et al 1995). The critical exponent z in our model could be related to t if we knew the exact relationship between weight and volume fractions. Roughly, ignoring the change in volume with moisture content, and estimating the specific gravity of wood as 0.5 and that of water as 1, the volume percolation model (Eq 4) gives an exponent, t , of 2.3, close to the expected value of 2.

Physical Interpretation of w_c

The most important parameter of this percolation model in terms of a physical picture of electrical conduction in wood is the percolation threshold, w_c , the moisture fraction at which there is a continuous path of water in which

ionic conduction occurs, which we refer to hereafter as “conducting water.” Using the percolation model and salinity/conductivity data for water, we found reasonable agreement between the conductivity of bulk water and conducting water in wood. The model has $w_c = 0.14$ (16% MC), which is far below the traditional fiber saturation point of nominally 30% MC (Forest Products Laboratory 1999), in which the cell walls are completely saturated and additional water begins to fill the lumina. It is also unlikely that truly bound water would be able to solvate the ions necessary for conduction because the water would not be able to orient its dipole along the electric field if it were hydrogen-bonded to the cell wall. However, other processes in wood that require mobile water such as corrosion (Dennis et al 1995) and mold growth (Viitanen 1996; Viitanen and Ojanen 2007), also begin to occur between 15% and 20% MC. Additionally, the longitudinal shrinkage, which is also a function of moisture content, changes slope in this moisture content region (Ying et al 1994; Kretschmann and Cramer 2007).

Current wood-moisture models do not explain the threshold-like behavior of certain physical properties between 15% and 20% MC such as ionic conduction or corrosion. However, the percolation model is consistent with several other types of measurements that characterize the nature of water in wood, including sorption isotherms, low temperature differential scanning calorimetry (DSC), and nuclear magnetic resonance spectroscopy (NMR).

Many thermodynamic models have been used to describe moisture sorption in wood below fiber saturation. Although these models differ in their parameters and physical interpretation, they all divide water into two types, tightly bound water and less tightly bound water (Simpson 1980). For example, the well-known BET model (Brunauer et al 1938) describes sorption in terms of monolayer (Langmuir) adsorption at low relative humidities with multilayer sorption occurring at higher relative humidities. Other models include the phenomenon of capillary condensation at still higher relative humidities, because

the vapor pressure required for condensation is reduced in capillaries with small radii. Both multilayer sorption and capillary condensation models fit the data well (Simpson 1980). The percolation threshold (16% MC) corresponds to roughly three monolayers of water using the BET model or a capillary with radius 5 nm (Skaar 1988).

Several researchers (Nakamura et al 1981; Hatakeyama and Hatakeyama 1998; Takahashi et al 2003; Kärenlampi et al 2005; Park et al 2006) have used low temperature DSC to measure the solidification (or melting) of water in cellulose of various origins. Three distinct types of water can be inferred from DSC curves: Type I (free) water freezes at the same temperature as bulk water; Type II water freezes at a lower temperature than bulk water; and Type III water is tightly bound and does not freeze (also called "nonfreezing bound water") but is inferred from the total moisture content. The moisture content at which Type II water begins to appear in cellulose ranges between 10% and 20% MC. Nakamura et al (1981) suggested that the freezing temperature of Type II water was depressed because Type II water was loosely bound to the cellulose and termed it "freezable bound water." However, Park et al (2006), who ran similar experiments, argued that Type II water is actually free water, but the freezing point is depressed because it is in capillaries with small diameters, which they calculated to be 20 nm.

Similarly, using NMR, Almeida et al (2007) found three states of water in hardwoods: free water, bound water, and a state intermediate between bound and free water. The lowest moisture content at which intermediate water was observed depended on species, however, it was present at 16 to 17% MC but absent at 11 to 12% MC for sugar maple (*Acer saccharum*) and beech (*Fagus grandifolia*). They interpreted the intermediate water as unbound but in small capillaries within the wood.

In summary, data from sorption isotherms, DSC, and NMR suggest that there is a type of water intermediate between bound and free water,

which we refer to as Type II water. It is not clear from these experiments whether the Type II water is loosely bound or unbound in capillaries, but in either case, this water has different thermodynamic properties from those of bound (Type III) water. Conducting water and Type II water are likely related because they emerge in the same moisture content range, and Type II water is not tightly bound and would be able to participate in the conduction process.

Assuming that ionic conduction occurs in Type II water, percolation theory gives us the moisture content at which there is a continuous path of Type II water in wood, although at this time, it is unclear how this continuous pathway relates to the anatomical features of wood. Therefore, understanding Type II water is key to understanding "aqueous" processes that occur in wood below fiber saturation such as ionic conduction, mold growth, and corrosion. The amount of Type II water at a given moisture content could be determined by more DSC and NMR experiments on softwoods. However, to develop a sub-cellular model for conduction in wood, one would need to know whether Type II water is in capillaries or is loosely bound. Because both theories have been convincingly argued based on sorption isotherms, DSC, and NMR data, a new type of experiment would be needed to determine where Type II water resides.

SUMMARY AND CONCLUSIONS

A percolation model was used to describe the DC conductivity of wood, which partitioned the wood into two phases, an insulating wood phase and a conducting water phase. The conductivity of the conducting phase was related to the conductivity of salinated water. The model suggests that there is a continuous path of conducting water at 16% MC. This conducting water was related to Type II water found in sorption isotherms, and DSC and NMR experiments, which has properties intermediate between bound and free water. The percolation model suggests a physical picture of the distribution of water in wood in which a continuous path of Type II

water exists at the percolation threshold. Therefore, this model can be used to interpret the results of moisture experiments in wood. Likewise, these measurements can be used to better understand the physical mechanism of electrical conduction in wood.

ACKNOWLEDGMENTS

This research was funded by the USDA Forest Service, Forest Products Laboratory (FPL). We are grateful for critical reviews of this manuscript by current FPL employee, Alex Wiedenhoef, as well as former FPL employees, Anton TenWolde and William Simpson. We also appreciate discussions with the following FPL employees: David Kretschmann (on longitudinal shrinkage), Chris Hunt (on wood–water relations), and Patricia Lebow (on the best way to compare R^2 statistics between a power law model and an exponential model).

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APPENDIX A. RELATIONSHIP BETWEEN $\epsilon'(\omega)$ AND ρ

The complex dielectric constant $\epsilon(\omega)$ is related to the impedance, $Z(\omega)$, through:

$$\epsilon(\omega) = (j\omega C_c \cdot Z(\omega))^{-1} \quad (\text{A1})$$

(MacDonald and Johnson 1987) where ω is the angular frequency (rad s^{-1}) and C_c (F) is a geometric factor needed for unit analysis and represents the capacitance of an empty cell (ie, $C_c = \epsilon_o A/L$ where ϵ_o is the permittivity of free space $\{8.8 \times 10^{-12} \text{ F m}^{-1}\}$, A is the cross-sectional area $\{\text{m}^2\}$, and L is the electrode spacing $\{\text{m}\}$). $\epsilon(\omega)$ and $Z(\omega)$ can be written in terms of their real and imaginary components as $\epsilon(\omega) = \epsilon'(\omega) + j\epsilon''(\omega)$ and $Z(\omega) = Z'(\omega) + jZ''(\omega)$ where $j \equiv \sqrt{-1}$. The AC resistivity, $\rho(\omega)$, is related to the real component of the impedance through:

$$\rho(\omega) = (A/L) \cdot Z'(\omega) \quad (\text{A2})$$

Using Eq A1, we can now show how $\epsilon'(\omega)$ is related to ρ .

$$\begin{aligned} \epsilon(\omega) &= \frac{1}{j\omega C_c} \cdot \frac{1}{Z'(\omega) + jZ''(\omega)} \\ &= \frac{-1}{\omega C_c} \cdot \frac{Z''(\omega) + jZ'(\omega)}{(Z''(\omega))^2 + (Z'(\omega))^2} \quad (\text{A3}) \end{aligned}$$

Therefore, $\epsilon'(\omega)$ can be written as a function of $Z'(\omega)$

$$\epsilon'(\omega) = \frac{-1}{\omega C_c} \cdot \frac{Z''(\omega)}{(Z'(\omega))^2 + (Z''(\omega))^2} \quad (\text{A4})$$

and $Z'(\omega)$ is related to ρ through Eq A2. Furthermore $Z'(\omega)$ and $Z''(\omega)$ are related by the Kramers-Kronig relations (MacDonald and Johnson 1987). Therefore, $\epsilon'(\omega)$ and ρ are not independent.