Characterization and Dielectric Properties of Fly Ash / Polystyrene / Low Density Polyethylene Composites

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Abstract: Polystyrene (PS) and its blend with low density polyethylene (LDPE) were prepared by melting method. Fly ash was used as filler with different contents to form composites by the same way with PS/LDPE 80/20 blend. The obtained data of fly ash from X-Ray Fluorescence analysis displayed that the total mass summation of silicon, magnetite and aluminum (SiO$_2$ + Fe$_2$O$_3$ + Al$_2$O$_3$) is about 79.52 wt. % whereas the amount of calcium oxide is less than 10%. TGA curves showed that the sample of the more thermal stability was the composite that contains the highest amount of fly ash (50 wt.%). For all the samples, the dielectric constant and the dielectric loss were increasing by decreasing the applied frequencies while they were decreasing by increasing the temperatures. Also, increasing the fly ash content in the blend improved the ac conductivity up to a content of 40 wt.% then it decreased. The values of the real part of the electrical modulus (M') did not equal to zero at low frequencies and high temperature. Therefore, it is expected that the electrode polarization may take place for the all samples.

1-Introduction

Few decades ago, polymers and polymer-based materials have been used for various electrical applications. Numerous studies have concerned on the improvement of the polymeric material with low and high dielectric constant. For instance, polymers with high dielectric constant have the ability to be applied as artificial muscle and film capacitors. However, those with low dielectric constants can be utilized in the field of capacitors and dielectric materials [1-3]. Polystyrene (PS) is a commercial thermoplastic polymer. It is rather brittle, clear and has good mechanical properties and a low cost price. Thus, PS has a wide range of applications as construction materials, packaging, disposable cups, consumer electronics, cassette boxes, compact disks and medical uses [4, 5]. This study includes also another polymer which is the low density polyethylene (LDPE). It is nominated to be widely used in different applications as using it in the industries of cables and wires. This is a consequence of its high resistivity and breakdown strength. LDPE was used in some other applications as packaging film, decks and docks, window frames, tubes and also in the industry of the automobile [6].

A great interest of research was paid for the polymer matrix composite which has unusual modified physical properties and variant industrial applications [7-10]. The physical/chemical properties of the hosting polymer and the filler can affect greatly the properties of their composites [11]. Nowadays, a great attention has been paid for developing the electrical conductivity and the dielectric properties of polymer composites. Therefore, variant kinds of fillers have been employed for improving the conductive ability of the polymeric materials. Developing the physical properties of the polymers is produced from the amalgamation of fillers with them [12].
In fact, using fillers is preferable for variant applications in order to lower the high cost of some hosting polymers and modifying their properties [6]. The effects of fly ash, as fillers, on the physical properties of the polymer materials were examined widely in many literatures [13-16]. Day by day, the production of fly ash has increased which is considered as a waste that is formed from the thermal power plants. Fly ash has become one of the most popular wastes which are used as industrial raw material. The production of fly ash resulted in many environmental and economical problems. Therefore, the researchers work on recycling it in different industrial aspects [17]. Fly ash has possibly included in diverse applications as using it in manufacturing of ash bricks, metal recovery, water and atmospheric pollution control and also in agriculture [18]. Fly ash contents considerable amounts of silica (SiO₂), alumina (Al₂O₃), iron oxide (Fe₂O₃), calcium oxide (CaO). However, magnesium oxide (MgO), titanium oxide (TiO₂), alkaline (Na₂O and K₂O), sulfur trioxide (SO₃), phosphorus oxide (P₂O₅) and carbon have usually small percentages [19].

As mentioned previously, fly ash is considered as a waste material which has a negative impact on the environment. Therefore, in the present study, the authors tried to reuse this low cost inorganic waste by embedding it in polymeric blends and studying its effect on the physical properties of the host matrix. PS, PS/LDPE blend and composites of fly ash/PS/LDPE were prepared by melt mixing method. The effect of fly ash as filler on tuning the electrical properties of the hosting blend of PS/LDPE was investigated. The fly ash was characterized by XRF analysis. The thermal stability of the samples was examined by TGA analysis. AC conductivity, dielectric constant, dielectric loss and electrical modulus were deducted and analyzed at a wide range of applied frequencies and variant constant temperatures.

2-Experimental Techniques

2.1 Material and the Samples

For this study, polystyrene (PS) was purchased from Sigma Aldrich (Germany) and low density polyethylene (LDPE) was obtained from advent chembio (India). Fly ash (FA) powder was supplied from the Nile Sons Co. for brick manufacturing (Cairo, Egypt) and it was used without modifications.

The prepared samples included pristine PS and a blend of PS/LDPE 80/20 wt. % which labeled as PS and C0, respectively. Also, Fly ash was added to PS/LDPE 80/20 blend to form composites with different concentrations. The fly ash contents included 10, 20, 30, 40, 50 wt.% and have labels of C10, C20, C30, C40 and C50, respectively. The samples were prepared by direct mixing in an internal mixer (Bra-bender plasticorder, USA) at a temperature of 90 °C for LDPE and 215 °C for the others. The process of mixture was done with a rotor speed of 80 rpm for 10 min. By using the electrically hydraulic press (XLB-D vulcanizing press, Zhejiang Hongtu machinery factory, China), the melt-blended samples and their composites were compression molded. This was done at 190°C under a constant pressure of (13.5 Mpa) followed by air cooling. Each sample was placed in a stainless steel mold of dimension 100 x 100 x 2 mm³ to be compressed and molded between the two plates of the hydraulic press.
2.2 XRF Measurements

The chemical composition of fly ash was determined by using wavelength dispersive X–Ray Fluorescence (XRF, PANalytical, Axios mAX, Netherlands).

2.3 DTA and TGA Measurements

Thermal gravimetric analysis (TGA) and differential thermal analysis (DTA) were used to determine the thermal stability of the tested material. TGA was performed, using Shimadzu Thermobalance model DTG-60H, for the pristine PS, PS/LDPE blend, C10 and C50. The tested samples were heated under nitrogen gas atmosphere with a heating rate of 5 °C/minute started from the room temperature to 600 °C. The initial weight of the samples was 7.1 ± 1.48 gram for each one.

2.4 Dielectric Measurements

Ac conductivity and the dielectric properties were examined for the whole samples in the form of circular discs, with a diameter of 1 cm. For good conduction, silver past was used to coat opposite surfaces of each sample. The measurements were done in temperature range of 303 - 403 K at frequencies from 1 to 5000 kHz. RLC Bridge (IM 3570 impedance analyzer “Hioki, Japan”) was used to investigate these measurements.

The real part ($\varepsilon'$, dielectric constant) and the imaginary part ($\varepsilon''$, dielectric loss) of the relative complex permittivity $\varepsilon^*$, of a material is given by: $\varepsilon^* = \varepsilon' - j\varepsilon''$. The dielectric constant $\varepsilon'$ describes the amount of both induced and permanent dipole alignment. $\varepsilon' = C/C_o$; where C and C_o is the sample and vacuum capacitance, respectively. The relation of $C_o = \varepsilon_o \times \frac{\varepsilon_o \times a}{d}$, describes the value of $C_o$ where $\varepsilon_o$ is the free space permittivity ($\varepsilon_o = 8.86 \times 10^{-12}$ F/m), d is the sample thickness and a is the effective electrode. The dielectric loss $\varepsilon''$ gives the energy required to align dipoles or move ions. $\varepsilon'' = \varepsilon' \tan\delta$, where $\tan\delta$ is the dielectric loss tangent [7, 20]. Ac conductivity, $\sigma_{ac}$, was calculated from dielectric data through the relation of $\sigma_{ac} = \varepsilon''\varepsilon_o\omega$, where $\omega$ is the angular frequency and $\omega = 2\pi f$ [21].

3- Results and Discussion

3.1 XRF Analysis

Table 1 illustrates the composition of fly ash that was analyzed with X-ray fluorescence (XRF). Iron, calcium, silicon, magnesium, aluminum, and sulfur oxides are some minerals of the chemical composition of fly ash. Some other elements in the ash possess high melting points which remain in furnace during combustion process. Magnetite (Fe$_3$O$_4$), lime (CaO), and periclase (MgO) are formed from oxygen in the air combined with Fe, Ca, and Mg. The fly ash is divided into two classes, depending on American Society for Testing and Materials (ASTM) standard: class C and class F. These classes are mainly relying on the chemical composition of the material as the amounts of the oxides including silicon, alumina and iron oxides. Nevertheless, the major dissimilarity between class C and class F ashes is the sum of the oxides
(minimum 70% for class F ashes and 50% for class C ashes) [22]. In this study, the total mass summation of silicon, magnetite and aluminum (SiO$_2$ + Fe$_2$O$_3$ + Al$_2$O$_3$) is about 79.52 wt. % whereas the amount of calcium oxide (~ 9.5) is less than 10%. Consequently, the used fly ash in this study can be considered according to ASTM as class F or low calcium fly ash [23].

Loss on ignition (LOI) value unable measuring the unburned carbon in fly ash. Conversely, LOI value overestimates the amount of unburned carbon. This is due to some other reactions as dehydration of lime, decomposition of carbonates and oxidation of iron minerals through heating process. Therefore, more accurate ways as thermogravimetric analysis is nominated to replace LOI, which gives accurate data about the weight changes with temperature and time [24].

Table 1. Chemical composition of the fly ash.

| Main constituents | Amount (Wt. %) |
|-------------------|---------------|
| SiO$_2$           | 49.59         |
| TiO$_2$           | 1.469         |
| Al$_2$O$_3$       | 14.14         |
| Fe$_2$O$_3$       | 15.79         |
| MgO               | 2.926         |
| CaO               | 9.506         |
| Na$_2$O           | 1.882         |
| K$_2$O            | 1.515         |
| P$_2$O$_5$        | 0.459         |
| V$_2$O$_5$        | 0.642         |
| Other             | 2.711         |

3.2 DTA and TGA Analysis

Figure 1a illustrates the TGA curves in which the onset degradation temperature of PS is 157 °C that increased by blending it with LDPE to be 192 °C. Table 2 shows that the onset degradation temperature of PS/LDPE 80/20 blend (C0) increased by embedding it with 10 wt.% (C10) and 50 wt.% (C50) of fly ash to be 197 °C and 210 °C, respectively. The increase in values of the onset degradation temperature for the blend (C0) and its composites of C10 and C50 more than PS is due to the reduction in the mobility of polymer chains by adding LDPE and fly ash, respectively. The formation of free radicals at weak bonds (usually peroxide groups and/or chain ends) is usually resulted from the thermal decomposition of polystyrene and LDPE. Consequently, this is followed by chain transfer reactions of the primary free radicals that transmit until the entire matrix is influenced. Embedding fly ash in the blend leads to the motion restriction of the chain segments which prevents the chain transfer reactions. Therefore, the thermal stability of the composites improved [25]. Hence, the degradation process is affected by the embedded fly ash which delays the degradation. This is owing to increase the thermal parameter of the composites with respect to the pristine blend itself, as seen in Table 2.
Figure 1. TGA curves (a) and DrTGA curves (b) for PS, PS/LDPE 80/20 blend (C0) and fly ash/PS/LDPE 10/80/20 composite (C10) and fly ash/PS/LDPE 50/80/20 composite (C50). The inset is the magnification of DrTGA curves from 150 to 400 °C.

Figure 1b shows clearly the thermal decomposition behavior of the samples obtained from the derivative thermogravimetric (DrTGA) curves. For PS, the weight loss started slowly from 157 °C to 321 °C (region 1) followed by a rapid degradation process up to 454 °C (region 2). PS has only one sharp peak, which means that the weight loss of PS takes place in a one-step degradation process from 321 °C to 454 °C, as observed in figure 1b. This was confirmed by the observed values of the weight loss in Table 1 which showed the low value in the 1st region and the high value in the 2nd region. On the other hand, there are sharp and broad (or shoulder) peaks depending on the second and the third decomposition regions for the blend and its composites, as seen in figure 1b. Table 2 illustrates that the positions of the peaks in figure 1b rely on thermal degradation of LDPE, PS and fly ash. A slow degradation (region 1) was observed below 345 °C for the blend and its composites. Afterwards, a sharp degradation (region 2) was observed in temperature ranges of (318 - 407°C), (345 - 405°C) and (345 - 394°C) for the blend, the composites of C10 and C50, respectively. Then, broad peaks (region 3) take place for the blend and the composites of C10 and C50 at (407 – 478 °C), (405 - 548 °C) and (394-600 °C), respectively. The weight loss at the 2nd region was higher than that at the 3rd region for the blend and its composites as clearly observed in Table 2. As mentioned in a published literature [26], that the weight loss of LDPE was happened in a range of temperatures between 400 °C and 500 °C. Therefore, an overlap of degradation is expected to be existed between the 2nd region of both of PS and LDPE. This was responsible about the observed sharp peaks of the blend and its composites, as seen in figure 1b. For all the examined samples, the observed degradation in region 1 can be a result of the elimination of the absorbed moisture [27]. Table 2 shows that there was no decomposition residue for PS and its blend while it was about 9% and 33% for C10
and 50 C50, respectively. Therefore, the sample of the more thermal stability is the composite that contains the highest amount of fly ash (50 wt.%, C50).

**Table 2.** The degradation temperature, weight loss (%), total residue (%), the temperature of mass loss at 10% ($T_{10\%}$), 50% ($T_{50\%}$) and 65% ($T_{65\%}$), and maximum degradation temperature from DrTGA curves ($T_{max}$).

| Sample (wt.%) | Degradation temperature ($^\circ$C) | Weight loss (%) | Total residue (%) | $T_{10\%}$ ($^\circ$C) | $T_{50\%}$ ($^\circ$C) | $T_{65\%}$ ($^\circ$C) | $T_{max}$ from DrTGA ($^\circ$C) |
|---------------|--------------------------------------|----------------|-----------------|----------------------|----------------------|----------------------|-----------------------------|
| Ps            | Region 1: 157-321 Region 2: 321-454  | Region 1: 5    | Region 2: 95  | 0 357 382 387       | 387                  |                      |                             |
| PS/LDPE 80/20| Region 1: 192-318 Region 2: 318-407 Region 3: 407-478 | Region 1,2: 84 Region 3: 16 | 0 369 388 393 | 388 434             |                      |                             |
| Fly ash/PS/LDPE 10/80/20 | Region 1: 197-340 Region 2: 340-405 Region 3: 405-548 | Region 1,2: 73 Region 3: 18 | 9 358 385 393 | 383 433             |                      |                             |
| Fly ash/PS/LDPE 50/80/20 | Region 1: 210-340 Region 2: 340-394 Region 2: 394-600 | Region 1,2: 32 Region 3: 35 | 33 367 416 477 | 392 439             |                      |                             |

### 3.3 Dielectric Properties

The polarity as being polar or non-polar has a great effect on dielectric properties of polymers. Polystyrene (PS) and Low density polyethylene (LDPE) are considered as non-polar polymers [7, 28]. The variation $\varepsilon'$ with frequency at different constant temperatures for PS and C10, as an example for all the composites, are depicted in figures 2a & 2b, respectively. Regarding to these figures, the permittivity illustrated a discrepancy in relation to the high frequency and a similar characteristic at low frequency. At a lower frequency, < 50 kHz, the values of $\varepsilon'$, for PS, its blend and its composites, was considerably high. This can be attributed to the asymmetry at phenyl side group of PS, thus it has a small dipole moment (0.26D), despite of being a non polar polymer [29, 30]. The high values of $\varepsilon'$ at low frequencies can be interpreted on base of the interfacial polarization i.e., the building up of bound charges at interfaces within the material of the samples. This interfacial polarization occurs in heterogeneous structures due to the accumulation of charges at the interfaces between variant regions which are not similar to each other in its conductivity. Besides, at the interfaces between the sample and the electrodes, there is an accumulation of bound charges which is included in the net polarization (electrode polarization). An extra capacitive component is produced into the system by this space charge accumulation [31]. Conversely, for all the applied constant temperatures, the values of $\varepsilon'$, decreased
with increasing the applied frequency, >50 kHz, for all the composites, see figure 2b. This can be due to the dipole polarization which requires a considerable long time as the side groups of the main chains of the polymers lose slowly their capability to track the applied field. Consequently, their contribution to the polarization dies down and this resulted in decreasing the dielectric constant of the composites [31, 32]. However, the values $\varepsilon'$ became independent of frequency for PS and its blend, at higher frequencies, >50 kHz, due to non-polarity structure of PS and LDPE. Also, figures 2a & 2b showed that, for PS and its composites, $\varepsilon'$ decreased by increasing the applied temperature due to the low polarity of PS and LDPE [33].

![Figure 2](image-url)

**Figure 2.** The variation of dielectric constant ($\varepsilon'$) and dielectric loss ($\varepsilon''$) with frequency for PS (a and d, respectively) and fly ash/PS/LDPE 10/80/20 composite (b and e, respectively). The variation of dielectric constant ($\varepsilon'$) and dielectric loss ($\varepsilon''$) with frequency, for all the samples, at constant temperature of 61°C (c and f, respectively).

The composition of the material as well as the reliance on the metal electrode type can obviously affect the dielectric properties of the polymers. For instance, the imaginary part ($\varepsilon''$) of the relative complex permittivity $\varepsilon^*$, depends on two important factors. The first one is the electrode resistance which can be minimized through using electrode material with high conductivity. However, the second one is the properties of the polymer itself. These two factors show frequency and temperature dependence [34]. The variation of $\varepsilon''$ with frequency, at variant constant temperatures, for PS and C10, as an example for all the composites, are presented in figures 2d & 2e, respectively. For PS and its composites, $\varepsilon''$ values were decreasing from 1 kHz up to 50 kHz followed by a stability until 5 MHZ for PS and up to 1 MHz for the
composites. This can be attributed to the slower polarization mechanism as a result of stretching and twisting of the long carbon chains attached to the main polymeric chain [35]. However, a slight increase in the values of $\varepsilon''$ were observed at frequencies $> 1$ MHz for all the composites. This can be a result of increasing the relaxation loss of the dielectric materials in case of applying a high frequency. In addition, at higher frequencies, ionic polarization can take place and consequently this encourages the charge carriers to instantly cross the surface of the composite. This can lead to an additional improving in the values of $\varepsilon''$ at higher frequencies [36]. Figures 2c & 2f showed the variation of $\varepsilon'$ and $\varepsilon''$, respectively, with frequency for PS, the blend (C0) and the composites (C5 - C50), at 61°C. By increasing the content of fly ash in the blend the values of $\varepsilon'$ and $\varepsilon''$ increased up to the composite of C40 then they decreased for the higher fly ash composite (C50). The observed increase in $\varepsilon'$ and consequently $\varepsilon''$ can be due to the higher values of the dielectric constant of the metal oxides of the fly ash as SiO$_2$ (dielectric constant $\approx 3.9$) and Al$_2$O$_3$ (dielectric constant $\approx 9$) [17, 37].

The ln - ln plot of $\varepsilon''$ versus frequency, at different constant temperatures, are illustrated in figures 3a, 3b & 3c, for PS, C20, as an example for the composites, and all the samples at 61°C, respectively. Figures 3a & 3b shows a linear variation of plotting ln $\varepsilon''$ versus ln $\omega$. Thus, the frequency dependence of the imaginary part of the complex permittivity $\varepsilon''$ can be presented by the law of fractional power as the following [38]:

$$\varepsilon'' = B\omega^n$$

Where $B$ is a constant and $n$ is a frequency power factor. The relation between ln $\varepsilon$ and ln $\omega$ is known as Maxwell – Wagner formula for the heterogeneous media which is obtained by re-arranging Eq. 1 as the following [39]:

$$\ln \varepsilon'' = \ln B + n \ln \omega$$

$$n = -\frac{4\pi kT}{W_M}$$

Where $W_M$ is the maximum barrier height which is defined as the energy required to move the electron from a site to the infinity. The values of $n$ were determined from the negative slope of the straight lines obtained from ln – ln plot of $\varepsilon''$ versus frequency, see figures 3a, 3b & 3c. Table 3 shows that the deduced values of $n$ and $W_M$ were obviously temperature dependent as they were increasing by increasing the applied temperature. Also, Table 3 exhibits the effect of the fly ash content on the values of $n$ and $W_M$ for the composites which were all higher than those of the blend (C0) itself. Dominating polarization in a material by mobile hopping charge carriers may not display Debye type loss peak in their dielectric curves. Instead, a monotonic rise of both dielectric constant and loss values with decreasing frequency take place and hence it follows the fractional power law of $\omega^n$. This attitude was firstly known by Jonscher [40-42] which is called the universal dielectric response.
3.4 AC conductivity

The values of ac conductivity (\(\sigma_{ac}\)) were obtained from the measured capacitance and the calculated dielectric loss [43]. The variation of ac conductivity, with a wide range of frequencies, from 1 kHz to 5 MHz, was examined at different temperature from 35 to 131\(^\circ\)C. Figures 3d, 3e and 3f illustrate the frequency dependence of \(\sigma_{ac}\) for PS, C20 composite, as an example for all the composites, and all the different samples at 61\(^\circ\)C, respectively. For each constant temperature, the ac conductivity exhibits a linear increasing in its value with increasing the applied frequency as a common behavior for all the samples. This behavior can be due to the presence of charge carriers that supply to the conduction mechanism along the polymer chains [7]. In addition, for all the samples, figures 3d & 3e showed that \(\sigma_{ac}\) was decreasing by increasing the applied temperatures. This behavior was due to the fact that the hopping distance increases with the thermal expansion of the polymer and polymer blend matrices. However, the more effect of the applied temperature on \(\sigma_{ac}\) was clearly displayed at low frequencies. However, \(\sigma_{ac}\) was nearly temperature independent for higher frequencies that existed above 1 MHz, for all the samples.
Table 3. The values of the exponent (n) and the barrier potential (W_m), at different temperatures, for PS, PS/LDPE blend and variant composites of fly ash/ PS/LDPE.

| T°C | Parameter | PS  | C0  | C5  | C10 | C20 | C30 | C40 | C50 |
|-----|-----------|-----|-----|-----|-----|-----|-----|-----|-----|
| 41  | n         | 0.158 | 0.041 | 0.154 | 0.226 | 0.172 | 0.591 | 0.149 | 0.309 |
|     | W_m      | 0.192 | 0.169 | 0.191 | 0.209 | 0.196 | 0.396 | 0.190 | 0.234 |
| 53  | n         | 0.162 | 0.055 | 0.172 | 0.208 | 0.196 | 0.654 | 0.143 | 0.239 |
|     | W_m      | 0.201 | 0.178 | 0.203 | 0.212 | 0.209 | 0.487 | 0.196 | 0.221 |
| 61  | n         | 0.170 | 0.083 | 0.266 | 0.223 | 0.236 | 0.603 | 0.142 | 0.325 |
|     | W_m      | 0.208 | 0.188 | 0.235 | 0.222 | 0.226 | 0.434 | 0.201 | 0.255 |
| 71  | n         | 0.176 | 0.141 | 0.263 | 0.193 | 0.301 | 0.643 | 0.165 | 0.358 |
|     | W_m      | 0.215 | 0.207 | 0.241 | 0.220 | 0.254 | 0.498 | 0.213 | 0.277 |
| 81  | n         | 0.23  | 0.216 | 0.320 | 0.193 | 0.323 | 0.581 | 0.146 | 0.403 |
|     | W_m      | 0.237 | 0.233 | 0.269 | 0.226 | 0.270 | 0.436 | 0.214 | 0.306 |
| 91  | n         | 0.255 | 0.266 | 0.351 | 0.245 | 0.361 | 0.580 | 0.170 | 0.551 |
|     | W_m      | 0.252 | 0.256 | 0.289 | 0.249 | 0.294 | 0.448 | 0.226 | 0.419 |
| 101 | n         | 0.232 | 0.343 | 0.372 | 0.298 | 0.390 | 0.670 | 0.128 | 0.570 |
|     | W_m      | 0.251 | 0.294 | 0.307 | 0.275 | 0.317 | 0.585 | 0.221 | 0.449 |
| 111 | n         | 0.217 | 0.406 | 0.390 | 0.281 | 0.338 | 0.630 | 0.138 | 0.686 |
|     | W_m      | 0.253 | 0.334 | 0.325 | 0.276 | 0.299 | 0.536 | 0.230 | 0.632 |
| 121 | n         | 0.185 | 0.471 | 0.558 | 0.352 | 0.621 | 0.535 | 0.24  | 0.687 |
|     | W_m      | 0.249 | 0.385 | 0.460 | 0.314 | 0.537 | 0.438 | 0.268 | 0.650 |

Also, at constant temperature of 61°C, figure 3f shows that increasing the fly ash content in the blend can improve \(\sigma_{ac}\) up to the composite C40 then it decreased. This can be attributed to the fact that increasing the fly ash content enables forming excess of charge carriers. So, the hopping conduction mechanism is dominant up to the composite of C40. However, for higher content of fly ash as C50, the filler particles may become large enough to be as obstacles for the hopping of charge carriers between the sites [44].

3.5 **Complex Electrical Modulus Properties**

Interfacial polarization plays an important role in the polymeric materials due to the additives, fillers or even impurities that make these systems to be heterogeneous. Generally, in some systems, interfacial relaxation is obscured by conductivity and the high dielectric permittivity at low frequencies. To overcome such a problem in studying the interfacial polarization, it has been decided to use the formalism "electric modulus" to study the polymer conductivity relaxation behavior [21]. In fact, for numerous cases, the dynamics of interest are better to be understood through plotting data in one of some choices, which are counterpart representations. The data illustration in M* (the electric modulus which is equal to \(1/\varepsilon^*\)) is physically comparable with the data depiction in \(\varepsilon^*\) formalism. Nevertheless, in some cases, the data represented in M* becomes more
helpful as the effects of the space charge can be restrained. Therefore, the conductivity current relaxation can be studied well [31, 45]. The complex electric modulus can be obtained according to the deduced formula by McCrum et al. [46] as the following:

\[
M' = \frac{1}{\varepsilon} = M' + iM'' = \frac{\varepsilon'}{\varepsilon' + i\varepsilon''} + i \frac{\varepsilon''}{\varepsilon' + i\varepsilon''}
\]  

(4)

Where \(M'\) and \(M''\) are the real and imaginary parts of the electric modulus, respectively. Owing to being the complex permittivity is in the denominator of Eq 4, its effect in dominating \(M'\) and \(M''\) has effectively lessened. Consequently, this permits more broad analysis of the dielectric data [21]. Herein, the complex modulus analysis relied on plotting \(M'\) and \(M''\) over a wide range of frequencies and different constant temperatures. The frequency dependence of \(M'\) and \(M''\) for PS, the composite C10, as an example for all the composites, and all the different samples at 61°C, are presented in figures 4a & 4d, figures 4b & 4e and figures 4c & 4f, respectively.

For all the samples, the \(M'\) values do not equal to zero at low frequencies and high temperature. Consequently, it is expected that the electrode polarization may take place for the all samples [47], see figures 4a, 4b & 4c. It can be noted that \(M'\) is increasing with increasing the applied frequency and with increasing the temperature for PS, its blend and composites. The \(M''\) frequency dependence for PS and its blend (C0), as seen in figures 4d & 4f, shows the existence of relaxation peaks at low frequency. However, the values of \(M''\) for the composite C10 were high at low frequency. Afterwards, they decreased up to \(\approx 500 \text{ kHz}\) followed by an obvious increasing at high frequencies without appearing any peak of relaxation, see figure 4e. This behavior is common for all the composites except for the composites of C5 and C50 which have a peak of \(M''\) at \(\approx 59 \text{ kHz}\) and 40 kHz, respectively. For all the observed relaxation peaks, the low-frequency side of the peak describes the region of frequencies where the hopping process of the charge carriers can occur from one site to the neighboring one. However, the charge carriers are confined to their potential wells and they can make localized motion within the potential well at the high frequency side of \(M''\) peak. The range of frequencies, where the peak itself is appeared, can explain the transition from long-range to short range mobility of the charge carriers [48, 49]. Figures 4c & 4f prove that the frequency dependence of \(M'\) and \(M''\), respectively, for all the samples, are depending on the filler content.
Figure 4. The variation of real (M') and imaginary (M'') parts of electric modulus for PS (a and d, respectively) and fly ash/PS/LDPE 10/80/20 composite (b and e, respectively). The variation of real (M') and imaginary (M'') parts of electric modulus for all the samples at constant temperature of 61°C (c and f, respectively).

4- Conclusion

The simple melt mixing method was used for preparing PS, PS/LDPE blend and composites of fly ash embedded in the blend with different contents. Analyzing the obtained data of XRF for the used fly ash showed that it can be considered according to ASTM as class F fly ash. TGA analysis illustrated that the degradation of the composites is affected by the filler content which delays the degradation process. This is due to increasing the thermal parameter of the composites with respect to the pristine blend itself. DrTGA curves showed that PS has only one sharp peak, which means that the weight loss of PS takes place in a one-step degradation process. However, there were sharp and broad peaks depending on the different decomposition regions for the blend and its composites. The values of ε' became independent of frequency for PS and its blend, at higher frequencies, >50 kHz. This can be attributed to the non-polarity structure of PS and LDPE. By increasing the content of fly ash in the blend, the values of ε' and ε'' increased up to the composite of C40 then they decreased for C50 composite. The frequency dependence of M' and M'', respectively,
for all the samples, are depending on the embedded content of the filler in the PS/LDPE blend.

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