

# The Influence of a Commercial Few-Layer Graphene on Electrical Conductivity, Mechanical Reinforcement and Photodegradation Resistance of Polyolefin Blends

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## Background theory and the calculation of critical shear rate to ensure coalescence during deformation

The morphology of an immiscible polymer blend is governed by polymer composition and polymer processing conditions. During processing, polymer blends undergo breakup and coalescence phenomena. If processed under low stress and steady uniform shear flow, the deformation of droplets, in a droplet-dispersed morphology, is directly related to capillary number and viscosity ratio.

$$\text{Capillary number, } C_a = \frac{\eta_m R_v \dot{\gamma}}{\alpha} \dots\dots\dots (1)$$

$$\text{Viscosity ratio, } \rho = \frac{\eta_d}{\eta_m} \dots\dots\dots (2)$$

Here,  $\eta_m$  is the viscosity of matrix phase,  $\eta_d$  is the viscosity of dispersed phase,  $R_v$  is the volume average radius of droplets,  $\dot{\gamma}$  is the applied shear rate, and  $\alpha$  is the interfacial tension between matrix and dispersed phase.

As mentioned before, coalescence and droplet break up phenomenon take place during processing. However, break up is dominant when capillary number is above a critical value,  $C_{ac}$ . Below  $C_{ac}$ , coalescence takes place. Grace [43] outlined a relation between critical capillary number and viscosity ratio. The experimental fit of Grace's curve was modeled by Tucker and Moldenaers [44] by following equation:

$$\log C_{ac} = -0.506 - 0.0994 \log \rho + 0.124 (\log \rho)^2 - \frac{0.115}{\log \rho - \log 4.08} \dots\dots\dots (3)$$

In this work, following parameter (shown in Table S1) was used to calculate the shear rate to ensure droplet coalescence during the coalescence test, using the rheometer, MCR 501.

Table S1: The parameters and corresponding values to calculate the shear rate to ensure droplet coalescence during the coalescence test.

Parameter	Value
$\eta_m$	880 Pa.s
$\eta_d$	374 Pa.s
$\rho$	0.43
$R_v$	4.94 $\mu\text{m}$
$\alpha$	1.21 N/m [30]

Calculated shear rate was  $0.13 \text{ s}^{-1}$ . A shear rate  $< 0.13 \text{ s}^{-1}$  ( $0.05 \text{ s}^{-1}$ ) was used during coalescence test to ensure the occurrence of coalescence during the deformation.

### **FTIR spectrum of the samples before UV exposure**

In Figure S1, the absorbance by neat PE, PE/PP -20/80, PE/PP – 60/40 and PP has been plotted against wave number.

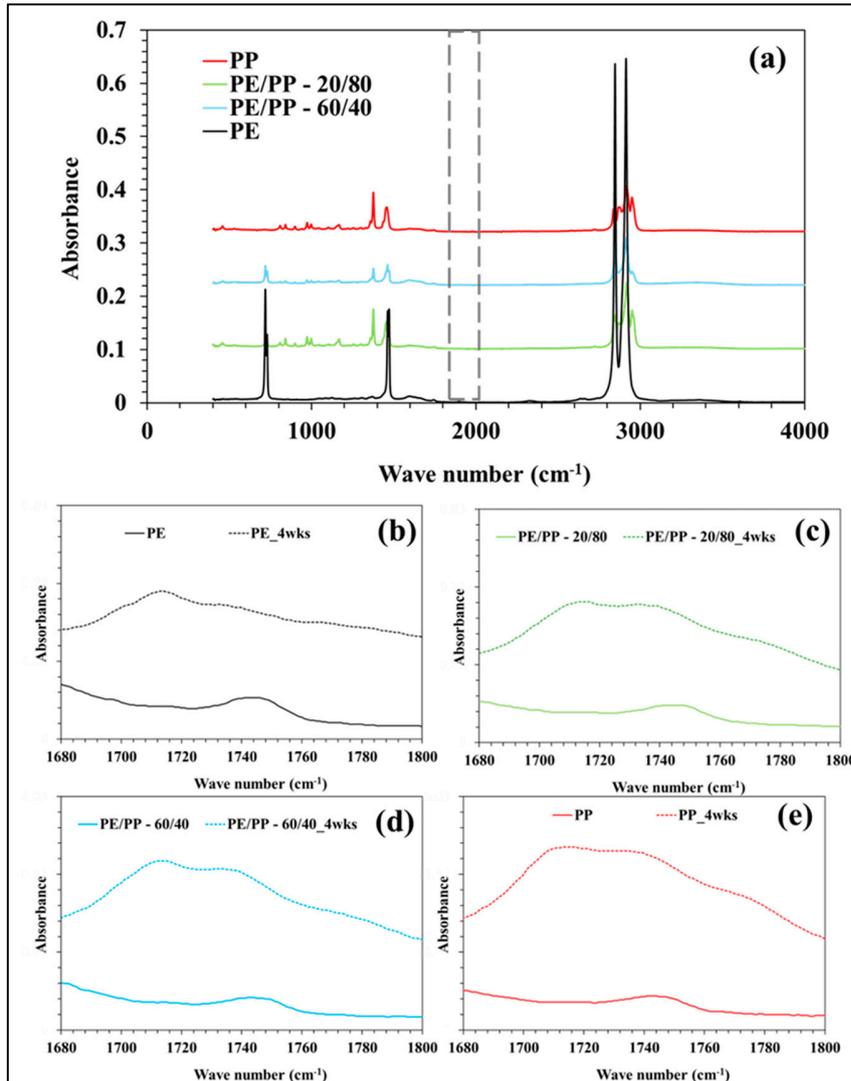


Figure S1: FTIR spectra of neat (a) PE, PE/PP – 20/8, PE/PP – 60/40 and PP samples, and FTIR spectra of neat (b) PE, (c) PE/PP – 20/8, (d) PE/PP – 60/40 and (e) PP samples indicating the carbonyl growth after UV exposure within the wave number range of 1680 to 1800 cm<sup>-1</sup>.

Figure S1 (a) shows that the peak intensity and area under the significant peaks vary based on the composition of the samples.

### Effect of different order of mixing of FLG on the electrical conductivity

Figure S2 illustrates the impact of different FLG mixing strategies on the electrical conductivity of a PE/PP – 60/40 blend composite containing 5 wt.% of FLG. Notably, the composite exhibits significantly higher electrical conductivity when FLG is pre-mixed with the PP phase compared to when it is pre-mixed with the PE phase. It is worth mentioning that FLG has a thermodynamic

preference for PE over PP. Consequently, by pre-mixing FLG with the thermodynamically less favorable PP phase, there is a possibility of capturing FLG at the PE/PP interface as it tends to travel from the PP to the preferred PE phase. This, in turn, has a positive effect on the electrical conductivity of the composite. Similar findings have been reported before [30]. This observation motivated us to carry on the rest of the study with composites, where FLG was pre-mixed with the PP phase.

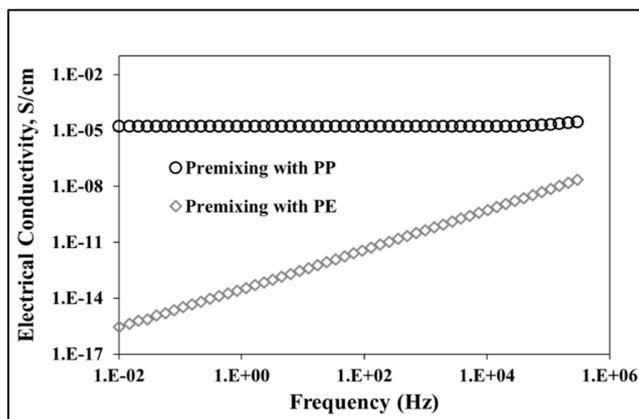


Figure S2: Electrical conductivity of PE/PP – 60/40 blend composite with 5 wt.% FLG; premixed with PP phase and premixed with PE phase.

### Influence of deformation on the morphology of blend composite

Figure S3 illustrates the distribution of PP droplet size (number average droplet radius,  $R_N$ ) in neat and FLG-filled PE/PP – 20/80 blend composite after deformation test. The shear-induced deformation results in the coalesced of the droplets, resulting is bigger droplets. It is worth noting that addition of FLG initially inhibit the coalescence of the PE droplets during processing which result in to smaller droplets of PE in FLG-filled PE/PP – 20/80 composite. However, this coalescence inhibition effect of FLG does not withstand during intense deformation by applied shear.

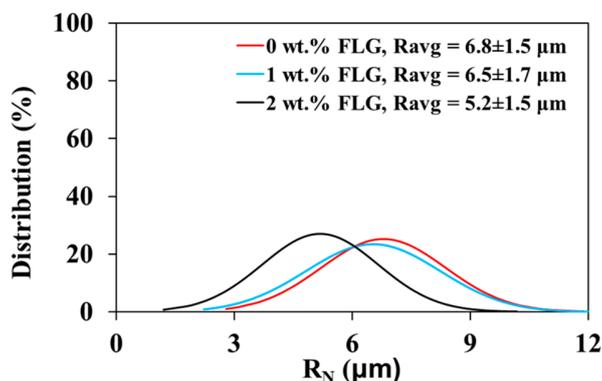


Figure S3: Distribution of PE droplet size in neat and FLG-filled PE/PP – 20/80 blend composite with 1 and 2 wt.% of FLG, respectively, after deformation by 250 strain.

Notably, the filler was initially premixed in the predominant PP phase. Due to thermodynamic affinity, FLG migrated from the PP phase to the PE phase during blend preparation, a phenomenon well-documented by other authors. Since extrusion is a rapid mixing process, a complete shifting of FLG from PP to PE during blend preparation is not possible. During the application of deformation in the rheometer via applied shear/strain, further transfer of FLG to thermodynamically favorable PE droplets from the PP matrix phase took place. In Figure S4, the SEM image of the fractured surface of the PE/PP – 20/80 blend, display a remarkable presence of FLG on the PE phase, corroborating above-mentioned observation.

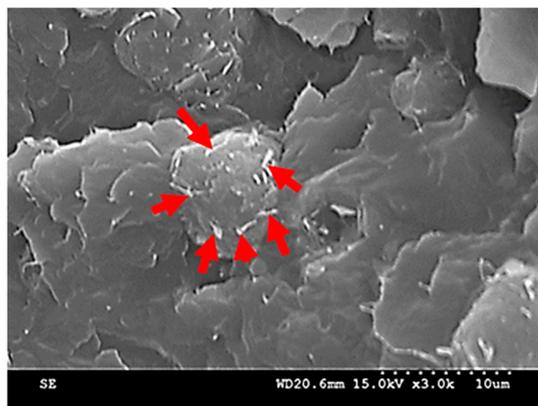


Figure S4: SEM image of 2 wt.% FLG-filled PE/PP – 20/80 blend composite after deformation, red arrows indicate the deformation-driven migration of FLG from PP phase to PE droplet surface.

### Tensile strength of the samples as a function of FLG concentration

In Figure A-I-5, the tensile strength of the PE, PE/PP -20/80, PE/PP – 60/40 and PP compounds has been plotted as a function of the concentration of FLG.

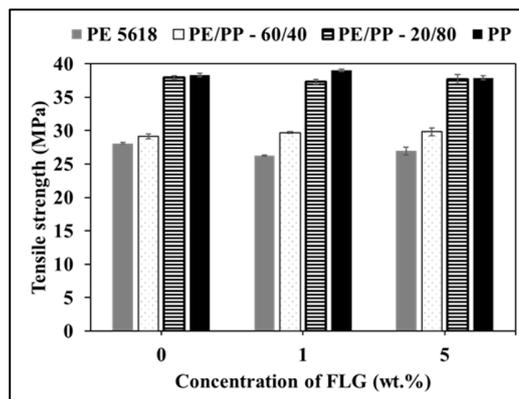


Figure S5: Tensile strength of PE, PE/PP blends and PP composites as a function of FLG concentration.

**SEM images of the surface appearance of a polyolefin sample before UV exposure.**

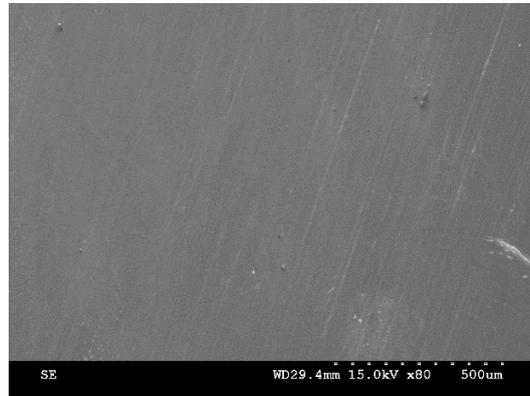


Figure S6: SEM images of the surface appearance of a polyolefin sample before UV exposure.

References

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