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Electro mechanical properties changes of LDPE doped with industrial type MgO for cable insulation purposes

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ABSTRACT

This manuscript introduces the changes of a comprehensive electromechanical properties bundle for low density polyethylene compounded to microscale magnesia (LDPE/MgO) to obtain electrical cables insulating material. Composites of various filler loading weight ratios were prepared by melt intercalation technique; multiple samples were produced in sets as they were cut with definite dimensions as per recommendations of the related testing standard then electrically and mechanically examined following the instruction dictated by the code while preserving typical test condition for all sets. Dielectric strength, volume resistivity, capacitance, and loss angle were the tests of the electrical test pack, while elongation, tensile strength, and melt flow rate were the mechanical and rheological tests applied. Test's findings were compared to each other's and to the base material to identify the differentiation. Electrical test results show improvements in the composite features at low loading percentages, whereas the mechanical tests revealed a deterioration in the mechanical properties along with all ratios under investigation. The research aims to determine the compositing benefit extents and drawbacks when a conventional compounding method and inexpensive filler are used, incurring marginal cost impact.

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1. INTRODUCTION

Polyethylene, that famous thermoplastic made from ethylene monomer polymerization, is considered one of the most successful insulating materials technically and commercially [1], [2]. It possesses convenient electrical isolation characteristics, including high dielectric strength and low permittivity; moreover, it has an industrial preference for its availability and low price. Despite that, PE encounters few flaws like temperature restrictions during processing and insulation deteriorations that need antioxidant additives to overcome [3]. Crosslinking solved the majority of these disadvantages; however, XLPE can't be annealed and remolded in recycling operation [4].

In the context of the relentless endeavor to develop cable insulators, the term (composites) has prominently emerged in recent decades as an interactive solution to intractable problems. Compositing allows varying the inherent polymer properties by exploiting specific compositing techniques to combine one or more fillers selected from the wide range of organic or inorganic fillers into the polymeric base material. The features of the original host stand on one hand and on the other hand, the type, size, shape, and proportion of fillings added [5], in addition to the method of filler surface treatment and silane utilized in the

functionalization process [6], are among the variables that affect the morphological, mechanical, electrical and rheological properties of the excretory compound.

Plenty of researches have investigated broadly the Nano compositing and particularly LDPE-MgO combination [7], [8] Nanocomposite, where they concluded the modifications occurred to different characteristics like space charge [8] repression, breakdown voltage enhancement [9], and volume resistivity improvement [10] yet, from a feasibility point of view, filler cost need to be considered. The price of one gram of pure Nano magnesia in the local Egyptian market is around 65 EGP (4 USD) while, the cost of one gram of the same material of industrial type is almost 0.16 EGP, bearing in mind that the industrial magnesia is produced in micro scale size with less purity and homogeneity; therefore, it is expected that the features improvements shall not be as good as their counterparts acquired when using fine-scale filler, however, due to that tremendous monetary variance, it is worth to study the main electromechanical properties obtained for polymer-based composites doped with such industrial micro filler and to evaluate whether is it cost efficient or not. This article presents an attempt to examine the morphology and key electromechanical features for LDPE doped with industrial MgO in a marketable vision that links material grade and processing expenses to the quality of the final product.

2. EXPERMINTAL WORK

2.1. Materials

Pure low density polyethylene was procured from SABIC KSA with a density of 0.924 g/cm3 and characterized by a melt flow rate of 2.00 g/10 min at 2.16 kg at 190 C provided in pellets form. It was crushed to finer particles of almost 0.2 mm diameter to facilitate the compounding. While micro scale MgO of industrial application type was obtained from the local market.

2.2. Micro composites preparation

The adopted technique was Melt Intercalation which was selected for its popularity in industrial approaches [11]. In melt intercalation, shear stresses are applied to the polymer that is heated to melting temperature in the presence of the filler, causing homogenous distribution of the filler into the polymeric host matrix. Step by step specimen's sheets fabrication process is illustrated by the flowchart in Figure 1.

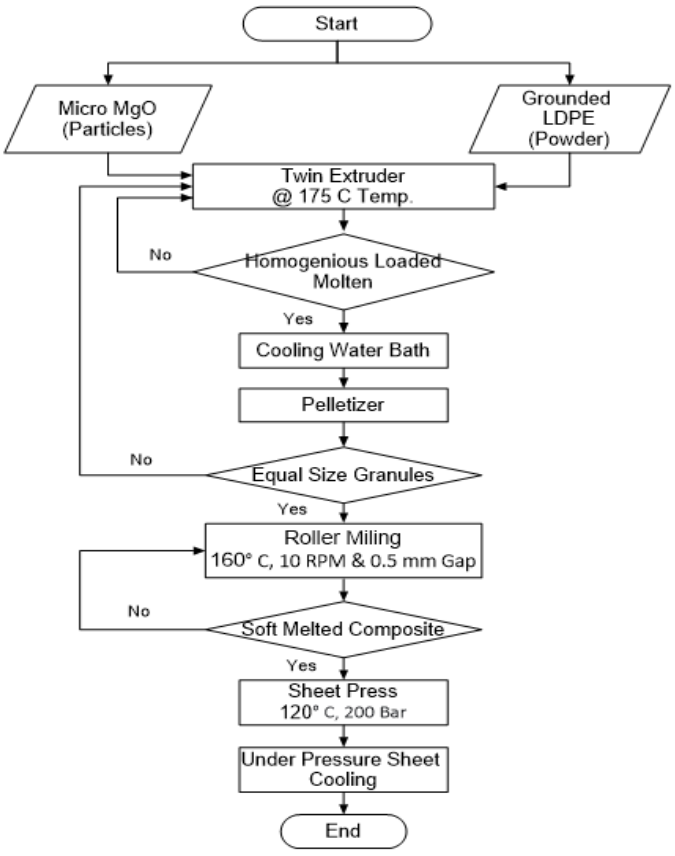


Figure 1. LDPE - MgO sheets preparation

Induced stresses were provided by (Brabender Lab Station) Twin Extruder; then, the soft kneading was quenched in a water bath and trimmed by a pelletizer. Pellets were softening by roller miller, and the end product was obtained by compression molding of the pre-soften fabricated granules using (Gibitre Lab Press) sheet compressing plates [12]. Finally, 200 x 200 mm square sheets with roughly 1 mm thickness are obtained. Many sheets, each was made of different composites with various loads, were acquired in order to execute the designated tests on them. Table 1 depicts the compositions prepared.

Table 1. Different samples compositions

Sample	Code	LDPE (wt. %)	Micro MgO (wt. %)
LDPE	B	100	0
LDPE + 5% MgO	M1	95	5
LDPE + 10% MgO	M2	90	10
LDPE + 15% MgO	M3	85	15
LDPE + 20% MgO	M4	80	20

2.3. Morphology characterization

T Philips x'pert x-ray diffraction (XRD) was used to check the purity and crystallinity phase of the filler powder. TEM, JEOL JEM-2100, was utilized to take high resolution captures to determine size and shape of the particles. SEM captures were taken for composites by JEOL JSM 5400 LV Scanning Electron Microscopy to examine the filler dispersion.

2.3.1. X-ray powder diffraction (XRD)

Cubic phase structure is recognized from X-Ray Diffraction by comparing the obtained peaks to their standard correspondent. The obtained pattern is presented in Figure 2, it shows an acceptable magnesium oxide purity level.

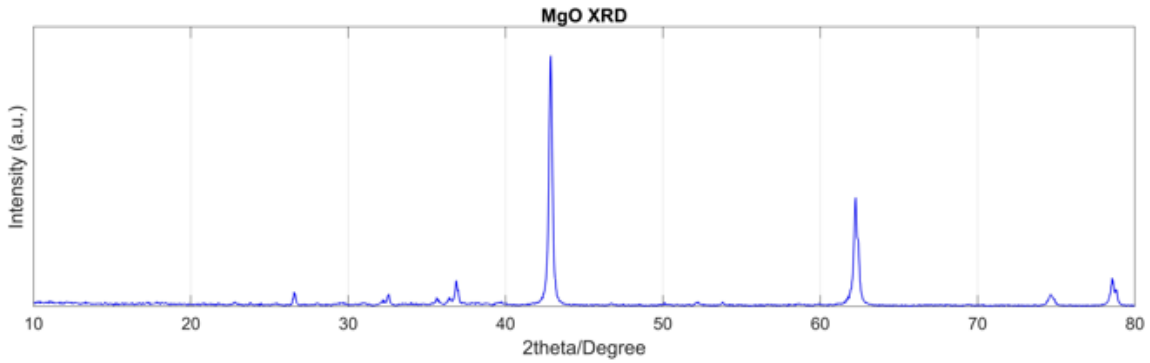


Figure 2. XRD of industrial type MgO

2.3.2. Transmission electron microscope (TEM)

Industrial MgO is a mixture of spherical and quasi-spherical configuration, powder form and white in color. TEM photos, as in Figure 3, show that the diameter of the particles is 250nm on average.

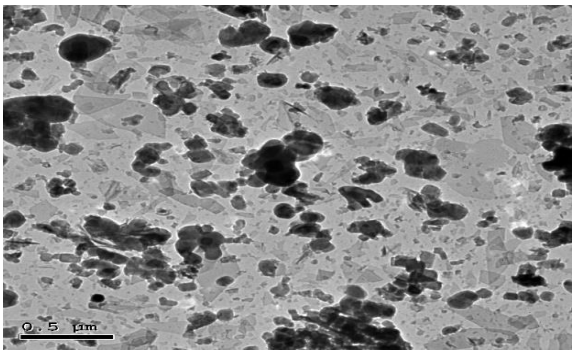
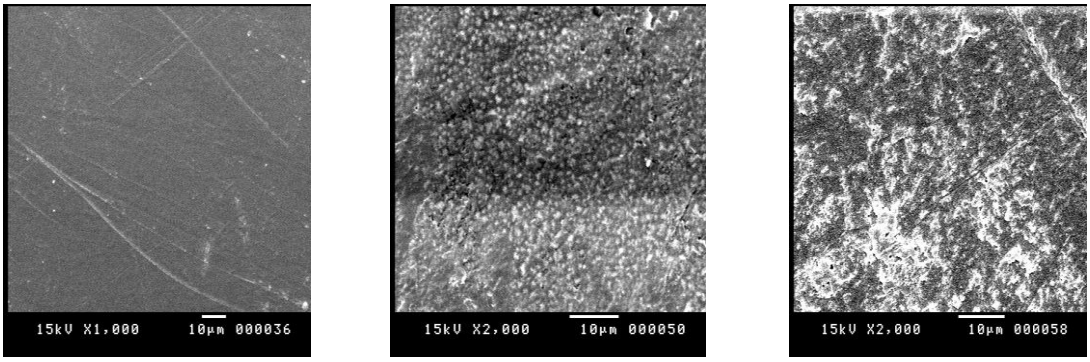


Figure 3. TEM of industrial type MgO

2.3.3. Scanning electron microscopy (SEM)

Micro size magnesium oxide dispersion in the LDPE after the completion of the compounding process was checked using SEM. Figures 4(a)-(c) illustrate the micrographs of neat LDPE, composites of 5wt% reinforced and that of 20wt% doping, respectively. By scrutinizing captures, a, b and c, it could be noticed the good dispersion of the filler at low filling ratios; conversely, the clusters agglomerated at high loading ratios.



(a) (b) (c)

Figure 4 (a). SEM of sample B, b). SEM of sample M1, c). SEM of sample M4

2.4. Electrical tests

2.4.1. Dielectric strength

Dielectric strength (DS) was done following ASTM D-150 using TERCO HV Lab. Kit, Sweden. Number ten circular discs were extracted from sheets of designated concentration (B & M1 to M4), the diameter of all samples was greater than 5 cm to avoid flashover during the test. Externally applied voltage has been ramped up gradually with 2 kV/sec rate on the electrodes till failure occurrence. Using the kit options, maximal voltage acquired before the breakdown incident was held for recording.

Field strength was computed by (1).

$$E = {}^{V}/_{d} \quad (1)$$

Where:
E is the Dielectric Strength in kV/mm
V is Breakdown peak potential recorded in kV
d is Thickness of Specimen in mm
Data analysis was done with Weibull [13], two parameters, probability function presented in (2).

$$P(E) = 1 - exp\left[-\left(\frac{E}{\alpha}\right)^{\beta}\right] \quad (2)$$

Where:
E: Experimentally recorded Dielectric Strength kV/mm
P(E): Cumulative breakdown failure probability
α: Breakdown gradient at failure probability of 63.2%
β: Shape parameter

For low counts of data population, probability (Pi) related to the i -th sample breakdown incident (i) is obtained as introduced in (3).

$$P_i = \frac{i-0.44}{n+0.25} \quad (3)$$

Least square method of linear regression technique was used to determine α & β Weibull variables.

2.4.2. Volume resistivity

It was tested according to ASTM D-275-99 using TOA, Ultra mega-ohmmeter, model SM-8210, Japan.

2.4.3. Electrical capacitance and dissipation factor

They were determined in accordance to ASTM D-669 and D-150, respectively, by using Ceast Qmeter, model 0194C, Italy.

Permittivity was calculated at 1 MHz frequency using measured capacitance and (4).

$$\varepsilon_r = C_p \frac{t}{\varepsilon_0 A} \quad (4)$$

Where,
(Cp) is practically measured capacitance, Ɛ0 = 8.854 × 10 $^{-12}$ F/m is the permittivity of free space, (A) is the area of electrodes, and (t) is the thickness of the samples.

2.5. Mechanical/rheological tests

2.5.1. Tensile strength and elongation at break

Tensile strength at break (TSb) and elongation at break (Eb) tests were determined according to DIN 53504 by using Gibtre Tensor electro-dynamic tester with a cell load capacity of 5 kN at 50mm/min speed. Specimens were cut in dumbbell shape with dimensions shown in Figure 5 [14]. The shape is obtained by a die press cutter with a specific die blade. For each filler concentration prepared, six samples were clipped and tested, the average of the registered values was calculated in order to minimize the error via tester software.

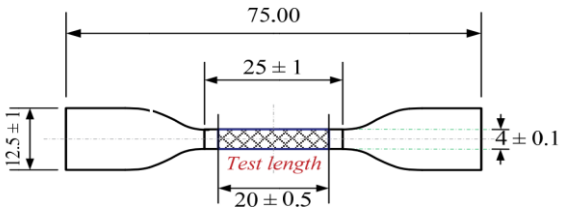


Figure 5. Sample in dumbbell shape according to DIN-53504

2.5.2. Melt flow rate

Melt flow rate (MFR) test was executed according to ASTM D-1238 by using Zwick/Roell LTM electro-dynamic tester. MFR is defined as the polymer mass that flows through a particular die size caused by applying a specific pressure at a certain temperature for a limited time. It represents the rheological properties of a material in the molten state.

3. RESULTS AND DISCUSSION

3.1. Electrical tests

3.1.1. Dielectric strength

Cumulative probability of breakdown gradient as a function of micro MgO percentage is introduced in Figure 6, whereas the dielectric strength and Weibull shape parameter are summed up in Table 2.

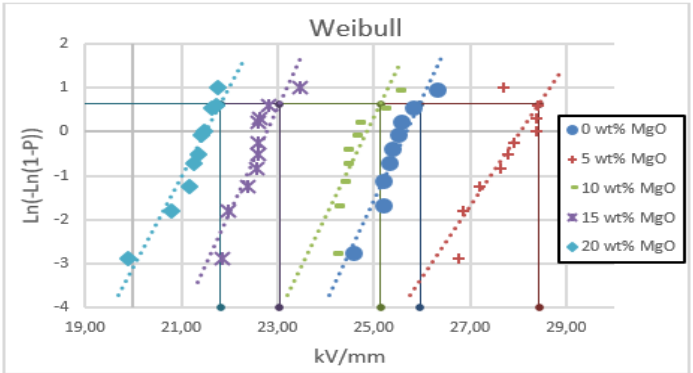


Figure 6. Weibull distribution of all filler concentrations

Table 2. Breakdown voltage at accumulated probability of 62.3 and corresponding weibull shape factor

Unit	MgO wt.%				
	0	5	10	15	20
kV/mm	25.96	28.44	25.14	23.04	21.81
β	2.32	1.63	2.23	2.41	2.06

Results were interpolated to illustrate the behavior of the dielectric strength, as shown in Figure 7. It could be noticed that a limited increase in the breakdown voltage is achieved only at low loading percentages. At around 4 percent filler weight, the composite possesses the highest breakdown voltage. This incrimination is followed by severe discrimination rate with further concentration degrees.

Generally, doping a polymer by inorganic filler introduces traps that have the ability to capture charge carriers. As proved by TEM scans, the size of the used filler is considered to be small within the micro scale segment. At low filler loading, MgO particles exhibit uniform distribution into the matrix, with such inner volume fraction, the emerged traps density is sufficient to marginally suppress the charges mobility and enhance the breakdown voltage. With more and more filler in the mixture, particles coalesce occurred, and the gross surface area of magnesia lumps can only engender low density traps with shallow depth; consequently, the dielectric strength is drastically diminished [15].

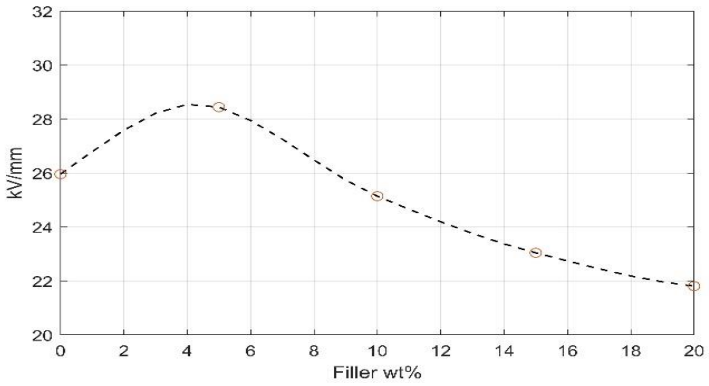


Figure 7. Micro LDPE-MgO composite dielectric strength

Table 3 represents a dielectric strength comparison between values of the micro compound under investigation and results obtained from earlier research, conducted on nano composite having typical composition (LDPE/MgO) and processed in the same technique; however, the used filler was in nano size [16].

Table 3. Breakdown anticipated when using micro and nano sizes of MgO filler-based LDPE

MgO	Filler Size	Max. Predicted Dielectric Strength		Dielectric Strength Improvement	Corresponding Filler wt.%
		Neat LDPE	LDPE/MgO		
Unit	nm	kV	kV	%	wt.%
Nano	70	25.96	38.25	47.3%	1.4%
Micro	250	25.96	28.52	9.9%	4.0%

3.1.2. Volume resistivity

Figure 8 demonstrates volume resistivity test results against filler loading ratio. By scanning the graph, it is easy to realize that volume resistivity increased with the addition of filler up to 10% concentration; after that, ohmic value dropped down gradually with extra filler addition till it reached almost the same value of the neat base material at 20% filler concentration.

The result could be reasonably justified by the trapped de-trapped recursive action. At small loading levels, a portion of the injected charges is trapped at the area of contact to the electrode, causing a space charge. This cloud will act as an electrostatic barrier, and with the assist of the other scattered traps, they will counter the carrier's mobility, hence improving the volume resistivity. Oppositely, at excessive filler contents, agglomeration probabilities increased and trapping energy minimized; consequently, the conductance improves and the resistivity decreases [17], [18].

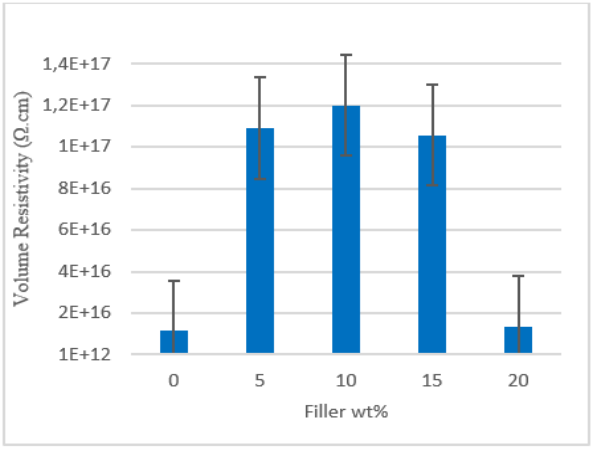


Figure 8. Micro LDPE-MgO composites volume resistivity

3.1.3. Relative permittivity and loss factor

Both Relative Permittivity and Loss Factor obtained findings were demonstrated in Figure 9. Relative permittivity increased with increasing the filler ratio because the MgO permittivity value itself, estimated to be around 10, is much higher than that of LDPE, which is around 2.1; consequently, the resultant value will keep rising with the augmentation of filler concentration. Another factor is the impact of the Maxwell-Wagner interfacial polarization that will normally increase the loss angle of the composite [19], [20].

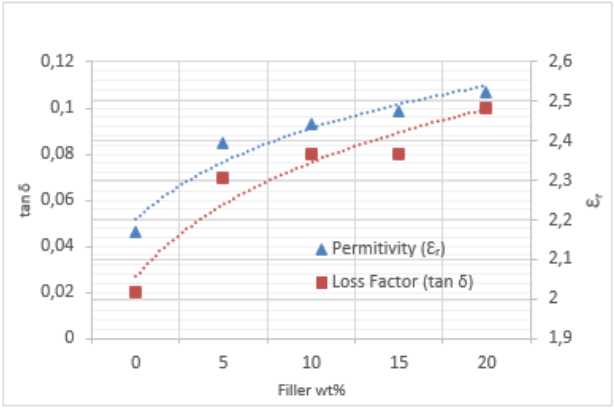


Figure 9. LDPE-MgO permittivity and loss factor

3.2. Mechanical tests

3.2.1. Tensile strength & elongation at break

Tests results for the loaded composites are shown in Figure 10. It could be observed that tensile strength and elongation properties have an inverse proportional relation with the filler concentration. This behavior could be attributed to non- uniform distribution of the filler due to its propensity to aggregate [21]. With such comparable high percentage of loading that reinforces filler mutual attraction and facilitates the agglomeration of the particles, the cohesion forces between those infested filler clusters and the host matrix are lessened significantly. This will yield a reduction of the sectional load standing capacity. By applying meager external stresses, forces will be projected to those embedded masses creating stress focal regions consequently the weak links will be debonded easily [22], [23].

Elongation decay with boosting the filler ratio was due to MgO hydrophobic characteristics. In the presence of natural humidity, moisture content is inherently associated and during compositing process that mainly incorporates heating to melting temperature, these contents evaporate causing pores appearance that act as internal notches and facilitate the fracture. In this case, it could be said that the material brittleness increased [3], [24].

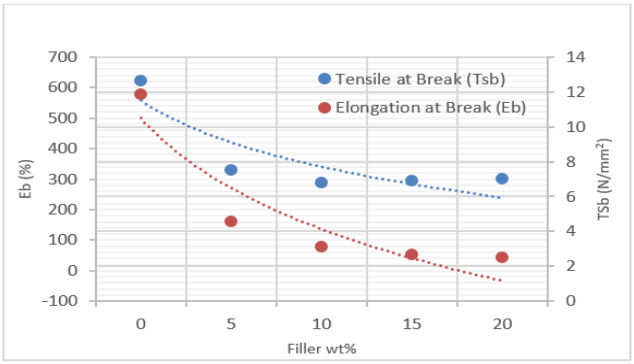


Figure 10. Tensile strength & elongation at break

3.3. Rheological properties

3.3.1. Melt flow rate

Figure 11 introduces the melt flow rate practical measurements of the prepared micro composites at 2.1kg load and 190◦ C. it could be observed the decay in melt flow rate with the addition of filler percentage. This could be ascribed to that, melt flow main reliance falls on the molecular chain mobility of the matrix which is motivated by the balance of temperature, versus entanglements. In composites with higher filler loading degrees and temperature 200º C, the densely filler particles have significant mutual interfacial forces that are capable to suppress the motility of the chains, leading to reduction in melt flow index value [25].

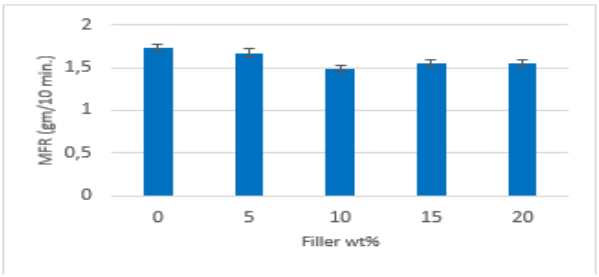


Figure 11. Micro LDPE-MgO composite melt flow rate

4. CONCLUSION

In this study, electro-mechanical and rheological properties of different weight ratios of industrial type Micro MgO composited to LDPE were examined. The conclusions are briefed: a) Electrical tests conducted represented in dielectric strength and volume resistivity exhibited improvements in low loading levels only, optimum dielectric strength could be achieved by almost 4% filler ratio. Similarly, volume resistivity had a 90% increase at 10% filler ratio, but with the continuous addition of the filler, both measured features decremented to lower levels. b) Mechanical tests were tensile strength and elongation. Both indicated undesirable decrescent mensuration. Tragic drop down was observed in (Eb) as it dropped from 600% registered in pristine LDPE test to just 40% in samples loaded with 20% filler. c) Around 11% melt flow rate reduction could be attained. Lower MFR is a good sign of fewer difficulties in the processing of this composite. d) Overall, despite the low cost of the ingredients and processing technique, the composites appeared not to be suitable for cable insulation applications.

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