

CHARACTERISATION OF THE MECHANICAL AND INTERFACIAL ADHESION PERFORMANCE OF NATURAL FIBRES IN POLYOLEFIN COMPOUNDS FOR LIGHTWEIGHT AUTOMOTIVE APPLICATIONS

J. L. Rudeiros-Fernández^{a*}, J. L. Thomason^a, M. Soliman^b, A. K. Taraiya^c

^aAdvanced Composites Group, Department of Mechanical and Aerospace Engineering, University of Strathclyde, 75 Montrose Street, Glasgow, G1 1XJ, United Kingdom

^bSABIC Technology Center, SABIC, Geleen, The Netherlands

^cSABIC Research & Technology Pvt., SABIC, Bangalore, India.

*Jose.Rudeiros-Fernandez@strath.ac.uk

Keywords: Natural fibres, Mechanical properties, Adhesion Polypropylene, Lightweight-Automotive.

Abstract

This paper presents a study on the measurements of the interfacial adhesion of coir fibre with various polypropylene (PP) matrices along with the mechanical properties of their related injection moulded composites. The interfacial adhesion between coir fibres and homopolymer and copolymer polypropylene, including the effect of coupling agent, was investigated using the pull-out method. The addition of coupling agent significantly increased the interfacial shear strength of coir fibre polypropylene. Results are presented from tensile and impact testing for characterisation of tensile properties and notched and un-notched impact strength of coir reinforced polypropylene. Results showed a positive effect of the coupling agent on stress at yield, Young's modulus and un-notched impact strength.

1. Introduction

In the past few years, the automotive industry has been implementing light-weighting strategies to the design of automotive components. The main target of this approach is to reduce the carbon emissions over vehicle lifetime. To this respect, the applications of mineral fibres and fillers reinforced thermoplastic composites have played a major role. Much of the success in this area is due to improvements in fundamentally understanding the influence of fibre content, fibre diameter, fibre length and fibre-matrix adhesion in the composite. Nevertheless, their relatively high density and high levels of energy required in their production have left room for natural fibres to compete in certain applications. Natural fibres denote a renewable and environmentally friendly reinforcement with low density and high specific strength and stiffness, with an overall reduced carbon footprint required during their production. It has been shown how they have potential to compete with mineral fibres and fillers in certain areas [1], [2]. To this respect, it has been reported how modifications of matrix and/or fibre in natural fibre composites have lower impact on the composites mechanical performance in comparison to mineral fibres composites [3]. Furthermore, from an end-use composite performance point of view, there are some issues – especially impact resistance (both notched and un-notched) – holding back the large-scale implementation of

natural fibre compounds in many applications. Understanding the mechanical behaviour of natural fibres and the influence of fibre-matrix adhesion is essential to successfully improve the mechanical performance of natural fibre reinforced thermoplastic compounds.

This paper presents a study on the measurements of the interfacial shear strength (IFSS) of coir fibre with various polypropylene matrices along with the mechanical properties of their related injection moulded composites. The interfacial adhesion between coir fibres and homopolymer and copolymer polypropylene including the effect of coupling agent was investigated using the pull-out method [4]–[6]. The influence of adhesion on the performance of injection moulded composites will be discussed. Results will be presented from tensile and impact testing for characterisation of modulus, strength and impact strength of coir reinforced polypropylene.

2. Experimental

2.1. Materials

Coir fibre used for this study was provided by Sabic. Coir fibres were used as delivered for pull-out testing. The matrixes used for composites and pull-out testing were homopolymer PP SABIC® PP 579S and copolymer PP SABIC® PP 513MNK10 with a melt flow rate (230 °C and 2.16Kg) of 47 and 70 g/10 min respectively. Maleic anhydride grafted polypropylene (MAPP) Exxelor™ PO 1020 was used as coupling agent.

2.2. Pull Out

The interfacial shear strength of coir fibre and homopolymer and copolymer PP was characterised through pull-out technique. The matrix systems characterised were homopolymer and copolymer along with their respective modified version with 5 wt% MAPP. All the fibres were individually separated until no fraying could be seen with the naked eye. Pull-out samples were prepared in a built in-house rig-mould, which allows the fibres to be aligned perpendicular to the surface of the mould where the polymer pellets were placed. The mould was designed to produce a cylinder-shaped matrix with a fibre embedded along the axis of the cylinder. In order to reduce the thermo-oxidative degradation of PP and coir fibres, samples were prepared in a nitrogen atmosphere. Once the fibres and the polymer were placed on the rig, the assembly was covered with a metallic chamber connected to a nitrogen cylinder. The nitrogen flow was established at 200 ml/min (1.5 bar). The temperature in the mould's surface was monitored by a thermocouple connected to the chamber. The whole system was then placed on a hot plate at 230 °C for 18 minutes. Subsequently, the rig-mould was taken off of the hot plate, the chamber was opened and the samples were left to cool down at room temperature.

Pull out testing was carried out using an Instron 3342 tensile testing machine with a 100 N load cell. Crosshead extension rate was established at 0.5 mm/min. The pull out samples were fixed by a built in-house device, Figure 1, and the fibre was clamped at a gauge length of approximately 5 mm.

Natural fibres' cross section area (CSA) is non-circular and non-constant along fibres' length [7]. In this study, as an initial approximation, the CSA of coir fibres was assumed to be circular.

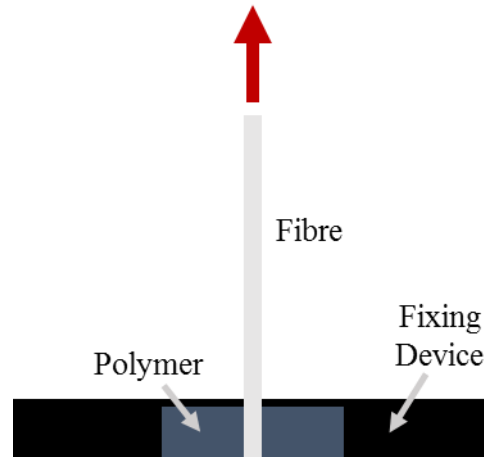


Figure 1. Schematic representation of pull out testing.

For each sample, fibre's diameter (D) and embedded length (L_e) was characterised after the sample was tested. The embedded length was measured on the polymer cylinder using a calliper. Fibres' embedded area were transversally photographed using an Olympus GX51 microscope. For each fibre, diameter was measured using the software ImageJ at three different points. Fibres' average diameter was based on these three measurements. The peak load from the load-displacement curve was used along fibres' diameter and embedded length to calculate a comparative approximation of IFSS (τ), based on the apparent IFSS, equation 1.

$$\tau = \frac{F_{\max}}{\pi D L_e} \quad (1)$$

A minimum of 18 samples, with different fibre diameters and embedded lengths, were tested for each system. Average IFSS values will be presented with 95 % confidence limits. Samples that failed during testing were further investigated using a Field Emission Scanning Electron Microscope (FE-SEM) HITACHI SU-6600.

2.3. Composites

Formulations of both PP (homopolymer and copolymer) and their respective 5 wt% MAPP modifications along with coir fibres loading of 10, 20 and 30 % by weight were made. These formulations were then melt mixed between 180-200 °C using an intermeshing, twin screw extruder of Coperion make (Model ZSK-25). A 25 mm screw diameter was used for compounding and screw rotation was maintained at 300 revolutions per minute (RPM) during the melt mixing. All the formulations were extruded into strands, which subsequently were cut into cylindrical shaped pellets using an inline strand cutter.

Compounded pellets were dried at 80°C for a minimum of 4 hours in a hot air circulated oven. Subsequently, pellets were injection moulded into standard test specimens using LT Demag 100 ton injection moulding machine of L&T Make. Barrel zones were electrically heated and were maintained between 180-200°C and the screw speed was 80 RPM. The mould was maintained at ambient temperature.

Tensile testing of composites was carried out according to ISO 527-2/1A/1 standard, using an Instron 5969 with a 50 KN load cell. Notched and un-notched charpy impact strength was

measured at room temperature according to ISO 179-1:2010 with edgewise impact using a Tinius Olsen Model Impact 503. Notched samples were manufactured based on ISO 179-1/1eAb. All results are illustrated with error bars representing 95% confidence limits. The fracture surface of the composites was observed using a Tungsten Filament Scanning Electron Microscope (W-SEM) HITACHI S-3700.

3. Results and Discussion

3.1. Pull out

Figure 2 illustrates the values of peak load obtained from the load-displacement curves from pull out testing and the embedded area of the fibre in the polymer for PP 579S. Results showed good linear correlation between the two variables. Figure 3 shows the average IFSS values for homopolymer and copolymer systems. The addition of 5 wt% MAPP improved the average IFSS by 54 and 141 % in relation to unmodified system for homopolymer and copolymer respectively.

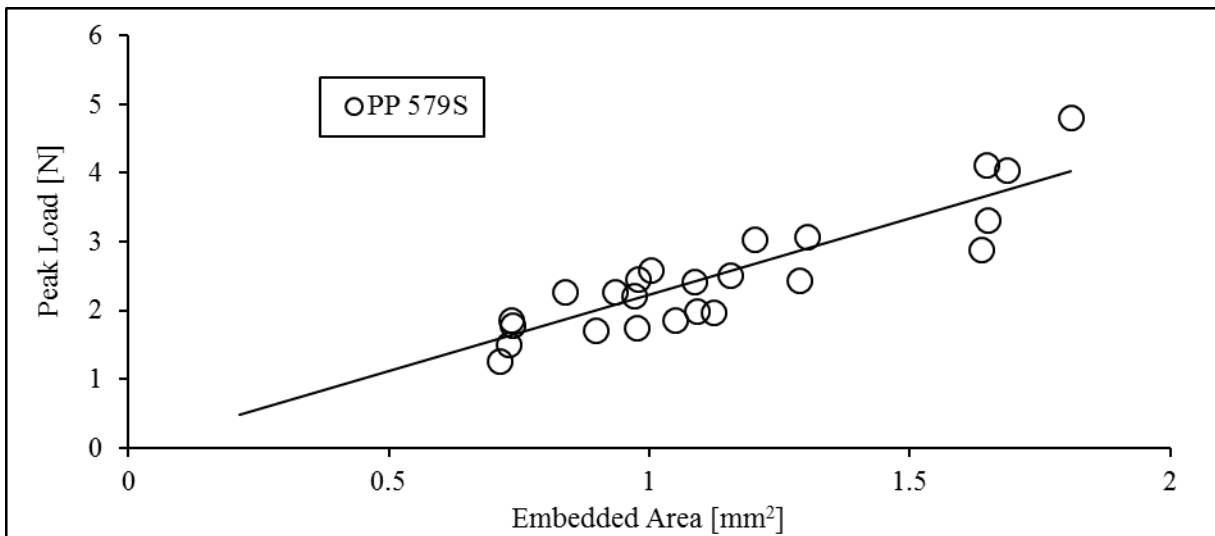


Figure 2. Peak load vs. embedded area from pull out tests on coir fibre-PP579S.

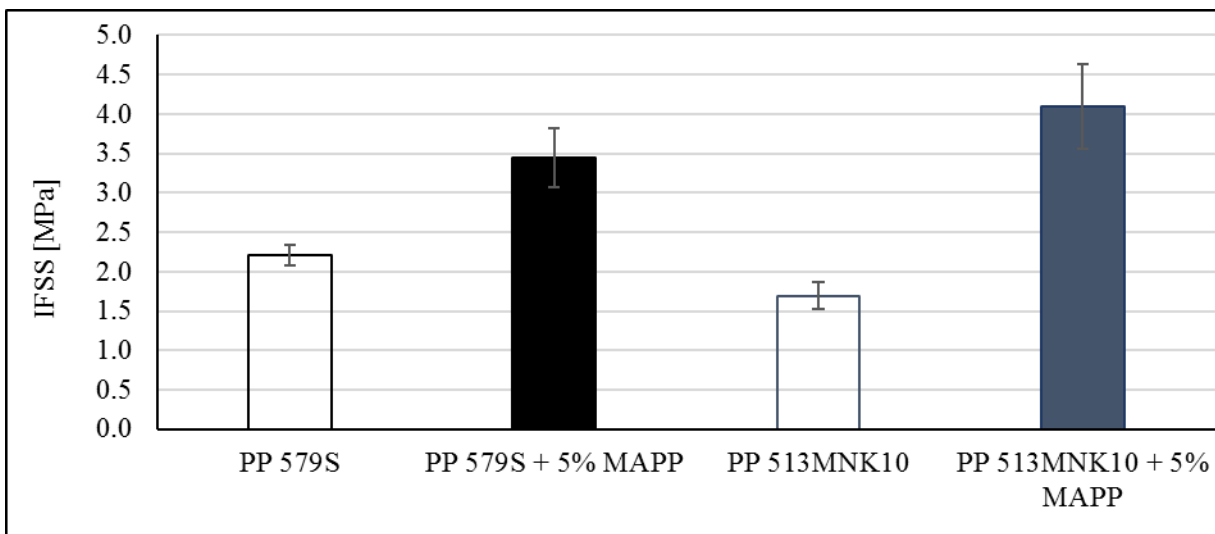


Figure 3. Average IFSS values for coir fibre and the four different systems.

Figure 4 illustrates two different fibre failures at the highest IFSS system (PP 513MNK10 + 5 wt% MAPP). On the left, the fibre failed before complete debonding. On the right side, the fibre failed along its embedded length while the fibre was being pulled out.

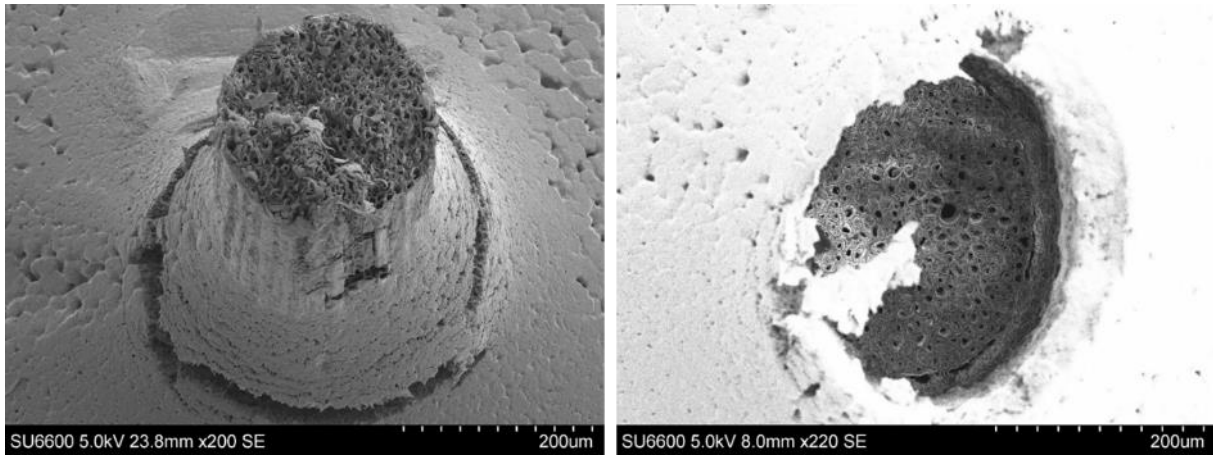


Figure 4. SEM photographs of fibre failure during pull out testing in coir fibre-PP 513MNK10+5 % MAPP.

3.2. Composites

The effects of fibre content on tensile properties are illustrated in Figure 5. Young's modulus increased for increasing fibre load in the four different systems. At 30 wt% fibre load, the addition of 5 wt% MAPP increased the Young's modulus of homopolymer composites by approximately 10 % in relation to unmodified composites. On the other hand, the opposite effect was observed in copolymer composites, where the addition of 5 wt% MAPP slightly decreased the composites Young's modulus. At lower fibre loadings, the addition of MAPP did not produced any significant change.

Regarding the composites yield stress, the trends in relation to fibre loading were not as clear. Unmodified PP 579S composites showed a decreasing stress at yield for increasing fibre load. The addition of MAPP led to the reverse trend, where stress at yield increased for increasing fibre load. In homopolymer composites with a 30 wt% fibre load, the addition of 5 wt% MAPP represented an improvement of approximately 55% over unmodified composites. In the case of copolymer composites, the stress at yield remains almost constant for increasing fibre content. At 30 wt% fibre load, the addition of MAPP led to an improvement of approximately 14% in comparison with unmodified composites. The effect of MAPP at lower fibre loads was negligible. In general terms, homopolymer composites – especially MAPP modified – showed higher stress at yield values. MAPP modified homopolymer and copolymer at 30 wt% fibre load obtained an average stress at yield of 40.15 and 23.03 MPa respectively.

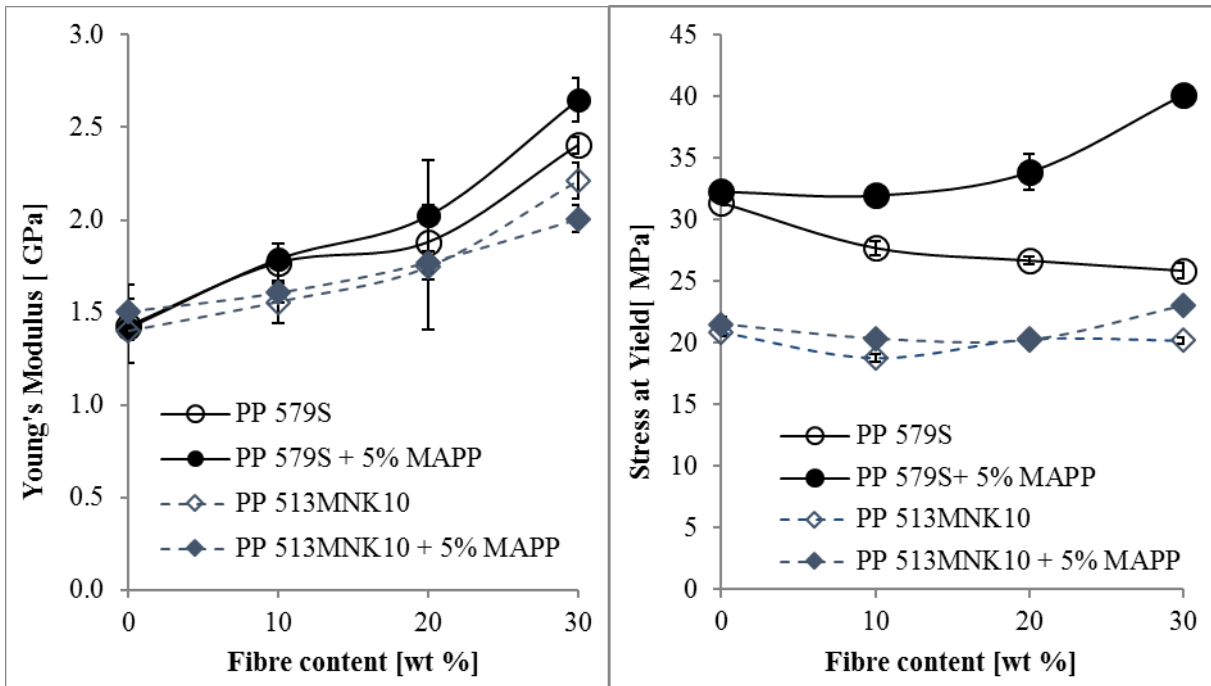


Figure 5. Composites Tensile Properties.

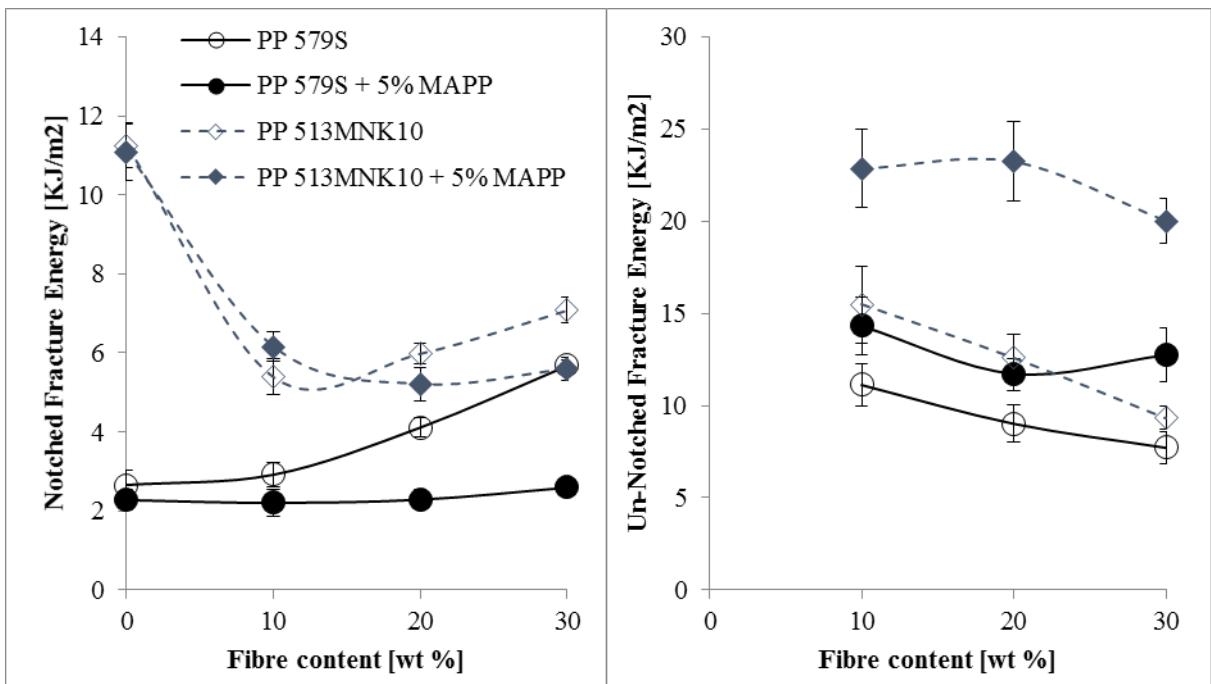


Figure 6. Composites impact properties.

The influence of fibre load and MAPP content on impact properties is showed in Figure 6. For copolymer composites, the notched fracture energy drastically decreased with the initial addition of coir fibre. From this point, the notched fracture energy of unmodified composites increased for increasing fibre load. At 30 wt% fibre load, unmodified composites showed an improvement of almost 27% over MAPP modified composites. The trend was clearer in relation to homopolymer composites. The notched fracture energy of MAPP modified composites remained almost constant for increasing fibre load. On the other hand, unmodified composites fracture energy increased for increasing fibre load. As showed in the pull out testing, the addition of MAPP lead to an increase IFSS. Energy absorbed during crack

propagation in natural fibre thermoplastic is mainly associated with fibre pull out mechanisms. An improvement of the IFSS could lead to a reduced contribution of fibre debonding and pull out and an increase of fibre fracture, which ultimately leads to a lower energy absorbed during fracture.

The average un-notched impact strengths of unreinforced homopolymer, modified homopolymer, copolymer and modified copolymer were approximately 58, 71, 89 and 109 KJ/m² respectively. The addition of coir fibre led to a dramatic drop of the impact strength in all systems. From this point, in general terms the further addition of fibre led to a decreased impact strength. This effect could be attributed to an increased effect of fibres acting as crack initiators. To this regard, MAPP modified composites showed higher fracture energy in comparison with the respective unmodified composites, which could indicate that MAPP increased the energy required in crack initiation. As in the case of notched fracture energy, copolymer composites obtained higher values than the equivalent homopolymer composites. Figure 7 illustrates the effect of MAPP in coir reinforced composites. It can be clearly seen how the addition of MAPP drastically reduced fibre pull out in the case of homopolymer and copolymer composites, observing higher fibre fracture.

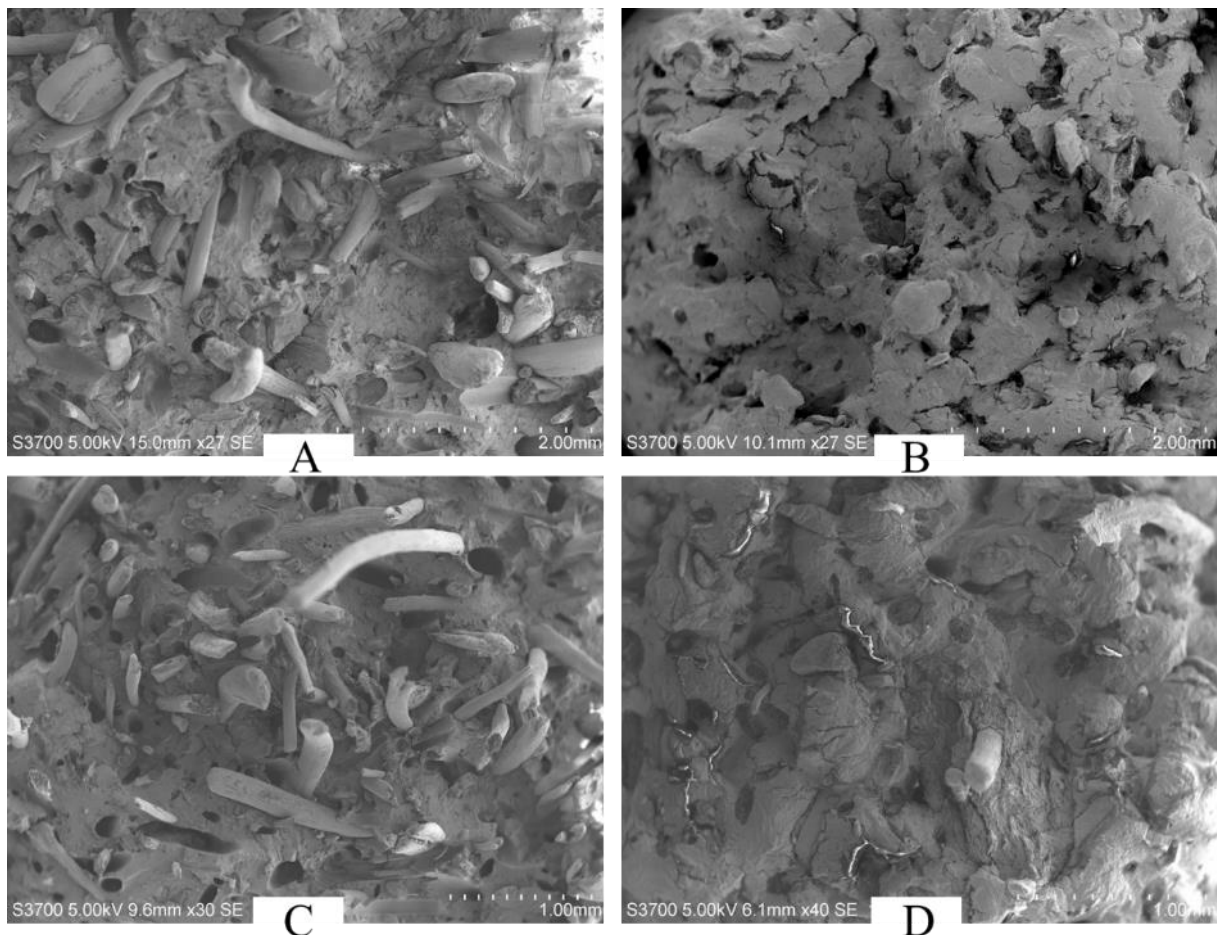


Figure 7. SEM photographs from un-notched composites fracture surfaces (30 wt% fibre load). A – PP 513MKNK10. B – PP 513MKNK10 + 5 % MAPP. C – PP 579S. D – PP 579S + 5 % MAPP.

4. Conclusions

This study characterised the adhesion of coir fibre in four different systems and the mechanical properties of their respective composites. It was demonstrated how the addition of MAPP clearly increases the average values of IFSS for homopolymer and copolymer systems. The values of IFSS increased from 2.2 and 1.7 to 3.4 and 4.1 MPa with the addition of 5 wt% MAPP for homopolymer and copolymer respectively. Further work will establish IFSS values for MAPP concentrations between 0 and 5 wt%. In relation to composites performance, it was shown how in general terms the addition of MAPP had a positive impact on composites' Young's modulus, stress at yield and un-notched impact strength. Regarding the impact performance, it was observed how the addition of MAPP drastically reduced fibre pull out in relation to unmodified composites.

Acknowledgements

The authors gratefully thank SABIC for the materials and financial support. The AMRL's assistance with the SEM micrographs is also greatly acknowledged.

References

- [1] A. K. Bledzki and J. Gassan. Composites reinforced with cellulose based fibres. *Progress in Polymer Science*, 24:221-274, 1999.
- [2] K. Goda and Y. Cao. Research and development of fully green composites reinforced with natural fibers. *Journal of Solid Mechanics and Materials Engineering*, 1(9):1073-1084, 2007.
- [3] J. L. Thomason. Dependence of interfacial strength on the anisotropic fiber properties of jute reinforced composites. *Polymer Composites*, 31(9):1525-1534, 2010.
- [4] A. Valadez-Gonzalez, J. M. Cervantes-Uc, R. Olayo, and P. J. Herrera-Franco. Effect of fiber surface treatment on the fiber – matrix bond strength of natural fiber reinforced composites. *Composites Part B: Engineering*, 30(3):309-320, 1999.
- [5] Y. Li, K. L. Pickering, and R. L. Farrell. Determination of interfacial shear strength of white rot fungi treated hemp fibre reinforced polypropylene. *Composites Science and Technology*, 69(7):1165-1171, 2009.
- [6] L. Yang and J. L. Thomason. Interface strength in glass fibre–polypropylene measured using the fibre pull-out and microbond methods. *Composites Part A: Applied Science and Manufacturing*, 41(9):1077-1083, 2010.
- [7] J. L. Thomason, J. Carruthers, J. Kelly, and G. Johnson. Fibre cross-section determination and variability in sisal and flax and its effects on fibre performance characterisation. *Composites Science and Technology*, 71(7):1008-1015, 2011.