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November 8, 2019

# Assessing water content in polymers using capacitive sensors: Impact of the water distribution

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**Abstract**— Capacitance analysis has been used as a sensing technique to evaluate moisture content in polymers. This approach relies on the dielectric constant dependence upon free water concentration. In this work, we anticipate that the way water is distributed inside polymer pellets may change the apparent permittivity, even though the total amount of water is unchanged. This bias roots in the non-linear dependence of the dielectric constant upon water concentration. In this context, an analytical model of diffusion in cylindrical body geometry is used to predict the relative error generated when performing current-voltage (CV) experiments.

**Keywords:** *polymer, capacitance, dielectric constant, diffusion, moisture, non-linearity.*

## I. INTRODUCTION

Moisture content in the polymer may lead to hydrolysis which affects the processability of the polymer, resulting in degraded mechanical properties and, consequently, in the of quality product. [1-6] The amount of moisture that a polymer can absorb depends upon its chemistry and the atmospheric conditions to which it is exposed. The drying time before processing required to avoid that the performance of the polymers is affected by the presence of moisture (maximum level is globally considered as 200ppm) also depends on the nature of the polymer [1-5]. In terms of temperature, polymers should be heated enough to let water molecules diffuse from the polymer chains and be transported to the free surface. To determine the moisture content accurately, the thermogravimetric method based on the weight loss of the polymer pellets that is measured before and after the evaporation of the moisture is the most commonly used. This kind of moisture analyzer uses a drying oven and a high precision balance [6]. However, for in-line measurements, indirect methods are preferred to calculate the material's moisture content. Among them, the capacitive method is the most common since it is cheap and easy to handle. In the capacitive approach, the measurement relies on the variation of the polymer's dielectric constant located between the sensing capacitor electrodes upon moisture content.

Reference [1] describes a high precision capacitive moisture sensor for polymers, which is part of a compact, fast, and economic moisture analyzer that reaches a precision of 1ppm. As discussed by Barrettino et al. [6], the sensing principle used to measure the moisture present within polymers is based on the variation of the capacitance arising from a variation of the polymers intrinsic dielectric properties. The electric field generated by the sensor's electrodes causes an electric displacement within the polymers generating a charge density on the sensor's electrodes. Since moisture (water) is a highly

polar compound and presents a very high dielectric constant ( $\epsilon_r \sim 80$ ), the presence of moisture within polymers will change the electrical charge on the electrodes, which can be detected in terms of change in the capacitance [6-10]. In Ref [11] the authors demonstrated that the importance of the orientation of the pellets in the accuracy of the measurements. Giving continuity to that work, in this paper we will demonstrate that, if the water is not distributed uniformly inside the polymer, and if the relationship between water concentration and dielectric constant is not linear, errors in the apparent dielectric constant could set the lower limit in terms of sensitivity. In following sections, we propose to model the diffusion of water in a single PA66 pellet and fix an upper limit to the non-linear dependence of dielectric constant, if any, with water concentration used to evaluate the capacitive sensor accuracy.

## II. MODELING

### 1) Modeling diffusion process in cylindrical coordinates.

To model the distribution of water in a polymer pellet, we adapt the solution developed in ref [12] dealing with diffusion in cylindrical entities. According to the Fick's second diffusion law, the concentration of water as the function of the time satisfies the differential equation:

$$\frac{\partial c}{\partial t} = D \nabla^2 \quad (1)$$

Where  $c$  is the concentration of water in our case,  $D$  is the diffusivity and  $t$  the diffusion time. In ref [12], equation (1) was solved in cylindrical coordinates with a constant surface concentration  $c(t; 0) = c_\infty$  and  $c(0; x) = 0$  for the initial condition. The solution was expressed in terms of Bessel function series that links the concentration with respect to time and radius:

$$\frac{c(t;r)}{c_\infty} = 1 - \sum_{m=1}^{\infty} \left[ \frac{2}{\alpha_m J_1(\alpha_m)} J_0\left(\alpha_m \frac{r}{r_a}\right) e^{-(\alpha_m \frac{\sqrt{Dt}}{r_a})^2} \right] \quad (2)$$

We will rely on this solution to study how the distributed water inside the polymer pellet may change the apparent permittivity *even though the total amount of water is constant.*

### 2) Polymer pellet with initial moisture content

As mentioned in the beginning of this paper, in practical cases, the polymer pellets must be dried before processing. To still use the derivation in ref [12] we define a new variable for the concentration that still verifies equation (1) with the limited conditions cited above:

$$c' = c_0 - c \quad (3)$$

Where  $c$  is the real water concentration in the polymer and  $c_0$

its initial water concentration. It is straightforward to see that  $c'$  still satisfies equation (1) with the same limit conditions as in the previous section and where  $c_\infty$  is 'written'  $c_0$ , meaning that the general solution is now:

$$\frac{c(t,r)}{c_0} = \sum_{m=1}^{\infty} \left[ \frac{2}{\alpha_m J_1(\alpha_m)} J_0\left(\alpha_m \frac{r}{r_a}\right) e^{-(\alpha_m \frac{\sqrt{Dt}}{r_a})^2} \right] \quad (4)$$

Assuming a water diffusion coefficient  $D=10^{-12}$  cm<sup>2</sup>/s [12] and a polymer rod cylinder with  $r_a = 2.5$  mm, the normalized water concentration  $c(t,r)/c_0$  versus the normalized cylinder radius  $(r/r_a)$  is plotted at different times in Figure 1.

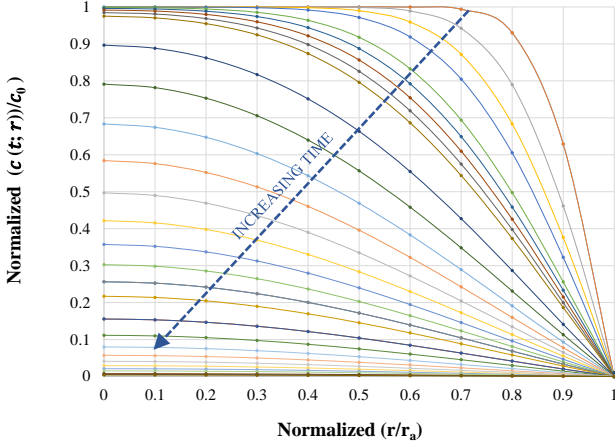


Fig. 1 Normalized water concentration versus normalized radius  $r/r_a$  for different normalized times  $\sqrt{Dt}/r_a$ .

Each line corresponds to a given time which varies from 10 to 1800 hours. The same numerical results can be plotted as function of the water concentration versus  $\sqrt{Dt}/r_a$  for different radius ratio  $r/r_a$  as shown in Figure 2.

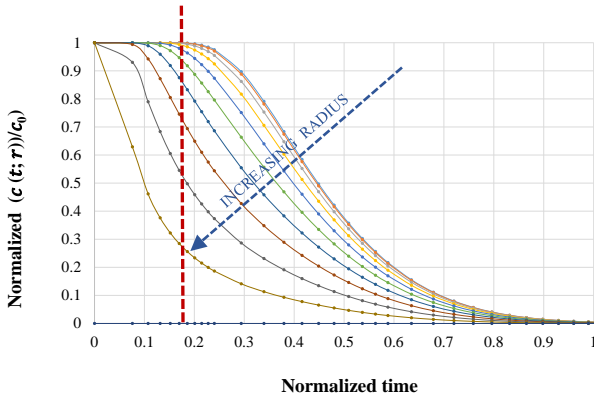


Fig. 2 Normalized water concentration versus normalized time  $\sqrt{Dt}/r_a$  for different normalized radius  $r/r_a$ .

Based on this preliminary study, we can conclude that the non-uniform distribution of water implies that *the dielectric constant will change along the radius*, and will change with time. Given that the capacitance accuracy required by moisture capacitive sensors is a few femto Farads, we will prove that neglecting how water is distributed inside the cylinder is

responsible for intrinsic errors which cannot be detected.

### 3) Modeling induced capacitance in presence of non-linear dielectric constant in a polymer rod.

In order to calculate the effective capacitance during the pellet drying process versus time, a cylindrical polymer pellet with the same length and diameter (namely  $2R$ ) is placed between two parallel plates with dimensions large enough to neglect the fringing fields as shown in Figure 3. For simplicity, the polymer rod is oriented perpendicular to the electric field.

Assuming an ideal capacitor comprising a polymer cylinder, according to the previous discussion, the dielectric constant  $\varepsilon(r)$  will vary with radius  $r$  and the capacitance can be written as:

$$C_T = \int_0^R 2\pi \cdot \varepsilon(r) \cdot \frac{r}{2R} \cdot dr = \int_0^R \pi \cdot \varepsilon(r) \cdot \frac{r}{R} \cdot dr \quad (5)$$

After normalization to the external radius  $R$  we obtain:

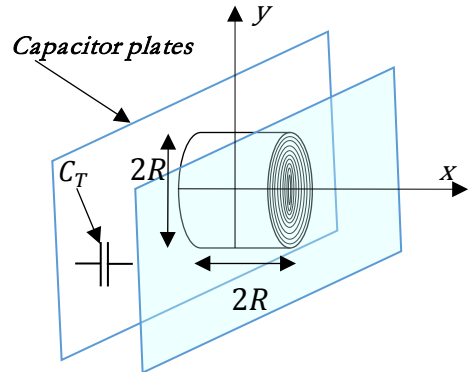


Fig. 3 Representation of the capacitor with the pellet oriented parallel to the electric field

$$C_T = \pi \cdot R \int_0^1 \varepsilon(x) \cdot x \cdot dx \quad (6)$$

Relation (6) is the total capacitance taking into account the non-uniform distribution of water along the radius as given by (4). Keeping the analysis generic, we consider that the dielectric constant can be trunked to the second order term of a polynomial expression, that is:

$$\varepsilon(H_2O) = \varepsilon^0 + a[H_2O] + b[H_2O]^2 \quad (7)$$

Where  $\varepsilon^0$  is the dielectric constant of the polymer without water. Inserting (7) in (6), the total capacitance writes:

$$C_T = \pi R \left\{ \int \varepsilon^0 x dx + a \int [H_2O(x)] x dx + b \int [H_2O(x)]^2 x dx \right\} \quad (8)$$

The total amount of water  $Q_{real}$  in the cylinder is:

$$Q_{real} = 2R \int_0^R [H_2O(r)] 2\pi r dr = 4\pi R^3 \int_0^1 [H_2O(x)] x dx \quad (9)$$

Replacing (9) in the second term of (8) gives:

$$C_T = \pi R \left\{ \int \varepsilon^0 x dx + \frac{a}{4\pi R^3} Q_{real} + b \int [H_2O(x)]^2 x dx \right\} \quad (10)$$

As in relation (4), we introduce the normalized concentration  $c(t,r)/c_0$ :

$$C_T = \pi R \left\{ \frac{\varepsilon^0}{2} + \frac{a}{4\pi R^3} Q_{real} + b c_0^2 \int \left( \frac{c(t,r)}{c_0} \right)^2 x dx \right\} \quad (11)$$

However, when measuring the capacitance, one would merely use relation (7) to get the *apparent* concentration of water

$[H_2O]_{app}$ , which means:

$$C_T = \frac{\varepsilon \pi R^2}{2R} = \frac{\pi R}{2} \varepsilon = \frac{\pi R}{2} \left\{ \varepsilon^0 + a[H_2O]_{app} + b[H_2O]_{app}^2 \right\} \quad (12)$$

#### 4) Error induced by on-linear effects.

Obviously, relations (11) and (12) are different. This reveals that for a given measured capacitance  $C_T$ , the apparent quantity of water  $Q_{app}$  may be different from  $Q_{real}$ , and this will depend on how the water is distributed inside the pellet (last term in relation (11)), and not only on the amount of water stored  $Q_{real}$ . Relations (11) and (12) imply that the real and apparent concentrations are linked to each other as:

$$a \frac{Q_{real}}{2\pi R^3} + 2bc_0^2 \int \left( \frac{c(t;x)}{c_0} \right)^2 x dx = a[H_2O]_{app} + b[H_2O]_{app}^2 \quad (13)$$

Rearranging the terms and noting that  $2\pi R^3$  is the volume of the pellet, we have:

$$[H_2O]_{real} - [H_2O]_{app} = \frac{b}{a} \left\{ \left( \frac{Q_{app}}{2\pi R^3} \right)^2 - 2c_0^2 \int \left( \frac{c(t;x)}{c_0} \right)^2 x dx \right\} \quad (14)$$

Here we introduce an approximation consisting in replacing the global value  $Q_{app}$  by  $Q_{real}$  in (14):

$$[H_2O]_{real} - [H_2O]_{app} = \frac{b}{a} \left\{ \left( \frac{Q_{real}}{2\pi R^3} \right)^2 - 2c_0^2 \int \left( \frac{c(t;x)}{c_0} \right)^2 x dx \right\} \quad (15)$$

Given that  $Q_{real}$  is also expressed in terms of the local concentration  $c(t;x)$  in (9), we obtain:

$$[H_2O]_{real} - [H_2O]_{app} \cong \frac{b}{a} c_0^2 \left\{ \left( 2 \int \frac{c(t;x)}{c_0} x dx \right)^2 - 2 \int \left( \frac{c(t;x)}{c_0} \right)^2 x dx \right\} \quad (16)$$

Relation (16) is plotted in Figure 4 which shows the mismatch between real and apparent water concentrations versus normalized time.

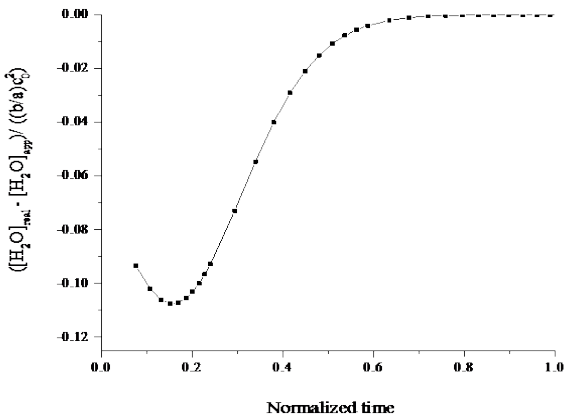


Figure 4. Mismatch between real and apparent water concentrations versus normalized time

Therefore, any non-linearity in the relationship between  $\varepsilon$  and

$[H_2O]$  (i.e. coefficient  $b$ ) could be responsible for a mismatch between the real and ‘measured’ amount of water in the polymer (note that when the water concentration is constant, the apparent and real values are equal whichever the non-linearity). This difference is proportional to the ratio  $b/a$ , to the initial water concentration  $c_0$ , and to a weighing term contained in the parenthesis. Interestingly, this term is expressed in normalized quantities and is independent of the material parameters. Its dependence with respect to the normalized time  $\sqrt{Dt}/r_a$  shown in Figure 4 reveals that it reaches a minimum around -0.11 when the normalized time is about 0.18, a worst case for the error between the real and apparent water concentrations. The total normalized water concentration at this point is found to be about 0.72. This time is shown by the red dotted line in Figure 2 where the different concentration with time inside the pellet are reported.

$$\Delta[H_2O]_{max} \cong 0.11 \frac{b}{a} c_0^2 \quad (17)$$

Assuming the water diffusion coefficient  $D=10^{-12}$  cm<sup>2</sup>/s and that the polymer rod cylinder has a radius  $r_a = 2.5$  mm, the maximum error will be generated after approximately 12 hours and 30 minutes.

#### 5) Intrinsic limitation of moisture evaluation.

According to reference [1], it is desirable to have an accuracy of 1ppm in terms of water concentration when measuring the moisture in polymers. Now, we consider that the polymer contains initially 200ppm of water, which is globally the minimum acceptable moisture content before it is submitted to processing [1]. However, since we are mainly concerned with a concentration of 50ppm which is the target in our case, this value will be reached at a normalized time of 0.5 (i.e. 34 hour 43 minutes), see figure 2. Then, according to Figure 4, the value reported on the y-axis is roughly -0.012.

In this case we would have to satisfy:

$$\Delta[H_2O]_{max} \cong 0.012 \frac{b}{a} c_0^2 \quad (18)$$

And then the ratio  $b/a$  should be lower than 2:

$$\frac{b}{a} < 2 \quad (19)$$

Figure 5 shows the variation in the total capacitance versus humidity at 20°C using Nylon-PA66 [1].

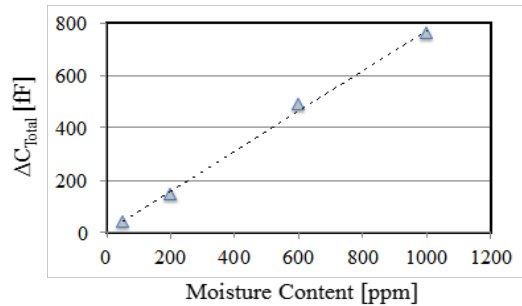


Figure 5. Capacitance variation upon moisture concentration at 20°C using Nylon-PA66.

In the work reported in [1], any capacitance change is linked to a change in the dielectric constant, meaning that we can write:

$$\Delta C_T = K \Delta \varepsilon \quad (20)$$

(K is a real number).

From equation (7), we can write:

$$\Delta \varepsilon = \varepsilon(H_2O) - \varepsilon^0 = a[H_2O] \left( 1 + \frac{b}{a}[H_2O] \right) \quad (21)$$

Which means that we have finally a relation between the variation in the measured capacitance and the apparent concentration of water:

$$\Delta C_T = K \Delta \varepsilon = K a [H_2O] \left( 1 + \frac{b}{a} [H_2O] \right) \quad (22)$$

In practice, this means that for a polymer pellet containing initially 200ppm as mentioned previously, the uncertainty would be  $\frac{b}{a}[H_2O]$ , that is  $10^{-4}$ . With respect to 1, this represents a maximum error of 0.01% when estimating the dielectric constant, or equivalently the capacitance in Figure 5. Therefore, it is very likely that any measurement of the moisture in polymers will be biased by some intrinsic error which cannot be predicted. In other words, this means that non-linear dependences, which may impede a moisture accuracy of 1ppm, could simply remain unnoticed. This looks like an intrinsic limitation in the accuracy coming from the limited knowledge of the apparent dielectric constant versus water concentration that bias its extraction to detect the moisture content. This approach could be used to verify if the moisture that is measured by the CV method gives the real total value. Note that this non linearity induced error will depend on the initial and final concentrations, as well as on the accuracy of interest. Depending on these parameters, if there is a non-linearity, it could be identified without ambiguity.

### III. CONCLUSION

We investigated how minor non-linearity in the relationship between the dielectric constant and the water concentration in polymers could settle a lower limit to the accuracy of the moisture measurement using capacitive sensors. This limitation is intimately linked to the non-uniform distribution of water during the drying process while the water content is monitored by CV experiments. The way this kind of system is treated here is quite general. The analysis could be applied to other systems exhibiting a non-linear behavior between the concentration of the parameter that we want to sense and the physical properties used to measure it while the parameter distribution is not uniform in the volume.

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