Dielectric Properties of Polymer Blends Using Least Square Method

Ahmed M. Mahmoud, Loai S. Nasrat and Abd Allah A. Ibrahim

Abstract—Poly vinyl chloride (PVC) is widely used in high and medium voltage cables insulation due to its low dielectric losses and its ability to improve cables properties in high temperatures. This paper aims to improve PVC electrical properties (dielectric strength) for high voltage and medium voltage cables in respect of mechanical characteristics by adding ethylene propylene diene monomer (EPDM). Blends of PVC with EPDM were prepared with 0%, 20%, 35%, 50%, 65%, 80%, and 100% by weight percentages concentration. The dielectric strength of the blends was tested in several conditions such as dry, wet and salty wet condition. Tensile strength test was applied to check mechanical properties of the blends. Results were analyzed using least square method in a way to determine the optimum percentage of mixing PVC/EPDM blend, which gives the best of electrical and mechanical properties under different conditions.

Index Terms—Polymers; PVC/EPDM Blend; Least Square Method; Dielectric Strength; Electrical and Mechanical Properties

I. INTRODUCTION

High-voltage rotating machines, underground cables, transformers and other electrical equipments play a significant role in generating electrical energy which faces an increasing demand for new, or the refurbishment of power stations. All these equipments have a common important and vital component, it is their electrical insulation. Due to its great importance in power system, electrical insulation has been studied and investigated for many decades however, it is the main reason behind about a quarter of all failures of electrical equipments [1]-[3].

Mainly, dielectric materials can be classified into two major types based on their nature; ceramic insulators and non-ceramic insulators. Ceramic insulators like porcelain and glass insulators. The other dielectric material which offers an alternative to the traditionally ceramic material as dielectric is polymers. Recently, polymer insulators are used increasingly due to their better characteristics over porcelain and glass types. They have many advantages like:

- Lighter weight.
- Higher impact strength.
- Good chemical stability.
- Better contamination performance due to surface hydrophobicity.

Their disadvantages like:
- Low thermal stability.
- Their dielectric constant is lower than ceramic materials.

Many studies and investigations have been done on their dielectric properties under contamination conditions[4]-[12]. A polymer can be defined as a long-chain molecule that is composed of a large number of repeating units of identical structure. Polymers could be classified according to different concepts.

According to origin or source, natural polymers and synthetic polymers. Natural polymers obtained from nature like; proteins, wool, cellulose, and silk. Synthetic polymer or man-made polymers prepared synthetically from low molecular weight compounds, includes polystyrene, polyethylene, and nylon, sometimes, natural polymers can also be produced synthetically like natural (Hevea) rubber, known as polyisoprene in its synthetic form.

All polymers can be assigned to one of two groups based upon their processing characteristics or the type of polymerization mechanism. A more specific classification can be made on the basis of polymer structure. Groupings are very important as they facilitate the understanding and discussion of properties [13].

Using composite materials, scientists can combine advantages of different materials. Recently, material scientists are looking for Nano-composites based on polymer matrix to get new advantages like, low production cost, easy processing, balanced physical and mechanical properties…etc.

A lot of scientific researches and studies have been done to enhance the optical and electrical properties of polymers through suitable doping [14]-[16].

Two different polymers are blended by specific percentages of weight to improve a specific property (electrical, mechanical, physical…etc.), this technique is called “polymer blending”.

Blending some polymers with poly vinyl chloride PVC can be done to improve chemical, physical and electrical properties as it has many advantages like good insulation resistance and high dielectric strength. It has adequate resistance to alcohols, acids, alkalies, moisture and abrasion. Also, it has flame retardant properties as it is chlorinated and low water absorption [17].

The polymer chain formed by the reaction of ethylene and propylene is called EPR. A terpolymer can be manufactured by polymerizing a diene monomer with ethylene and propylene. Therefore, ethylene propylene diene terpolymer is labeled EPDM by ASTM nomenclature. In EPDM, the “E”

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stands for ethylene, the "P" for propylene, the "D" for diene, and the "M" for the repeating CH2 units of the saturated polymer backbone [18].

Due to their stable, saturated polymer backbone structure; ethylene-propylene rubbers have a good resistance to heat, ozone, oxidation, and weather aging. This kind of polymers have good electrical resistivity and good resistance to polar solvents, such as water, alkalies, acid etc. [19].

The present work has been devoted to study dielectric strength under several conditions, and some mechanical properties of PVC/ EPDM to select the optimum blend percentage of EPDM and PVC which gives good performance as insulating material.

II. EXPERIMENTAL PROCEDURE

A. Blend Preparation.

Chemical ingredients of the materials used in the present investigation are as follows:

1. Polyvinyl chloride (PVC).
2. Ethylene Propylene Diene Monomer (EPDM rubber).

It was supplied by EGY Gates International trading S.A.E.

Seven blend samples have been prepared as: 100% EPDM, 100% PVC, 80% EPDM with 20% PVC, 65% EPDM with 35% PVC, 50% EPDM with 50% PVC, 35% EPDM with 65% PVC and 20% EPDM with 80% PVC. The mixing formulation are listed in Table I.

**TABLE I: THE MIXING FORMULATION OF EPDM WITH DIFFERENT PERCENTAGES OF PVC**

<table>
<thead>
<tr>
<th>Blend Number</th>
<th>EPDM content wt%</th>
<th>PVC content wt%</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>100%</td>
<td>0</td>
</tr>
<tr>
<td>2</td>
<td>80%</td>
<td>20%</td>
</tr>
<tr>
<td>3</td>
<td>65%</td>
<td>35%</td>
</tr>
<tr>
<td>4</td>
<td>50%</td>
<td>50%</td>
</tr>
<tr>
<td>5</td>
<td>35%</td>
<td>65%</td>
</tr>
<tr>
<td>6</td>
<td>20%</td>
<td>80%</td>
</tr>
<tr>
<td>7</td>
<td>0</td>
<td>100%</td>
</tr>
</tbody>
</table>

B. Dielectric Breakdown Strength Test.

The voltage at which current begins to flow through the insulator is defined as dielectric strength or breakdown voltage. Usually, the breakdown voltage of a polymer may be detected by an avalanche breakdown. It can be expressed in terms of voltage gradient like voltage per thickness (kV/mm) [20].

At polymers where the operation temperature is below the glass transition temperature, intense heating leads to a change of material properties during the glass transition point. For PVC; different behavior was found, but nevertheless the failure of the specimen occurs. Basically, the electrical conductivity increases rapidly with increasing temperature. Further, partial discharges occur due to different thermal expansion of insulation materials or adjacent electrodes or materials [21].

Sets of blend samples have been prepared and tested using A.C voltage. Samples are in the form of disc with 5 cm diameter and 1 mm thickness for the dielectric strength test as shown in Fig. 1 and the test is carried out according to ASTM D-149 [22].

The relation (equation) between different dielectric strength values and the content of PVC percentage in the blend was interpreted by the least square method using MATLAB.

![Fig. 1. Different percentages of EPDM/PVC Blend samples.](image)

Each sample has been tested several times to insure the results and the average is taken to minimize the error. All of the practical results have been precisely gathered and recorded in tabulated forms and then plotted to be easy for discussion and analysis.

Dielectric properties were investigated at three conditions; dry condition, wet condition and salty wet condition.

1. Dry Condition

The procedures and precautions required during the dielectric strength test are as follows:

- Test samples should be clean and dry before starting high voltage tests.
- The electrodes should be fixed into the samples, one at the top and the other on the bottom end.
- Circuit links should be correct before applying the electrical test to insure safety. Fig. 2 shows the schematic diagram which represents the dielectric strength testing circuit.
- The voltage is gradually increased at an almost constant rate of 2 kV/sec until the voltage breakdown occurs.
Test results are obtained, tabulated and discussed.

3. Salty wet Condition.
- A set of 7 specimens are used and should be placed in a container filled of a sodium chloride at room temperature, and should rest on edge for 24 hours.
- At the end of the 24 hours, all specimens should be removed from the water one at a time, all surface water wiped off with a dry cloth, and weighed to the nearest 0.001 g immediately and the weight gain is noted.
- percent increase in weight during immersion is calculated as follows:

\[
\text{Increase in weight} \quad \% = \left( \frac{\text{Wet Weight} - \text{Dry Weight}}{\text{Dry Weight}} \right) \times 100
\]

- The graph percentage weight gain with against PVC content in the blend is plotted.
- Then the samples are taken and inserted one by one between two electrodes performing the dielectric breakdown test with the same procedure as in dry condition test.
- Test results are obtained, tabulated and discussed.

C. Mechanical test
Another important concept in engineering and material science is polymer tensile strength. It can be defined as the ability of a material to withstand a tensile force. It is usually expressed in terms of force per cross-sectional area or (MPa).

The tensile strength of a material also may be defined as the maximum amount of tensile stress that a material can withstand before failure, like breaking or permanent deformation. Tensile strength specifies the point when a material goes from elastic to plastic deformation.

Mechanical performance of polymers is dependent on some factors like the molecular wt., temperature and loading rate [26],[27].

The dimensions of the sample are 5 cm length and 1 mm thickness for tensile strength in the form of dumbbell shape according to ASTM D 412 – 06a and tests are carried out also under three conditions (dry, wet, and salty wet condition) [28].

The dumbbell shaped sample is placed in the grips of the testing machine. The machine will start with grip separation rate of around 500 mm/min and the force is increased gradually till the sample rapture and the force is recorded. The tensile strength is calculated as follows:

\[
TS = \frac{F}{A}
\]

Where:
\(TS\) is the tensile strength, the stress at rupture (Mpa), \(F\) is the force magnitude at rupture (MN) and \(A\) is the cross-sectional area of unstrained specimen (m2).

III. LEAST SQUARE METHOD
Experimental data is exact about specific points but it is needed to predict intermediate values which can’t be easily carried out. A convenient method for these cases is to derive a mathematical model or a function which fits the shape of the data without the necessity of matching all points.
Least square method is used to minimize the discrepancy between the data points and the curve, which will be used in analyzing the obtained results [29].

A. Problem statement.

The main objective of the method is to adjust the parameters of a model function to find the best fit of a data set. A simple data set consists of n points \((x_i, y_i), i = 1, ..., n\), where \(x_i\) is an independent variable and \(y_i\) is a dependent variable.

A regression line is defined as a straight line which describes how a dependent variable \(y\) changes when an independent variable \(x\) changes. It is used to predict the value of \(y\) for a given value of \(x\) [30].

If this fit is a straight line, then

\[ f(x) = ax + b \]  

(1)

\[ Error = \sum_{i=1}^{n} d_i^2 = [y_1 - f(x_1)]^2 + [y_2 - f(x_2)]^2 + [y_3 - f(x_3)]^2 + [y_4 - f(x_4)]^2 \]  

(2)

\[ Error = \sum_{i=1}^{n} y_i - f(x_i) = \sum_{i=1}^{n} (y_i - (ax_i + b))^2 \]  

(3)

The ‘best’ line has minimum error between line and data points, this is called the least squares approach, since square of error is minimized.

\[ \text{Minimize} \ \{Error\} = \sum_{i=1}^{n} [y_i - (ax_i + b)]^2 \]  

(4)

Taking the derivative of the error with respect to \(a\), set the equation to zero

\[ \frac{\partial (Error)}{\partial a} = \frac{\partial \left( \sum_{i=1}^{n} [y_i - (ax_i + b)]^2 \right)}{\partial a} = 0 \]  

(5)

\[ \frac{\partial (Error)}{\partial a} = -2 \left( \sum_{i=1}^{n} x_i [y_i - (ax_i + b)] \right) = 0 \]  

(6)

Making the same with respect to \(b\)

\[ \frac{\partial (Error)}{\partial b} = \frac{\partial \left( \sum_{i=1}^{n} [y_i - (ax_i + b)]^2 \right)}{\partial b} = 0 \]  

(7)

\[ \frac{\partial (Error)}{\partial b} = -2 \left( \sum_{i=1}^{n} [y_i - (ax_i + b)] \right) = 0 \]  

(8)

Solve for the \(a\) and \(b\) so that, (6) and (8) both be 0

\[ a \sum_{i=1}^{n} x_i + b \sum_{i=1}^{n} x_i^2 = \sum_{i=1}^{n} x_i y_i \]  

(9)

\[ a \sum_{i=1}^{n} x_i + b n = \sum_{i=1}^{n} y_i \]  

(10)

Put these into matrix form

\[ \begin{bmatrix} \sum_{i=1}^{n} x_i^2 & \sum_{i=1}^{n} x_i \\ \sum_{i=1}^{n} x_i & n \end{bmatrix} \begin{bmatrix} a \\ b \end{bmatrix} = \begin{bmatrix} \sum_{i=1}^{n} x_i y_i \\ \sum_{i=1}^{n} y_i \end{bmatrix} \]  

(11)

\[ a = \frac{N \sum_{i=1}^{n} x_i y_i - \sum_{i=1}^{n} x_i \sum_{i=1}^{n} y_i}{N \sum_{i=1}^{n} x_i^2 - \left( \sum_{i=1}^{n} x_i \right)^2} \]  

(12)

\[ b = \frac{\sum_{i=1}^{n} x_i^2 \sum_{i=1}^{n} y_i - \sum_{i=1}^{n} x_i \sum_{i=1}^{n} x_i y_i}{N \sum_{i=1}^{n} x_i^2 - \left( \sum_{i=1}^{n} x_i \right)^2} \]  

(13)

B. Polynomial Regression.

Representing engineering data by a line isn’t convenient for many cases so, it would be preferable to fit the data by a curve using polynomial regression.

The least square procedure can be extended to fit the data to a higher-order polynomial. So, fitting a second-order polynomial as [30]:

\[ y = a_0 + a_1 x + a_2 x^2 + e \]  

(14)

\[ Error = \sum_{i=1}^{n} d_i^2 = \sum_{i=1}^{n} (y_i - a_0 - a_1 x_i - a_2 x_i^2)^2 \]  

(15)

Applying the same procedure of the linear regression, taking the derivative of (15) with respect to each of unknown coefficients of the polynomial \(a_0, a_1\) and \(a_2\). The equations are then set equal to zero, with all summations from \(i=1\) through \(n\):

\[ AX = B \]  

(16)

\[ X = A^{-1} * B \]  

(17)

Where:

\[ A = \begin{bmatrix} n & \sum_{i=1}^{n} x_i & \sum_{i=1}^{n} x_i^2 \\ \sum_{i=1}^{n} x_i & \sum_{i=1}^{n} x_i^2 & \sum_{i=1}^{n} x_i^3 \\ \sum_{i=1}^{n} x_i^2 & \sum_{i=1}^{n} x_i^3 & \sum_{i=1}^{n} x_i^4 \end{bmatrix} \]

(18)

The coefficients: \(a_0, a_1\) and \(a_2\), can be then easily calculated from the measured data. The two-dimensional case can be easily extended to an \(m\)-th order polynomial as:

\[ y = a_0 + a_1 x + a_2 x^2 + \cdots + a_m x^m + e \]  

(19)

C. Coefficient of determination (R2).

Coefficient of determination (\(R^2\)) is the square of multiple correlation coefficient between \(y\) and \(x_1, x_2, \ldots, x_n\).

Its value describes how well the regression line fits to the actual measured data. Which is considered a measure of goodness of the fitting for the model.

Then

\[ R^2 = 1 - \frac{SS_{res}}{SS_T} \]  

Where \(R^2\) is the Coefficient of determination, \(SS_{res}\) is the sum of squares due to residuals and \(SS_T\) is the total sum of squares [31].

IV. RESULTS AND ANALYSIS

A. Electrical Results

1. Dry Condition Calculations

The samples are tested in a dry condition. The results are taken to be a reference in case of wet and salty wet tests.
Fig. 4 shows that pure PVC (7th sample) has the maximum value of dielectric strength 34.12 kV/mm. While pure EPDM (first sample) has the minimum value of dielectric strength 12.91 kV/mm. The blend samples have values in between.

Comparing by the 20% blend sample; It can be seen that, increasing the amount of PVC in the blend by 35%, 50%, 65% and 80% increases the dielectric strength of the blend samples by 32.68%, 52.05%, 61.07% and 69.86% respectively. As the amount of PVC increases, the dielectric strength increase.

Applying least square criterion using Matlab program (curve fitting tool box), the best curve fitting for the obtained results from the test is shown in Fig. 5. Representing the data by a 4th degree polynomial equation to minimize the error as possible:

\[ y = a_1 x^4 + a_2 x^3 + a_3 x^2 + a_4 x + a_5 \]  

Where:
- \( y \) is the dielectric strength under dry condition value,
- \( x \) is the percentage of PVC in blend,
- \( a_1 \) is a constant = 83.438,
- \( a_2 \) is a constant = -167.69,
- \( a_3 \) is a constant = 82.131,
- \( a_4 \) is a constant = 23.442,
- \( a_5 \) is a constant = 12.855 and \( R^2 = 0.9984 \)

Applying least square criteria for the two cases of porosity test:
1. Fig. 6 and (22) representing the percentage weight gain under first condition (immersion in distilled water).
2. Fig. 7 and (23) representing the percentage weight gain under second condition (immersion in NaCl Solution).

### Table II: The Weight of the Samples for All Conditions

<table>
<thead>
<tr>
<th>PVC Content in the blend (%)</th>
<th>Weight (gm) Dry</th>
<th>Weight (gm) After immersion in Water</th>
<th>Weight (gm) After immersion in NaCl Solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>5.31</td>
<td>5.89</td>
<td>5.97</td>
</tr>
<tr>
<td>20</td>
<td>5.12</td>
<td>5.58</td>
<td>5.65</td>
</tr>
<tr>
<td>35</td>
<td>4.88</td>
<td>5.21</td>
<td>5.29</td>
</tr>
<tr>
<td>50</td>
<td>4.31</td>
<td>4.55</td>
<td>4.62</td>
</tr>
<tr>
<td>65</td>
<td>5.28</td>
<td>5.49</td>
<td>5.56</td>
</tr>
<tr>
<td>80</td>
<td>5.45</td>
<td>5.55</td>
<td>5.68</td>
</tr>
<tr>
<td>100</td>
<td>4.34</td>
<td>4.36</td>
<td>4.42</td>
</tr>
</tbody>
</table>

2. Porosity Test Results

The studies carried out in this work lead to know the effect of water on dielectric strength of blend samples. This effect can be clearly illustrated from the results tabulated in Table II which includes the two cases of porosity test (wet condition and salty wet condition).
Fig. 7. Curve fitting results for the weight gain of blend samples under salty wet condition.

\[ y = a_1 x^4 + a_2 x^3 + a_3 x^2 + a_4 x + a_5 \]  

Where:
- \( y \) is the percentage weight gain value after immersion in NaCl solution,
- \( x \) is the percentage of PVC in blend,
- \( a_1 \) is a constant = \(-1.4273 \times 10^{-07}\),
- \( a_2 \) is a constant = \(2.6679 \times 10^{-5}\),
- \( a_3 \) is a constant = \(-0.0014243\),
- \( a_4 \) is a constant = \(-0.08743\),
- \( a_5 \) is a constant = 12.433 and
- \( R^2 = 0.9983 \)

3. Wet Condition Dielectric Strength Calculations

After immersion in a container of distilled water for 24 hours as a simulation of wet weather and rains the samples are tested for dielectric strength.

Fig. 8 shows that pure PVC (7th sample) has the maximum value of dielectric strength 28.44 kV/mm. While pure EPDM (first sample) has the minimum value of dielectric strength 9.21 kV/mm. The blend samples have values in between.

Comparing by 20% blend sample; It can be seen that, Increasing the amount of PVC in the blend by 35, 50, 65 and 80% increases the dielectric strength of the blend samples by 30.79%, 51.72%, 60.78% and 67.80% respectively.

Applying least square criterion using Matlab program (curve fitting tool box), the best curve fitting for the obtained results from the test is shown in Fig. 9. Representing the data by a 4th degree polynomial equation to minimize the error as possible:

\[ y = a_1 x^4 + a_2 x^3 + a_3 x^2 + a_4 x + a_5 \]  

Where:
- \( y \) is the dielectric strength under dry condition value,
- \( x \) is the percentage of PVC in the blend,
- \( a_1 \) is a constant = \(5.3001 \times 10^{-07}\),
- \( a_2 \) is a constant = \(-9.7681 \times 10^{-5}\),
- \( a_3 \) is a constant = \(0.0030937\),
- \( a_4 \) is a constant = \(0.33017\),
- \( a_5 \) is a constant = 9.1911 and
- \( R^2 = 0.9996 \)

4. Salty wet condition dielectric strength calculations

As a simulation of coast countries and areas near seas, samples are immersed in a container of NaCl solution for 24 hours and then tested for dielectric strength.

Fig. 10 shows that pure PVC (7th sample) has the maximum value of dielectric strength 21.88 kV/mm. While pure EPDM (first sample) has the minimum value of dielectric strength 6.24 kV/mm. The blend samples have values in between.

Comparing by 20% blend sample; It can be seen that, Increasing the amount of PVC in the blend by 35, 50, 65 and 80% increases the dielectric strength of the blend samples by 23.4%, 47.42%, 73.84% and 79.18% respectively.

Applying least square criterion using Matlab program (curve fitting tool box), the best curve fitting for the obtained results from the test is shown in Fig. 11. Representing the data by a 4th degree polynomial equation to minimize the error as possible:

\[ y = a_1 x^4 + a_2 x^3 + a_3 x^2 + a_4 x + a_5 \]  

\[ y = a_1 x^4 + a_2 x^3 + a_3 x^2 + a_4 x + a_5 \]
B. Mechanical Results

Tensile strength tests (TS) are carried out in order to illustrate the ability of the blend samples to withstand the mechanical forces.

<table>
<thead>
<tr>
<th>PVC Content in the blend (%)</th>
<th>TS under Dry condition (MPa)</th>
<th>TS under wet condition (MPa)</th>
<th>TS under salty wet condition (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>7.2</td>
<td>6.1</td>
<td>5.2</td>
</tr>
<tr>
<td>20</td>
<td>5.9</td>
<td>5.0</td>
<td>4.5</td>
</tr>
<tr>
<td>35</td>
<td>4.5</td>
<td>3.9</td>
<td>3.4</td>
</tr>
<tr>
<td>50</td>
<td>3.8</td>
<td>3.1</td>
<td>2.6</td>
</tr>
<tr>
<td>65</td>
<td>2.4</td>
<td>1.9</td>
<td>1.3</td>
</tr>
<tr>
<td>80</td>
<td>1.9</td>
<td>1.2</td>
<td>0.8</td>
</tr>
<tr>
<td>100</td>
<td>1.2</td>
<td>0.7</td>
<td>0.3</td>
</tr>
</tbody>
</table>

V. Conclusion

It can be concluded that,

1. The dielectric strength was improved by increasing of PVC percentage in the blends. [80%PVC,20%EPDM] blend sample had the highest dielectric strength of:
   - 32.8 kV/mm (69.86% improvement) under dry condition,
   - 27.25 kV/mm (67.80% improvement) under wet condition and
   - 20.14 kV/mm (79.18% improvement) under salty wet condition.

2. The porosity test showed that increasing the content of EPDM in the blend samples led to more water absorption which decreased the dielectric strength of the blend.

3. Increasing the content of EPDM in the blend samples enhanced the mechanical properties of the blend.

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