Impedance Analysis and Breakdown Voltage of Dielectric Materials

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Contents

List of Figures

List of Tables

Chapter 1

Introduction

1.1 Summary

The goal of this project was to measure the dielectric and breakdown properties of thin film samples of insulating materials and to assess their suitability for applications in electronics, particularly as gate dielectrics in transistors. The primary method employed was impedance spectroscopy, which was used to determine dielectric constants and conductivities. We successfully performed these measurements on a series of gadolinium-scandate thin film samples prepared by the process of pulsed laser deposition (PLD) and found that the performance of this material as an insulator is not adequate to be useful in applications.

1.2 Motivation and Goals

The dielectric properties of thin film materials are very important for materials to be employed in electronics applications because they determine the extent to which the material can be used as an insulator, as well as its effectiveness as a medium in capacitors. In particular, a successful high dielectric constant thin film material could see applications in two major areas. First, in the construction of semiconductor devices (such as transistors and logic gates), there is a need for high-quality, very thin insulators for use in increasingly small devices where current leakage is a critical issue. Second, there is the possibility of a successful transparent insulator, which could see application as a building block in transparent electronic devices, which are a new and promising area of development. [1]

Our primary goal, in addition to actually measuring the insulator properties for a number of thin film samples, is to develop the techniques to continue these measurements in the future. If we can develop the methods and experimental protocols within the group to do these dielectric measurements, they can become easier and more routine in the future. Previous to this project, neither the knowledge of how to perform the actual measurements nor the knowledge and tools to interpret and analyze the results existed in the group.

1.3 Background - Insulators

Before discussing impedance spectroscopy, we should comment on what constitutes a good insulator for our purposes.

- The most obvious property of a good insulator is that it is not a good conductor. A good insulator will have a low conductivity σ or a high resistivity ρ. This is equivalent to saying that it has a small leakage current.

- A related requirement is that a good insulator should have a high breakdown voltage. Many insulating materials will fail and become conductive if a large enough electric field is applied to the material (or equivalently if a large enough voltage is applied across the material); this process is termed dielectric breakdown. In a good insulator, the field strength of voltage where breakdown occurs will be large.

- A good insulator will have a large dielectric constant ϵ_{r}. This dielectric constant determines how easy it is for an applied electric field to induce a polarization in the material. Strictly speaking, this is a property of a good *dielectric* rather then a good insulator: for purely insulating applications, this is not important. For applications as a gate dielectric in transistors, the dielectric constant is critically important. Because gate dielectrics are a major application for thin film insulators, this is a very important quantity.

These last two qualities, breakdown field strength and dielectric constant, can be combined to determine a figure of merit for the use of the material as

[1] This paragraph is based on text written for the URISC grant proposal for this project.

a gate dielectric. This figure of merit, which we will call the quality factor, is a the product of the breakdown field strength and the dielectric constant. Because the dielectric constant is dimensionless, this figure of merit has units of electric field strength, typically in megavolts per centimeter (MV/cm). Silicon dioxide (SiO_2) thin film insulators have a breakdown voltage of approximately 10 MV/cm and a dielectric constant of approximately 4, making a quality factor of 40MV/cm. In order to be a good candidate for a gate dielectric, a new material should be at least as good as this, preferably better. Because the relevant quantity is the product of the breakdown field strength and the dielectric constant, a large breakdown can make up for a small dielectric constant or vice versa. For application as a gate dielectric it is also important that the leakage current due to the conductivity σ be small, but this is not incorporated numerically into the figure of merit.

For capacitor applications, the dielectric constant, breakdown field strength, and leakage current are likewise important, depending on the application; a larger dielectric constant will *not* necessarily make up for a smaller breakdown voltage as it can in the gate dielectric application. For use in capacitors, the dielectric constant determines the capacitance for a given area; thus it is easier to make higher value capacitors with a larger dielectric constant. The breakdown voltage determines the maximum voltage to which the capacitor can be charged to and limits the operating voltage of a circuit incorporating the capacitor. However, in many applications a capacitor not operate at high voltages, so this parameter is less important in these cases. The leakage current likewise determines the time a capacitor can hold its charge; if the capacitor is not being used for energy storage or is being charged and discharged on time scales much shorter then the discharge time due to leakage, this property will likewise not be important.

1.4 Background - Impedance Spectroscopy

In order to measure the small conductance σ and dielectric constant ϵ_r discussed in the previous section, we will employ the technique of impedance spectroscopy. The goal of impedance spectroscopy is to map out the complex impedance (a complex generalization of resistance that includes capacitive and inductive effects as well) Z of the sample as a function of frequency ω. This is done by applying a small oscillatory voltage to the sample and measuring the resulting oscillatory current through the sample. The phase and amplitude difference between the voltage across and the current through the sample determines the impedance of the sample at that frequency. By varying the frequency we can map out the impedance spectrum. Once the impedance spectrum is known, it can be fit against analytical formulas for impedance derived from circuit models, yielding an equivalent resistance and capacitance, which can be converted into resistivity and dielectric constant if the sample geometry is known.

1.5 Previous Work

This follows directly from work done by Briony Horgan in her senior thesis[6]. Horgan did similar impedance spectroscopic analysis on bulk pellet samples. In bulk pellet materials, the influence of grain boundary effects tends to overshadow the intrinsic properties of the material, making it difficult to use this method to make dielectric constant measurements of a particular material. This difficulty in determining dielectric properties of bulk pellets provides the need for using this method in thin film materials.

In the literature, impedance spectroscopy mostis often employed in two areas. The first primary area of application is in the analysis of electrolytic cells in chemistry. These cells are not mainly dielectric in nature so the analysis is employed in different ways[1]. Impedance spectroscopy is also employed in a more similar application in ceramic and bulk materials[2, 8, 4]. In this case the analysis is similar, but effects such as grain boundaries tend to dominate; see [7] for a derivation of the brick layer model for grain boundaries in these systems.

1.6 Methods

1.6.1 Sample preparation

The thin film samples to be tested were produced by the method of pulsed laser deposition, the details of which are beyond the scope of this document. In order to measure properly calibrated dielectric constants and breakdown field strengths, it is necessary to measure the thickness of the sample optically before metallic contacts are deposited. If this is not done, it is still possible to determine the figure of merit for gate dielectric applications (the product

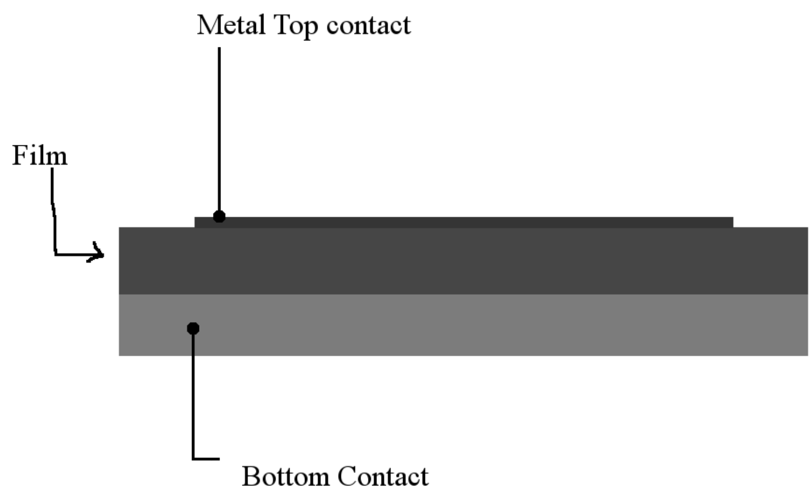

Figure 1.1: A side view of the sample with top and bottom contacts and film indicated. Not to scale. Each top contact is a circular dot when viewed from above.

of breakdown field strength and dielectric constant) but not to determine either independently.

The next critical step in sample preparation was to deposit metallic top contacts onto the sample. These define the area of the sample under test, act as a top capacitor plate, and allow us to make electrical contact to the top of the sample. These contacts were deposited using a mask to create small circular aluminum dots as top contacts. For a side view of the sample indicating top contact, bottom contact and film, see Figure 1.1.

A critical and potential troublesome step is making a bottom contact to the sample. The samples are deposited on conducting substrates, either a metallic layer or doped silicon depending on the sample. During the PLD process, some portions of the substrate are covered by clips and are not covered by the film. During the top contact deposition, these become covered in large metallic contacts that appear to be in electrical contact with the substrate of the film, apparently making an easy point of contact for the measurement. These clip mark areas should *not* be used for making contact to the substrate! In our experience there appears to be an insulating layer that is formed on the substrate, so the clip marks do not make a good contact to the substrate and at best exhibit very large contact resistances.

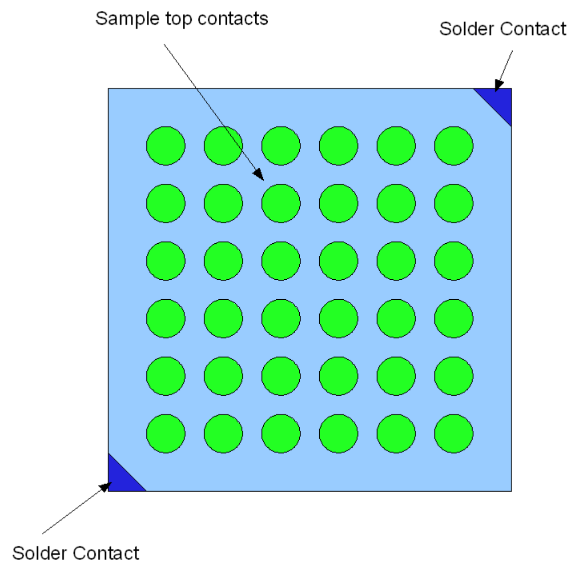

Figure 1.2: A diagram of a typical film in top down view with solder contacts and top contacts indicated

Contact should be made properly after depositing the top contacts by scribbling through the film in an unused area (typically a corner) to expose the substrate directly and making a small solder contact to fill the hole (see Figure 1.2 for a diagram of a typical sample). Although only one back contact point is needed, it is best to create at least two on opposite corners. This allows us to measure the resistance across the substrate (this resistance should be less then 100Ω) and verify that the quality of the contacts is good.

1.6.2 Measurement

When taking measurements of either the breakdown voltage or dielectric constant, the sample will be mounted in a metal box and set on on an aluminum backing. Connection is made from the top via two movable probes attached to the bottom of the box with magnets. One probe has a sharp tip and the other a rounded tip. The probes each connect to the central conductors of two separate BNC connectors on the front panel of the box. The sharp (black) probe is used to connect to the soldered connection to the sample substrate, *not* to the sample top contacts. The rounded (red) probe is used to connect to the top contact of the dot under testing. Because the sample thickness varies across the surface, dots near the center should be used. Because the top contacts are very thin, using the sharp probe risks punching holes in the sample.

Once the probes are located on a particular sample dot, all connections for different measurements should made by changing the connections to the front panel connectors, and all measurements for that dot must be made at that time, without moving the contacts. Because of the fragility of the film, removing and replacing the contacts may damage the film and change the results. Furthermore, the impedance spectroscopy measurement for the dielectric constant must be taken first, before the dielectric breakdown measurement. The breakdown is a destructive measurement and it may not be possible to get good measurements of the dielectric constant after breakdown has been measured for a given sample dot. Also the breakdown measurement can be taken only once. Once the dot has broken down, it will no longer exhibit insulator behavior and will not exhibit a proper breakdown in future measurements. Because of these considerations, measurements at a particular sample point are *not* repeatable.

1.6.3 Data Analysis and Curve fitting

The data resulting from the impedance spectroscopy measurement were processed and fit against a model with a contact resistance and parallel RC resistance representing the film. This was done using software developed in MatLAB by the author for this project, which is described elsewhere. The curve fitting is done using a multi-dimensional optimization routine.

Chapter 2

Theory

In this chapter we will derive a number of results that are important for our use of impedance spectroscopy. First, we will discuss the concept of impedance in circuit analysis. Second, we will derive the impedance of several important model circuits we will use in our analysis. Third, we will show that the simple model of a resistor in parallel with a capacitor can be derived by using the standard formula for the impedance of a capacitor and allowing it to have complex dielectric constant. Fourth we will derive the capacitance of a finite plate above an infinite plate and show that this configuration is equivalent with the standard parallel plate capacitor result. Lastly we will consider a system consisting of a capacitor having two layers and show that if one layer is much thinner than the other, all the relevant quantities we are interested in measuring reduce to those of the thicker layer. This is relevant, for example, in the case of a substrate with a thin intrinsic oxide layer with a sample layer on top of it. This result lets us neglect such layers provided our films are of reasonable thickness.

2.1 Impedance Basics

Impedance (typically denoted Z) is a generalization of resistance by allowing complex values. A strictly real impedance Z is identical to a resistance R. This generalization allows us to include affects of reactive elements such as inductors and capacitors in our circuit analysis. When dealing with impedance, the familiar form of Ohm's law $V = IR$ is replaced by:

$$V(\omega) = I(\omega)Z(\omega) \tag{2.1}$$

In engineering references it may also be written as $V(j\omega) = I(j\omega)Z(j\omega)$, or even $V(s) = I(s)Z(s)$. This is because electrical engineering circuit analysis texts sometimes do their analysis in the Laplace Transform domain, which is a generalization of the Fourier to allow complex frequencies, which are often denoted by s. The imaginary part of s corresponds to the real frequency ω that appears in the Fourier transform and setting $s = j\omega$ reduces the equations to the conventional real frequency form. In this document we will consider only real frequencies ω and will not concern ourselves further with the Laplace transform representation. Regardless of presentation, Ohm's Law for impedances is typically abbreviated to $V = IZ$, although it is important to remember that in general all three quantities are functions of frequency.

The values of the impedance of the standard passive circuit elements (resistors, capacitors and inductors) are well known and can be found in many books, for example [9, pg. 393-396]. The impedance of a resistor is simply the value of the resistor itself.

$$Z_{\text{Resistor}} = R \tag{2.2}$$

The impedance of a capacitor is entirely imaginary, and is given by:

$$Z_{\text{Capacitor}} = \frac{1}{i\omega C} = \frac{-i}{\omega C} \tag{2.3}$$

Note that engineering references frequently replace i with j to represent the imaginary number.

In addition to the impedance of the basic circuit elements, we also require the rules for combining impedances in series and parallel[9, pg. 398]. Two impedances Z_1 and Z_2 combined in series have the total impedance:

$$Z_{\text{series}} = Z_1 + Z_2 \tag{2.4}$$

Two impedances combined in parallel have the combined impedance:

$$Z_{||} = Z_1 || Z_2 = \left(\frac{1}{Z_1} + \frac{1}{Z_2} \right)^{-1} = \frac{Z_1 Z_2}{Z_1 + Z_2} \tag{2.5}$$

It is sometimes helpful to use the notation $\Re\{Z\}$ to refer to the real part of the Z and $\Im\{Z\}$ to refer to the imaginary part.

Because the impedance combines information relating resistive elements and capacitive or inductive elements in a single quantity, it allows us to consider both leakage currents and dielectric behavior in the same model.

Essentially, impedance spectroscopy is the measurement of the mapping out of the impedance as a function of frequency so that $Z(\omega)$ is known. This is done by introducing a sinusoidal current or voltage into the sample at a known frequency and measuring the relative phase and amplitude of the other quantity at the same point. The impedance can be calculated mathematically as:

$$\frac{V(\omega)}{I(\omega)} = Z(\omega) \tag{2.6}$$

Provided that the response of the material is linear, both voltage and current will have the same frequency. If we consider a reference voltage with amplitude V_0 and zero phase, we can write this as $V = V_0 e^{i\omega t}$. In this case the resulting current will be given by $I = I_0 e^{i(\omega t+\phi)}$ where ϕ is the relative phase. This will result in:

$$Z(\omega) = \frac{V_0 e^{i\omega t}}{I_0 e^{i(\omega t+\phi)}} = \frac{V_0}{I_0} e^{-i\phi} \tag{2.7}$$

In this case a positive ϕ denotes current which is leading voltage. Thus $\phi = \pi/2$ corresponds to a negative imaginary impedance, a purely capacitive element, and $\phi = -\pi/2$ corresponds to a positive imaginary impedance, a purely inductive element. We will not consider inductive elements further in this document. See any standard text on circuit theory. This relation allows us to directly map out $Z(\omega)$ using sinusoidal excitations. This forms the fundamental theoretical basis of impedance spectroscopy. Although we do not directly measure these currents and voltages, instead using an impedance analyzer, understanding this relation between relative amplitude and phase and impedance is important to understand the theory employed in analyzing the results further.

2.2 Equivalent Circuit Models and Derivations

2.2.1 Models

Introduction

In order to apply impedance spectroscopy to the problem of determining the electrical properties of dielectric materials, it is useful to employ an idealized circuit model to describe the material and derive formulas describing its impedance. We will discuss four such models of increasing complexity. All of the models we will address are only appropriate in a small-signal regime where non-linear and breakdown effects can be neglected.

Ideal Capacitor Model

The simplest circuit model is a single ideal capacitor (See Figure 2.1). This will give dielectric behavior but assumes infinite breakdown voltage, no leakage current at DC, and zero contact resistance. This model is appropriate for modeling a single layer of a very good insulator with good contacts and no grain boundary effects. For a capacitor the impedance is [9, pg. 395-396]:

$$Z(\omega) = \frac{-i}{\omega C} \tag{2.8}$$

where ω is radian frequency.

Series Resistor and Capacitor Model

We can use a somewhat more complex model and treat the material as a series combination of a resistor and capacitor (see Figure 2.2). This will take into account contact resistances, but not leakage currents. This model is appropriate for a single layer of a good insulator with contact resistance and no grain boundary effects. For this model the impedance is:

$$Z(\omega) = R_\mathrm{c} - i\frac{1}{\omega C} \tag{2.9}$$

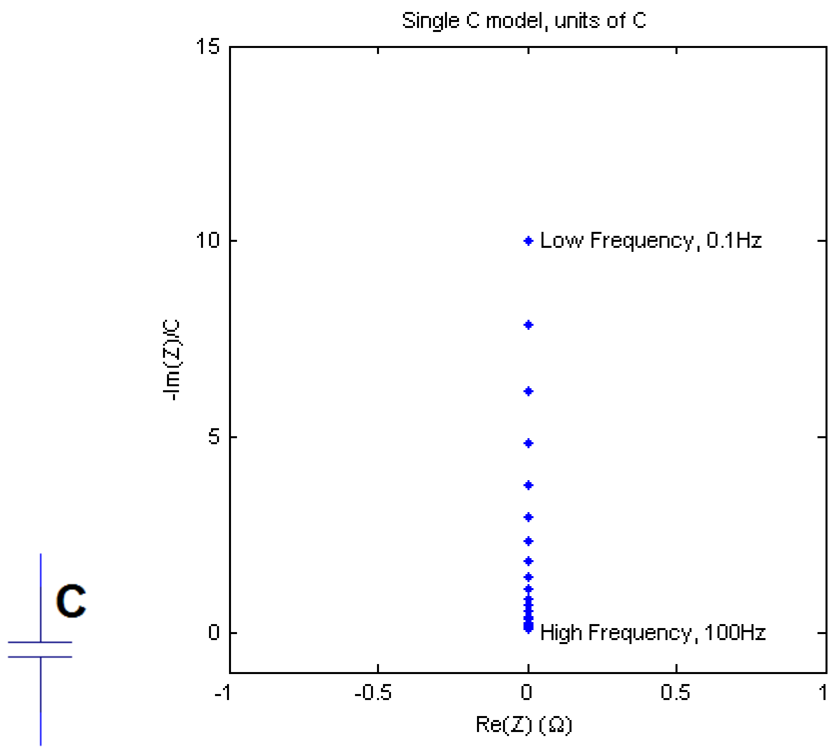

Figure 2.1: Circuit diagram of ideal capacitor model (left) and real versus imaginary impedance plot (right).

If the impedance $Z(\omega)$ is given, we can find the two parameters of this model, R_c and C, as:

$$R_c = \Re\{Z\} \tag{2.10}$$

$$C = \frac{-1}{\omega \Im\{Z\}} \tag{2.11}$$

Parallel Resistor and Capacitor Model

The next circuit model we may use to model a dielectric material is an ideal resistor in parallel with an ideal capacitor (see Figure 2.3). This takes into account leakage conduction in the material and allows us to determine both the small signal conductivity of the material and the dielectric constant. This model is appropriate for a leaky dielectric with good contacts and no grain

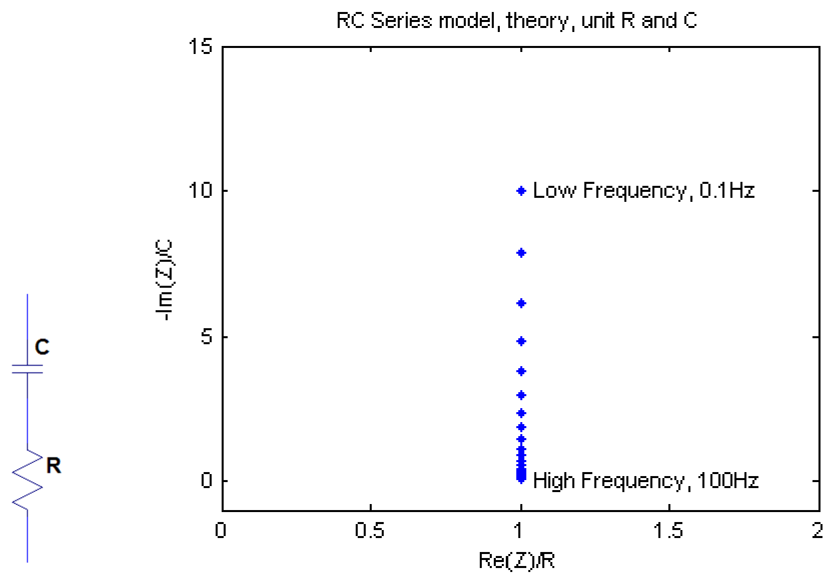

Figure 2.2: The circuit model for the simple series resistor capacitor model (left) and the real versus imaginary impedance plot for the same, in units of R and C (right).

boundary effects. For this configuration the impedance is:

$$Z(\omega) = \frac{R}{(\omega RC)^2 + 1} - i\frac{\omega R^2 C}{(\omega RC)^2 + 1} \tag{2.12}$$

If the real and imaginary parts of the impedance Z are known, we can find the parameters of the model as:

$$R = \frac{|Z|^2}{\Re\{Z\}} \tag{2.13}$$

$$C = \frac{-\Im\{Z\}}{\omega |Z|^2} \tag{2.14}$$

The conductance G is equal to R^{-1} so

$$G = \frac{\Re\{Z\}}{|Z|^2} \tag{2.15}$$

For the derivation of these results, see Section 2.2.2.

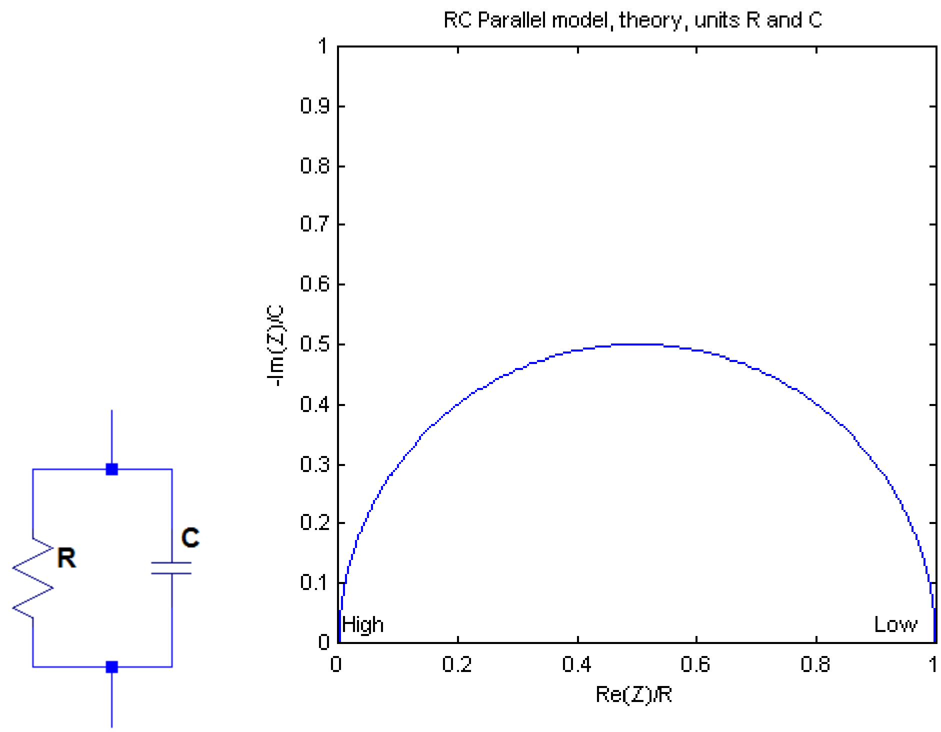

Figure 2.3: The circuit model for the simple parallel resistor capacitor model (left) and real versus imaginary impedance plot for the same, in units of R and C (right).

Parallel Resistor and Capacitor with Contact Resistance

Combining the two previous models yields the more complete model with a contact resistance in series with a parallel combination of a capacitor and resistor (see Figure 2.4). This model takes into account leakage currents and contact resistance and is appropriate for a single layer of a leaky dielectric with resistive contacts and no grain boundary effects.

$$Z(\omega) = R_c + \frac{R}{(\omega RC)^2 + 1} - i\frac{\omega R^2 C}{(\omega RC)^2 + 1} \tag{2.16}$$

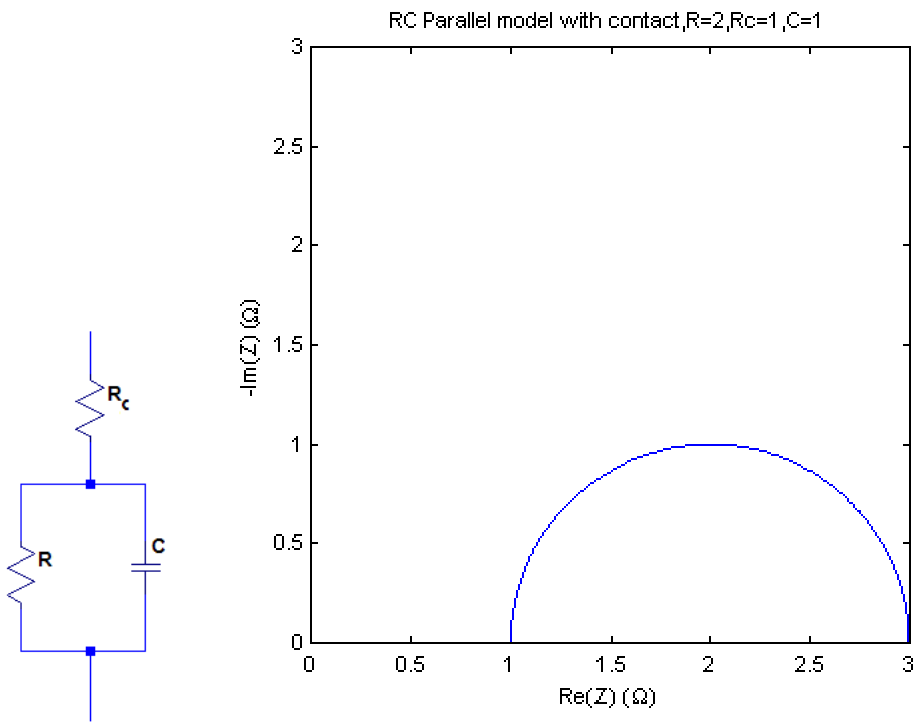

Figure 2.4: The circuit model for the parallel resistor-capacitor model with a contact resistance (left) and real versus imaginary impedance plot (right).

2.2.2 Deriving impedances for circuit models

Using the relations introduced in Section 2.1 the total impedance $Z(w)$ of any of the given circuit models can be calculated using the known impedances of the component elements in the model, in this case resistors (R) and capacitors (C) and the rules for parallel and series combinations of these elements.

Deriving the RC Series Model

The simplest model to derive the impedance of is a series combination of a resistor and capacitor because the elements are simply added.

$$Z_1 = R \tag{2.17}$$

$$Z_2 = \frac{-i}{\omega C} \tag{2.18}$$

And combining these in series yields:

$$\begin{aligned} Z &= Z_1 + Z_2 && (2.19) \\ &= R - i\frac{1}{\omega C} && (2.20) \end{aligned}$$

Or equivalently:

$$\Re\{Z\} = R \tag{2.21}$$

$$\Im\{Z\} = \frac{1}{\omega C} \tag{2.22}$$

Note that in this case the real part is independent of both frequency and capacitance, and the imaginary part is independent of resistance.

Deriving Parallel RC model

Deriving the impedance of a parallel combination of a a resistor and capacitor is a direct application of the impedance of the two elements and the rule for parallel combinations, with:

$$Z_1 = R \tag{2.23}$$

$$Z_2 = \frac{-i}{\omega C} \tag{2.24}$$

Applying the parallel combination rule gives:

$$\begin{aligned}
Z &= Z_1 \| Z_2 && (2.25)\\
&= \frac{Z_1 Z_2}{Z_1 + Z_2} && (2.26)\\
&= \frac{\frac{-i}{\omega C} \cdot R}{R - \frac{i}{\omega C}} && (2.27)\\
&= \frac{-Ri}{\omega RC - i} && (2.28)\\
&= \frac{-Ri(\omega RC + i)}{(\omega RC)^2 + 1} && (2.29)\\
&= \frac{R - \omega R^2 C i}{(\omega RC)^2 + 1} && (2.30)\\
&= \frac{R}{(\omega RC)^2 + 1} - i \frac{\omega R^2 C}{(\omega RC)^2 + 1} && (2.31)
\end{aligned}$$

We could equivalently write the above as:

$$\Re\{Z\} = \frac{R}{(\omega RC)^2 + 1} \quad (2.32)$$

$$\Im\{Z\} = -\frac{\omega R^2 C}{(\omega RC)^2 + 1} \quad (2.33)$$

We can see that both components of the impedance depend on R, C and the frequency ω.

Solving Parallel RC Model for R and C

We can also solve the model presented in the previous section for the parameters R and C in terms of Z, which is useful if the impedance is known (for example from impedance analyzer data) and one wishes to calculate the equivalent circuit parameters. We can begin by writing Equation (2.26) in a different form:

$$Z = \frac{1}{R^{-1} + i\omega C} \quad (2.34)$$

or

$$Z^{-1} = R^{-1} + i\omega C \quad (2.35)$$

Splitting Z^{-1} into its real and imaginary parts gives:

$$\Re\{Z^{-1}\} = R^{-1} \tag{2.36}$$

$$\Im\{Z^{-1}\} = \omega C \tag{2.37}$$

Now we must determine the value of $\Re\{Z^{-1}\}$ and $\Im\{Z^{-1}\}$. Note that in general $\Re\{Z^{-1}\} \neq \Re\{Z\}^{-1}$, and likewise for the imaginary part. To begin, by definition:

$$Z = \Re\{Z\} + i\Im\{Z\} \tag{2.38}$$

And so

$$Z^{-1} = \frac{1}{\Re\{Z\} + i\Im\{Z\}} \tag{2.39}$$

$$= \frac{(\Re\{Z\} - i\Im\{Z\})}{(\Re\{Z\} + i\Im\{Z\}) \cdot (\Re\{Z\} - i\Im\{Z\})} \tag{2.40}$$

$$= \frac{\Re\{Z\} - i\Im\{Z\}}{\Re\{Z\}^2 + \Im\{Z\}^2} \tag{2.41}$$

$$= \frac{\Re\{Z\} - i\Im\{Z\}}{|Z|^2} \tag{2.42}$$

Split into real and imaginary parts, this gives us:

$$\Re\{Z^{-1}\} = \frac{\Re\{Z\}}{|Z|^2} \tag{2.43}$$

$$\Im\{Z^{-1}\} = \frac{-\Im\{Z\}}{|Z|^2} \tag{2.44}$$

Plugging the first of these into Equation (2.36) gives us:

$$R^{-1} = \frac{\Re\{Z\}}{|Z|^2} \tag{2.45}$$

$$R = \frac{|Z|^2}{\Re\{Z\}} \tag{2.46}$$

Plugging Equation (2.44) into Equation (2.37) yields:

$$\omega C = \frac{-\Im\{Z\}}{|Z|^2} \tag{2.47}$$

$$C = \frac{-\Im\{Z\}}{\omega|Z|^2} \tag{2.48}$$

These are the two relations we wished to derive.

Deriving parallel RC model with contact resistance.

The model with contact resistance and a parallel RC circuit is simply a series combination of the a parallel RC circuit derived earlier and a resistor R_c representing the contact resistance.

$$Z_1 = R_\mathrm{c} \tag{2.49}$$

$$Z_2 = \frac{R}{(\omega RC)^2 + 1} - i\frac{\omega R^2 C}{(\omega RC)^2 + 1} \tag{2.50}$$

Using the series combination rule yields:

$$Z = Z_1 + Z_2 \tag{2.51}$$

$$= R_\mathrm{c} + \frac{R}{(\omega RC)^2 + 1} - i\frac{\omega R^2 C}{(\omega RC)^2 + 1} \tag{2.52}$$

Or equivalently:

$$\Re\{Z\} = R_\mathrm{c} + \frac{R}{(\omega RC)^2 + 1} \tag{2.53}$$

$$\Im\{Z\} = -\frac{\omega R^2 C}{(\omega RC)^2 + 1} \tag{2.54}$$

We can see that this is very similar to the model without contact resistance; only the R_c in the real part is changed.

2.3 RC parallel Model From Complex Dielectric Constant

In this section, we wish to show that one can derive the model of a material as a parallel combination of a resistor and capacitor by introducing a complex dielectric constant (permitivity) ϵ_c.

We begin with the complex form of the permittivity of a material, given on page 341 of [3] as:

$$\epsilon_\mathrm{c} = \epsilon - i\frac{\sigma}{\omega} \tag{2.55}$$

If we plug this into the standard form of the capacitance of a parallel plate capacitor to arrive at a "complex capacitance", we get the following:

$$C_\mathrm{c} = \frac{A}{d}\epsilon_\mathrm{c} \tag{2.56}$$

$$= \frac{A}{d}\left(\epsilon - i\frac{\sigma}{\omega}\right) \tag{2.57}$$

$$= \frac{A}{d}\epsilon - i\left(\frac{A}{d}\sigma\right)\omega^{-1} \tag{2.58}$$

$$= C - iR^{-1}\omega^{-1} \tag{2.59}$$

Note that in the last step I have simply identified the (strictly real) forms of R and C.

If we now proceed to calculate the complex impedance Z using this 'capacitance' C_c, we arrive at the following:

$$Z = \frac{1}{i\omega C_\mathrm{c}} \tag{2.60}$$

$$= \frac{1}{i\omega\left(C - iR^{-1}\omega^{-1}\right)} \tag{2.61}$$

$$= \frac{1}{i\omega C + R^{-1}} \tag{2.62}$$

$$= \frac{R}{i\omega RC + 1} \tag{2.63}$$

$$= \frac{R(-i\omega RC + 1)}{(\omega RC)^2 + 1} \tag{2.64}$$

$$= \frac{R}{(\omega RC)^2 + 1} - i\frac{R^2 C\omega}{(\omega RC)^2 + 1} \tag{2.65}$$

This is the same form of the impedance derived earlier from the parallel RC circuit model. We can also rewrite Equation (2.62) as:

$$Z^{-1} = i\omega C + R^{-1} \tag{2.66}$$

$$= \left(\frac{1}{i\omega C}\right)^{-1} + (R)^{-1} \tag{2.67}$$

Which is the formula for the parallel combination of an impedance $1/i\omega C$ and R. Thus we can model a material with permittivity given in (2.55) as a parallel combination of an ideal resistor and ideal capacitor.

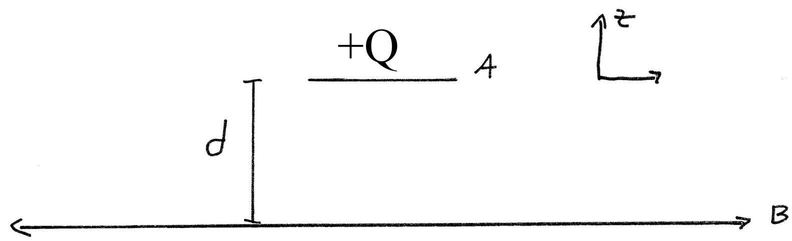

Figure 2.5: A capacitor formed by a small dot above a very large plane, with a charge Q transported from the lower plate to the upper plate.

2.4 Capacitance Derivation

In this section we wish to derive the capacitance C of the configuration of a small top contact of area A above an infinite ground plane bottom contact and show that this is the same as the result for a standard parallel plate capacitor with two plates of equal area.

This system can be seen in cross section in Figure 2.5. We model the upper plate as a thin conducting plane and the lower plate as a thick conducting grounded slab We introduce a charge Q on the upper plate. Because we are modeling the lower plate as a conducting slab, this will create an image charge $-Q$ at a distance d inside the plane. The real top contact and its image will both have charge densities $\sigma = \pm Q/A$ where the plus corresponds to the upper plate and the minus to the lower. The net electric field in the $\hat{z}$ direction will thus be:

$$\vec{E} = -\left(\frac{\sigma}{2\epsilon} + \frac{\sigma}{2\epsilon}\right)\hat{z} = -\frac{\sigma}{\epsilon}\hat{z} \tag{2.68}$$

We can now find the voltage across the the capacitor as:

$$V = -\int_{\mathrm{A}}^{\mathrm{B}} \vec{E} \cdot \mathrm{d}\vec{r} \tag{2.69}$$

$$= -\int_{z_0+d}^{z_0} \left(-\frac{\sigma}{\epsilon}\hat{z}\right) \cdot (-\hat{z})\, \mathrm{d}z \tag{2.70}$$

$$= \frac{\sigma}{\epsilon}\int_{z_0}^{z_0+d} \mathrm{d}z \tag{2.71}$$

$$= \frac{\sigma}{\epsilon}d \tag{2.72}$$

$$= \frac{Q}{A\epsilon}d \tag{2.73}$$

From this we can calculate the capacitance using the definition of capacitance:

$$C = \frac{Q}{V} \tag{2.74}$$

$$= \epsilon\frac{A}{d} \tag{2.75}$$

This is the standard result for a parallel plate capacitor. This shows us that the in a parallel plot capacitor, the effective area is dominated by the *smaller* of the two plates. Only the portion of the structure that lies between two plates contributes to the total capacitance.

2.5 Two Layer Capacitor Model

2.5.1 Introduction

We wish to calculate the capacitance C and conditions for dielectric breakdown for a capacitor made up of layers of two different materials. Let us assume that the first layer has thickness d_1, dielectric constant ϵ_{r1} and breakdown field strength E_{B1}, and that the second material correspondingly has thickness d_2, dielectric constant ϵ_{r2} and breakdown field strength E_{B1}.

In order to calculate the capacitance and field strength within the material, we need to introduce the 'electric displacement', denoted by $\vec{D}$ and for linear media given by[5, pg. 180]:

$$\vec{D} = \epsilon\vec{E} \tag{2.76}$$

where $\vec{E}$ is the electric field strength and $\epsilon \equiv \epsilon_{\mathrm{r}}\epsilon_0$. Gauss's Law can be written in terms of $\vec{D}$ as[5, pg. 175]:

$$\vec{\nabla} \cdot \vec{D} = \rho_{\mathrm{f}} \tag{2.77}$$

where ρ_{f} is the free charge density, or in integral form as

$$\oint \vec{D} \cdot d\vec{A} = Q_{\mathrm{f,enc}} \tag{2.78}$$

Where $Q_{\mathrm{f,enc}}$ is the total enclosed free charge. We can see from these formulations that $\vec{D}$ depends only on charge densities, not dielectric properties of the media.

2.5.2 Infinite plane charge

An useful intermediate result is the electric displacement due to an infinite plane charge with free surface charge density σ. If we enclose the plane charge in a gaussian pillbox and apply the integral form of Gauss's Law, we get:

$$\int_{\mathrm{top}} \vec{D} \cdot d\vec{A} + \int_{\mathrm{bottom}} \vec{D} \cdot d\vec{A} + \int_{\mathrm{sides}} \vec{D} \cdot d\vec{A} = \sigma A \tag{2.79}$$

$$DA + DA + 0 = \sigma A \tag{2.80}$$

$$D = \frac{\sigma}{2} \tag{2.81}$$

This result is analogous to the standard result for the electric field of an infinite plane charge in free space:

$$E = \frac{\sigma}{2\epsilon_0} \tag{2.82}$$

2.5.3 Capacitance

We can now use the result of the last section to calculate the capacitance of our two layered capacitor. Let us assume the capacitor is charged to a voltage V with a charge Q, and note that the capacitance C is defined as [5, pg. 104]:

$$C \equiv \frac{Q}{V} \tag{2.83}$$

If we treat the top and bottom contacts of our capacitor as infinite charged plates with charge densities of σ and $-\sigma$ respectively, then total electric displacements between the plates will be:

$$D = \frac{\sigma}{2} + \frac{\sigma}{2} = \sigma \tag{2.84}$$

and D will point from the positively charged plate to the negatively charged plate. In order to calculate the capacitance we next need to find the voltage between the two plates. In order to do this we need to know the electric field strength in the two different materials. We can solve Equation (2.76) for $\vec{E}$ and get:

$$\vec{E} = \frac{1}{\epsilon}\vec{D} \tag{2.85}$$

So in the the two materials we have:

$$E_1 = \frac{D}{\epsilon_1} \tag{2.86}$$

$$E_2 = \frac{D}{\epsilon_2} \tag{2.87}$$

We can now calculate the voltage V as:

$$\begin{aligned} V &= \int_{+\text{plate}}^{-\text{plate}} \vec{E} \cdot d\vec{r} && (2.88) \\ &= E_1 d_1 + E_2 d_2 && (2.89) \\ &= \frac{D}{\epsilon_1} d_1 + \frac{D}{\epsilon_2} d_2 && (2.90) \\ &= D\left(\frac{d_1}{\epsilon_1} + \frac{d_2}{\epsilon_2}\right) && (2.91) \\ &= \sigma\left(\frac{d_1}{\epsilon_1} + \frac{d_2}{\epsilon_2}\right) && (2.92) \end{aligned}$$

We can plug this result together with $Q = \sigma A$ into (2.83) to get a expression for the capacitance C.

$$\begin{aligned} C &= \frac{\sigma A}{\sigma\left(\frac{d_1}{\epsilon_1} + \frac{d_2}{\epsilon_2}\right)} && (2.93) \\ &= \frac{A}{\frac{d_1}{\epsilon_1} + \frac{d_2}{\epsilon_2}} && (2.94) \end{aligned}$$

This is a compact form of the result. We can continue to manipulate the expression to put it in terms of the separate capacitances of the two layers.

$$C = \frac{A}{\frac{d_1\epsilon_2}{\epsilon_1\epsilon_2} + \frac{d_2\epsilon_1}{\epsilon_1\epsilon_2}} \tag{2.95}$$

$$= \frac{A\epsilon_1\epsilon_2}{d_1\epsilon_2 + d_2\epsilon_1} \tag{2.96}$$

$$= \frac{\frac{A}{d_1 d_2} \cdot A\epsilon_1\epsilon_2}{\frac{A}{d_1 d_2} \cdot (d_1\epsilon_2 + d_2\epsilon_1)} \tag{2.97}$$

$$= \frac{\left(A\frac{\epsilon_1}{d_1}\right)\left(A\frac{\epsilon_2}{d_2}\right)}{A\frac{\epsilon_2}{d_2} + A\frac{\epsilon_1}{d_1}} \tag{2.98}$$

$$= \frac{C_1 C_2}{C_1 + C_2} \tag{2.99}$$

$$= \left(\frac{1}{C_1} + \frac{1}{C_2}\right)^{-1} \tag{2.100}$$

where the last step recovers the standard circuit theory formula for a series combination of two capacitors if C_1 and C_2 represent the capacitances of the layers treated separately.

$$C_1 \equiv \epsilon_1 \frac{A}{d_1} \tag{2.101}$$

$$C_2 \equiv \epsilon_2 \frac{A}{d_2} \tag{2.102}$$

The form in Equation (2.100) has the property that the result will be dominated by the smallest capacitance: if $C_1 \ll C_2$ then $C \approx C_1$. This result is useful because the capacitance of a given layer is inversely proportional to the thickness of that layer. Particularly, in the case with a thick layer of desired material and thin layer of unwanted material (for example, oxide layer on a silicon substrate), assuming the two materials' dielectric constants are of a similar order the thinner material will have a much higher capacitance and will largely not affect the overall capacitance.

2.5.4 Breakdown Field Strength

Let us assume that we have applied a known voltage V across the same capacitor discussed in the previous section and that we also now know the overall capacitance C for the capacitor. We are now in a position to calculate the electric field strength in the two regions and determine if breakdown will occur in either or both. We can first calculate the total charge as:

$$Q = CV \tag{2.103}$$

and surface charge density as:

$$\sigma = \frac{Q}{A} = \frac{CV}{A} \tag{2.104}$$

And since in this geometry we know from Equation (2.84) that $\sigma = D$ we also know the electric displacement and can now calculate the electric fields in either of the two layers as:

$$E_n = \frac{\sigma}{\epsilon_n} = \frac{CV}{A\epsilon_n} \tag{2.105}$$

where n can be either 1 or 2. So we can see that in the case where neither material has broken down, the electric field strength inside a given layer of the material depends on the dielectric constant of that material and the overall capacitance and applied voltage, but does not depend directly on the composition of the other layer. We can also place the dielectric constant on the other side of the equation:

$$E_n\epsilon_n = \frac{CV}{A} \tag{2.106}$$

We can recognize the left hand side as similar to the metric we are using to define the quality of an insulator: the product of the breakdown voltage and the dielectric constant of the material. This tells us that in this configuration, whichever material is a poorer insulator by that metric will breakdown first, regardless of the relative thickness of the two layers. Note that this is under the assumption of no ohmic leakage currents.

Let us now consider the case where the field strength rises such that one material breaks down and the other does not. Let us consider the case where the first material breaks down before the second. If we assume[1] that once breakdown occurs, charges in layer one are free to move and the layer will

[1] Temporary Note: I don't know if this is a good assumption or not, my E&M references don't talk much about breakdown mechanisms.

act as a conductor, then we essentially have only layer two remaining and $C' = C_2$, where C' is the total capacitance after breakdown.

If we further assume that one layer was much more capacitive then the other, this gives two cases for C'.

$$C' \approx C \text{ where } C_1 \gg C_2 \tag{2.107}$$

$$C' \gg C \text{ where } C_1 \ll C_2 \tag{2.108}$$

The first case corresponds to the situation where a very thin contamination layer breaks down before the sample layer. This will not change the overall capacitance of the system significantly and so will not change the electric field strength in the other layer; the second layer will not break down as a result of the break down of the first. The second case corresponds to the much thicker desired sample layer breaking down before the thin contamination layer. In this case the overall capacitance of the configuration will be dramatically higher then it was before the breakdown occurred and the electric field strength in the second material will likewise increase dramatically. Unless the second material is a much better insulator then the first, this dramatic increase in field strength will likely cause the second material to break down as well, thus breaking down the entire stack and ending the experiment.

2.5.5 Conclusions

Under the conditions that the desired sample material layer is much thicker then any contaminating oxide layers and has a dielectric constant as good as or better than the contaminating layer, the presence of a second contaminating layer will not have a large impact on the capacitance of the overall configuration. Under the same assumption and the added assumption that the breakdown strength of the two materials is of somewhat similar order, the overall configuration of materials will show breakdown only when the sample material itself breaks down.

Chapter 3

Data and Results

A series of samples were prepared and measured as described in Section 1.6. We obtained results for a series of gadolinium scandate film samples prepared under different conditions. The results of this experiment are given in the following two sections.

3.1 GSO Results

We measured seven films prepared under different conditions. The films are denoted by GSO-N, where N identifies a particular film. However, the N number refers to deposition order and not all deposited films were used in this experiment.

All of the GSO series films exhibit poor breakdown characteristics, with the tenth film, numbered GSO-10, having the highest average breakdown (0.91 MV/cm). Most films in the series had dielectric constants of approximately 25, while GSO-10 was somewhat lower with a dielectric constant of approximately 20 and GSO-9A and GSO-9B were aberrantly high with dielectric constants of approximately 60 and 48 respectively. The dielectric constants and breakdown can be seen plotted in Figures 3.1 and 3.2. The solid line in the figures represents combinations of breakdown and dielectric constant equal to that of SiO_2. The area outside of the solid line (above and to the right) represents materials that perform better then SiO_2, which we have not yet achieved. The same data and the deposition parameters of the films is given numerically in Table 3.1. The far left column of this table gives the product of the dielectric constant and breakdown field strength, which is a measure of insulator performance; for SiO_2 this value is approximately 40, whereas for GSO-10, which is our best film, it is only 18.

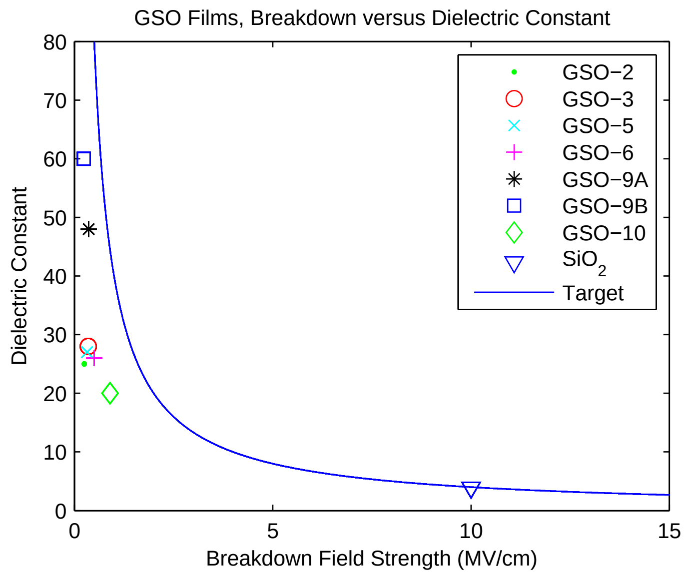

Figure 3.1: Plot of breakdown versus dielectric constant for all GSO films, far view. The solid line is the set of points having the same product of dielectric constant and breakdown field strength as SiO_2.

3.2 GSO-10 Results

3.2.1 Film Parameters

GSO-10 was the last $GdScO_3$ film we deposited and was deposited at a higher temperature than the earlier films. Because of the high temperature, the film could not be deposited on a tantalum wafer, as some of the earlier lower temperature films had been, but was deposited on a doped silicon substrate. The detailed parameters are given in Table 3.2.

3.2.2 Data

The impedance analyzer used in this experiment is configured to give results in terms of a resistance and capacitance, or equivalently, conductance and di-

Film	Substrate	T_{sub} (°C)	O_2 pp mtorr	d nm	ϵ_r	E_{break} MV/cm	Quality MV/cm
GSO-2	Ta-coated Si	140	5	141	25	0.25	6.25
GSO-3	Ta-coated Si	140	5	130	28	0.35	9.80
GSO-5	Ta-coated Si	230	5	≈140	27	0.32	8.64
GSO-6	Ta-coated Si	230	10	≈140	26	0.5	13.00
GSO-9A	ITO-coated Si	≈450	10	≈140	48	0.36	17.28
GSO-9B	bare Si	> 450	10	≈140	60	0.24	14.40
GSO-10	bare Si	≈650	10	≈140	20	0.9	18.00

Table 3.1: Table of GSO film parameters and results. Where T_{sub} is substrate temperature, d is film thickness, ϵ_r is the dielectric constant and E_{break} is the breakdown field strength.

Name	Value
ID	GSO-10
Material	$GdScO_3$
Thickness	$\sim$ 140nm
Substrate	Base Si
Substrate Temp	$\sim$ 650°C
O_2 pp	10 mtorr
Dot Area	0.011 cm^2

Table 3.2: GSO-10 Film and Deposition Parameters

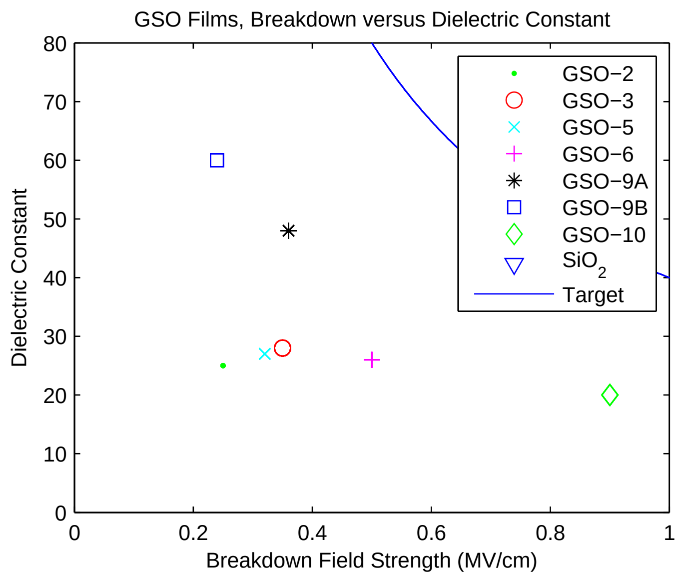

Figure 3.2: Plot of breakdown versus dielectric constant for all GSO films, close view.

electric constant as a function of frequency. This uses the simple two element model at every point in the data set and neglects contact resistance. The data in this form are given in Figures 3.3 and 3.4. The breakdown measurement is a separate measurement from the impedance analysis measurements and is shown in Figure 3.5. The conductance and dielectric constant as functions of frequency form the input of the curve fitting process to determine the correct parameters, taking into account contact resistance.

3.2.3 Results

Using the data in the previous section as input, the data were curve fit against the three element model with contact resistance. The dielectric constant and other small signal results are given in Table 3.3. These values were determined by using capacitance and conductances values given by the impedance analyzer to calculate the overall impedance of the film and fitting this impedance to a three element model including a contact resistance. The breakdown field strengths are given in Table 3.4. These were obtained directly from the breakdown measurement apparatus. A scatter plot combining the dielectric constant and breakdown strength is given in 3.6. A representa-

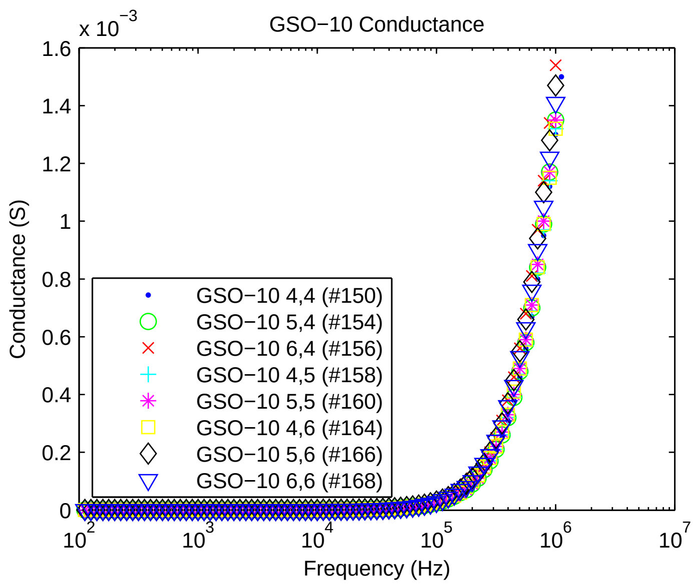

Figure 3.3:]
The conductance data for GSO-10 given by the impedance analyzer, without curve fitting.

tive plot of real versus imaginary impedance from one sample point is shown in Figure 3.7.

The GSO-10 film showed the highest breakdown field strength of any of GSO series of films and was our most successful attempt. Over the dots tested, the breakdown voltage[1] was 0.91 ± 0.37MV/cm. The dielectric constant was 20.0 ± 0.6. Multiplying this together to give an estimate of the quality of the insulator yields 18.2 ± 7.40MV/cm, whereas the same measure for SiO_2 is ~ 40MV/cm; although this film was one of the best of the GSO series, it is not a good enough insulator to be competitive with silicon.

3.3 MTO Results

We also performed a single measurement run on a magnesium tin oxide sample using the same procedure. This gave the results given in Tables 3.5 and 3.6. The raw breakdown data is shown in Figure 3.8. Note that unlike the

[1]Temporary Note: I've calculated the error bars as 1 standard deviation here. There may be a better metric for this.

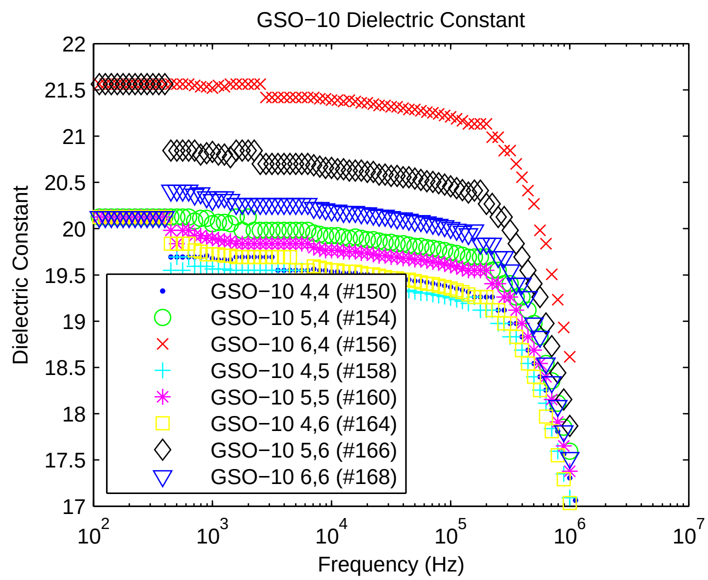

Figure 3.4:]
The dielectric constant data for GSO-10 from the impedance analyzer, without curve fitting.

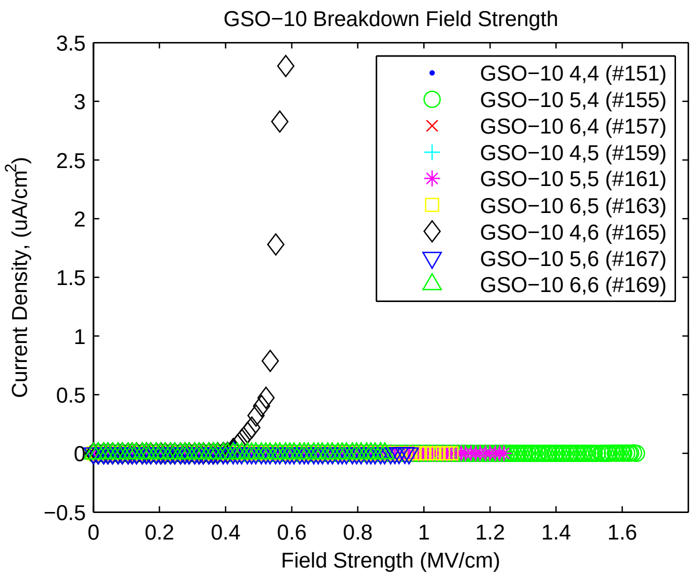

Figure 3.5: Data from breakdown measurement on GSO-10 sample.

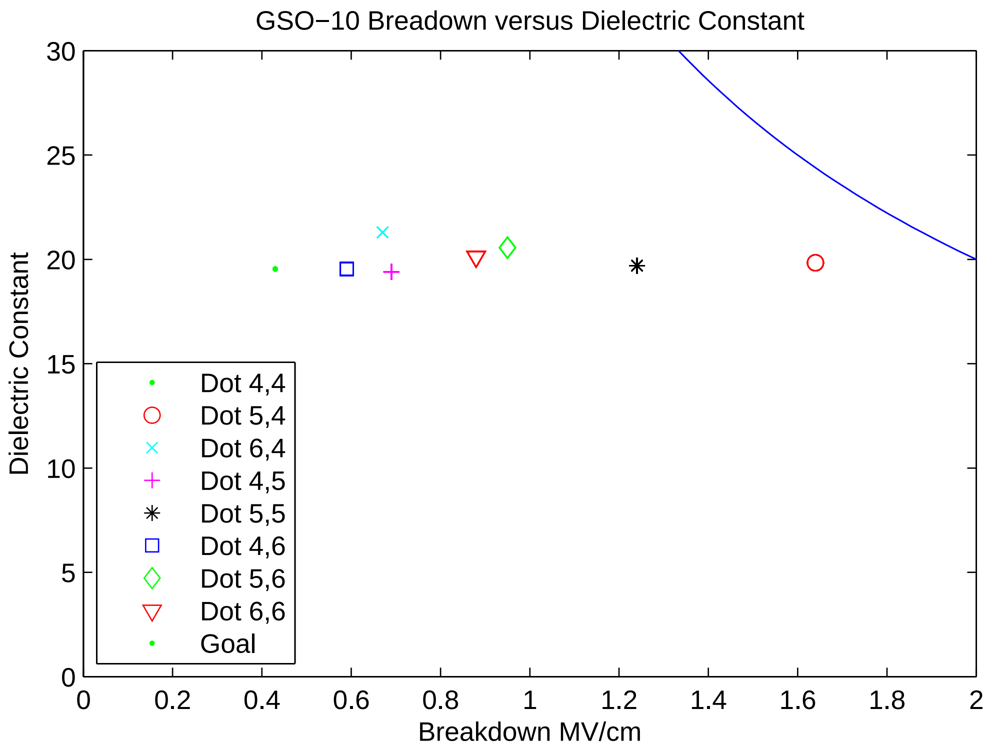

Figure 3.6: Plot of GSO-10 breakdown versus dielectric constant

Run#	Position	R (MΩ)	C (nF)	R_c (Ω)	RMSE (Ω)	ϵ_r	σ (nS/m)
150	4,4	1.69	1.36	30.7	10.2	19.5	75.2
154	5,4	1.60	1.38	32.6	7.2	19.8	79.4
156	6,4	1.47	1.48	32.9	9.7	21.3	86.7
158	4,5	1.79	1.35	33.5	8.3	19.4	71.1
160	5,5	1.95	1.37	33.5	6.1	19.7	65.2
164	4,6	1.43	1.36	34.3	11.3	19.5	89.2
166	5,6	1.44	1.43	35.5	9.7	20.6	88.2
168	6,6	1.84	1.40	36.1	6.8	20.1	69.1
Mean		1.65	1.39	33.7		20.0	78.0
St. Dev.		0.20	0.04	1.7		0.6	9.3

Table 3.3: Results of curve fitting GSO-10 small-signal data to the three element RC parallel model with contact resistance.

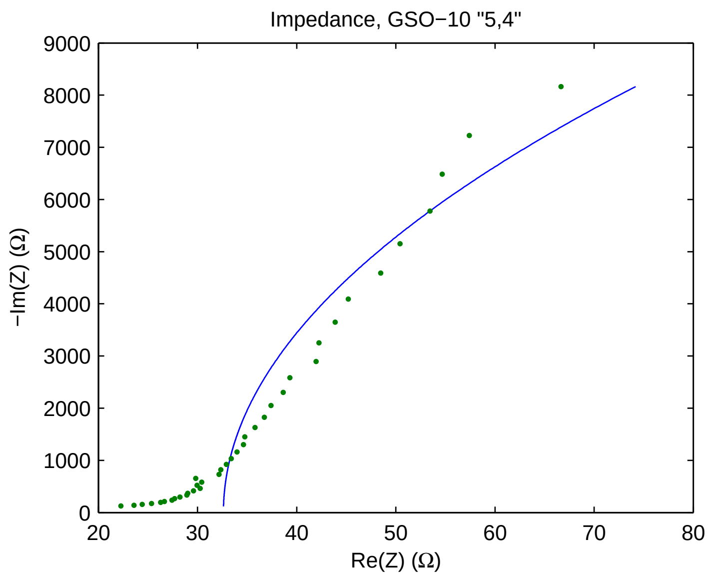

Figure 3.7: Impedance plot of the 5,4 dot on GSO-10

Run#	Position	Breakdown (MV/cm)
151	4,4	0.43
155	5,4	1.64
157	6,4	0.67
159	4,5	0.69
161	5,5	1.24
163	6,5	1.08
165	4,6	0.59
167	5,6	0.95
169	6,6	0.88
Average		0.91
St. Dev.		0.37

Table 3.4: GSO-10 Breakdown Results

Run#	Position	R (MΩ)	C (nF)	R_c (Ω)	RMSE (Ω)	ϵ_r	σ (nS/m)
209	4,5	1.10	0.49	21.1	6.5	18.2	299.0
211	5,5	1.30	0.46	22.0	6.7	17.0	253.3
213	6,5	1.28	0.44	22.7	6.5	16.5	258.2
215	7,5	1.25	0.42	23.7	7.6	15.8	265.0
217	8,5	1.14	0.44	24.0	7.6	16.5	288.4
219	5,6	1.06	0.48	20.2	7.5	17.9	312.6
221	6,6	1.09	0.46	21.3	7.6	17.1	301.8
Mean		1.17	0.46	22.2		17.0	282.6
St. Dev.		0.10	0.02	1.4		0.8	23.6

Table 3.5: MTO-1 Small Signal Results

GSO-10 sample shown earlier, this sample shows poor breakdown dielectric behavior by allowing significant leakage current even at small field strengths rather than very small current followed by an abrupt breakdown. As such the breakdown numbers quoted in Table 3.6 indicate the point where the sample passed the programmed threshold current rather then a true breakdown. The small signal dielectric constant and conductance values are likewise shown in Figures 3.9 and 3.10. Because of the poor breakdown characteristic, this material is not suitable as dielectric.

Run#	Position	Breakdown (MV/cm)
202	4,4	0.11
204	5,4	0.13
206	6,4	0.17
208	7,4	0.11
210	4,5	0.10
212	5,5	0.13
214	6,5	0.15
216	7,5	0.16
218	8,5	0.16
220	5,6	0.12
222	6,6	0.14
Average		0.13
St. Dev.		0.02

Table 3.6: MTO-1 Breakdown results.

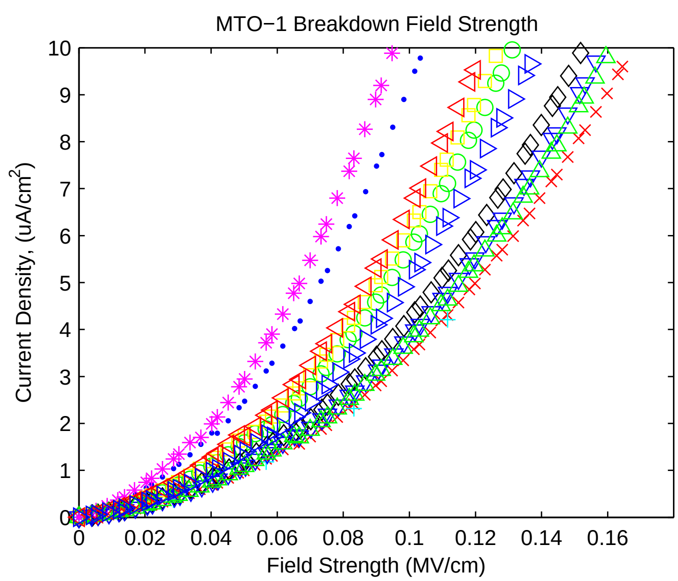

Figure 3.8: Breakdown data for MTO-1 showing poor dielectric behavior. Different symbols denote different sample points. The legend has been omitted to show the results more clearly.

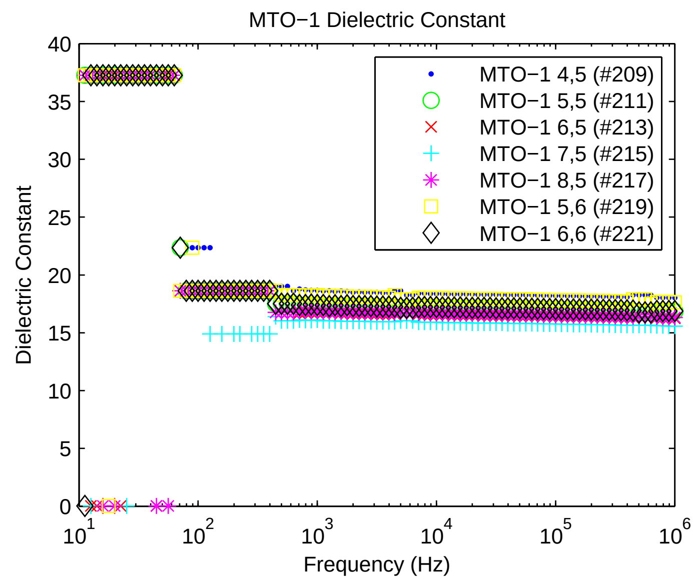

Figure 3.9: Raw dielectric constant data from MTO-1 sample.

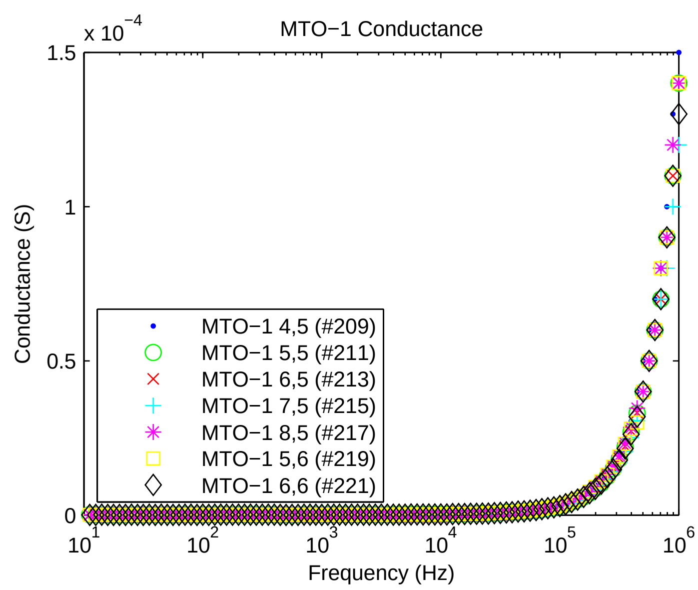

Figure 3.10: Raw conductance data from MTO-1 sample.

Chapter 4

Conclusion

In the preceding chapters we have analyzed the properties of a good dielectric material with an emphasis on applications as a gate dielectric. We have introduced the concepts and theory behind impedance spectroscopy as a method to determine dielectric properties of materials and developed several different circuit element models for representing a thin film dielectric material. We also derived the impedances of these models in order to compare them with experimental results and allow curve fitting. We also used impedance spectroscopy and these results to measure dielectric properties of samples of several materials. We also measured dielectric breakdown strength and combined these results to arrive at figure of merit. Most significantly, we measured a series of samples of gadolinium scandate oxide films prepared under different conditions and measured their dielectric properties. We found that our highest-performing sample had a dielectric constant of approximately 20 and a breakdown strength of 0.9 MV/cm, resulting in a figure of merit of 18 MV/cm. For comparison, SiO_2 has a figure of merit of 40MV/cm. Thus we found that gadolinium scandate oxide does not perform well enough to compete with existing dielectric materials. We also performed a single sample measurement on a magnesium tin oxide sample. However this sample did not show dielectric breakdown behavior, instead allowing conduction at any field strength, and thus was not suitable for dielectric applications.

4.1 Acknowledgments

This work was funded by URISC grant from the Oregon State University Research Office. The project was conducted under the supervision of Professor Janet Tate. Samples were produced in Prof. Tate's labs by Paul Newhouse. Testing and characterization were done in Prof. John Wager's lab. Some sample processing and preparation were done by Stephen Meyers. The author received training and advice in impedance spectroscopy from Daniel Heineck. Thanks to Jennifer Moser for proofreading and feedback on this document.

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