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Mechanical and Rheological Characterization of PA6 and ABS Blends-With and Without Short Glass Fiber

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Abstract: Polyamide-6 (PA6), acrylonitrile-butadiene-styrene and their blends are an important class of engineering thermoplastics that are widely used for electronic and automotive industries. Many efforts have been done to improve the properties of both pure components and the blends. It was for this reason that the mechanical and rheological properties of PA6/ABS blend systems compatibilised by acrylonitrile-butadiene-styrene-maleic anhydride was studied. Short glass fibre also was used to enhance the compatibilised blends. The systems were prepared using a twin screw extruder. The specimens for mechanical test were moulded using injection moulding. The compatibilised blends showed higher mechanical properties compared to uncompatibilised blend. However the impact strength reduced with the introduction of short glass fibre in the blends. The processing properties of the materials have been investigated using dynamic rheometers. The rheological measurements confirmed the increased interaction between the blend components with the incorporation of compatibiliser. The incorporation of SGF into the compatibilised blends increased the storage and loss modulus as well as the viscosity of the blends.

Key words: PA6, ABS, Short glass fibre, dynamic rheological, PA6/ABS composites

INTRODUCTION

Polyamide 6 (PA6) and acrylonitrile-butadiene-styrene (ABS) are widely used engineering thermoplastics. PA6 are a particularly attractive class of polymers due to the good strength and stiffness, low friction and excellent chemical and wear resistance. The beneficial properties have led to the wide range of usage especially in automotive, electrical and mechanical application. However, PA6 has some disadvantages associated with its processing instability such as high mould shrinkage and dimensional stability, due to its inherent properties of rapid crystallization (Jang and Kim, 2000) and high moisture sensitive because of its hygroscopic nature (Acierno and Puyvelde, 2005). Therefore, the strong reasons behind blending ABS with PA6 is that the low price of ABS compared to PA6, ease of processing, low water absorption and high impact strength. ABS has some strength over PA6 in low mould shrinkage, even if other mechanical and thermal properties are not as good as PA6.

Blending of PA6 with ABS offers an interesting route to achieve new materials with promising property combinations by optimizing their properties. Essentially, blends of PA6 and ABS are immiscible throughout the whole range of compositions and exhibit low impact toughness because the butadiene particles formed during the melt blending process are relatively large (Tjong *et al.*, 2002). Also, the immiscibility of PA6 and ABS in the melt state leads to the blends with poor interfacial adhesion, unstable phase morphology and low mechanical properties of the blends, which is limited their practical use. Therefore, reactive compatibilisation is the most promising way to enhance the interfacial adhesion and improve the compatibility of the blends. During melt blending, the compatibiliser with anhydride functional group can react with amine end-group of PA6, resulting in the reduction of interfacial tension and improvement of phase dispersion and adhesion between PA6 and ABS phases. The ABS-grafted maleic anhydride is expected to act as reactive compatibiliser between PA6 and ABS. Aoki and Watanabe (1992) reported that the addition of certain

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the addition of certain amount of modified ABS-ABS grafted with maleic anhydride-to the PA6 has improved the dispersion phase. However, the addition of ABS leads to a reduction in strength and stiffness. In contrast, reinforcing thermoplastics by Short Glass Fibre (SGF) improved both strength and modulus; but a high content of SGF is necessary to achieve high strength and high stiffness. Unfortunately, there is considerable loss in toughness and ductility when these SGF are incorporated in the ductile polymer matrices.

Rheological effect on the incorporation of compatibiliser and SGF into PA6/ABS blends is a subject that has not been discussed in depth. Generally, polymer rheological properties help to formulate a polymer system in respect to its processing characteristics. In addition, polymer rheological study will give an insight into physical properties of the system because there is interplay between the processing conditions, structure and properties (Varma and Dhar, 1989). In this study, the rheological properties of the blends have been investigated by means of melt flow index and dynamic rheometer.

MATERIALS AND METHODS

Samples preparation: Both Super high impact ABS resin and PA 6 with the grade of Amilan CM1017 was supplied by Toray Plastics (Malaysia) Sdn. Bhd. with a specified melt flow index of 14 and 35 g/10 min (at 220°C and 10 kg load). While, compatibilizer was obtained from Polyram, Ram-O Industries Limited, with a brand name Bondyram® 6000. This compatibilizer is a maleic anhydride grafted ABS (ABS-g-MAH) with melt index and density of ABS-g-MAH are 8 g/10 min (at 220°C and 2.16 kg load) and 1.05 g cm⁻³, respectively. Glass fibres were obtained from Taiwan Glass Industries Corporation, named as chopped strand TG183 with a filament diameter of 10 µm and an average length of 3.2 mm.

PA6, ABS, ABS-g-MAH pellets and SGF were dried at 80°C for 24 h. The PA6/ABS ratios were 60/40. The compatibilizer concentrations were 1, 2, 3 and 5% wt. of the matrix. The composites were reinforced with 10, 20 and 30 wt.% SGF, respectively. The blends and composites were prepared by melt mixing using a Brabender twin-screw extruder at a speed of 90 rpm and the temperature profile adopted was 220/230/240/250°C. The extruded strands were then air-dried and palletised. The specimens for mechanical characterization experiments were moulded by using injection moulding.

Mechanical properties measurements: The tensile tests for blends and composites were conducted at room

temperature at a crosshead rate of 5 mm/min with Instron Model 5567 computer-controlled testing machine. Tensile strength, modulus and elongation at break were simultaneously recorded. Izod impact strength was measured according to ASTM D256 using impact tester. 2.54 mm notch was generated on the sample surface in the thickness of 3.4 mm. Measurements of mechanical properties were performed 10 times per each and the average value was obtained for its determination.

Dynamic rheological testing: Small-amplitude oscillatory shear measurements were performed on a Rheostress RC501 in the parallel-plate geometry with diameter of 25 and 1.5 mm thickness. The dynamic viscoelastic properties were determined with frequencies ranging from 0.05 to 100 rad/s at 230°C, using strain and stress values determined the linear viscoelastic region. All the frequency sweeps were repeated at least three times to confirm the reproducibility.

RESULTS AND DISCUSSION

Tensile properties of polymer blends: Figure 1 and 2 show the effect of ABS-g-MAH content on the tensile properties of PA6/ABS blends. The tensile modulus of PA6/ABS blends increased with increasing of ABS-g-MAH ratio in the blends, especially at low filling rates. It shows that tensile modulus increased drastically from 1740 to 2250 MPa with the increment of 1 wt.% of ABS-g-MAH content. However, the tensile modulus decreased at 2% of ABS-g-MAH to about 2040 MPa. The addition of 3 to 5 wt.% of ABS-g-MAH content almost resulted in constant modulus. This is believed due to ABS-g-MAH became excess able amount and has not improved much in compatibility of PA6/ABS blends. This trend could also be found similar in tensile strength as shown in Fig. 2.

Figure 3 shows the elongation at break of PA6/ABS blends. The elongation can be used to represent the behaviour of toughness. It can be seen at lower amount of compatibilizer (from 0 to 3 wt.%), it seems that the elongation increased gradually. However the increase in percentage of ABS-g-MAH did not show any improvement and considered as maximum elongation about 9%. These results are in agreement with the result of impact strength (Fig. 7). Generally, the low elongation is attributed to the phase separation and resulting less compatibility between PA6 and ABS phases even with the presence of ABS-g-MAH as compatibiliser.

Tensile properties of polymer composites: Figure 4 and 5 show a tensile modulus and tensile strength of polymer

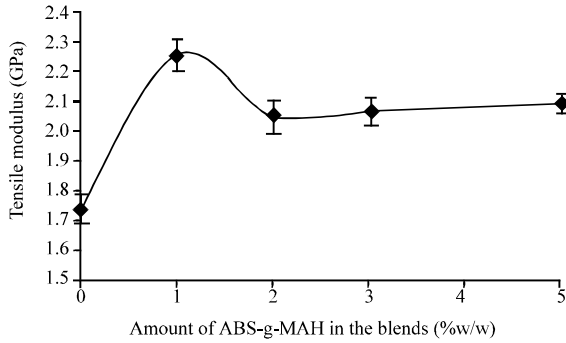


Fig. 1: Tensile modulus of PA6/ABS blends versus amount of ABS-g-MAH

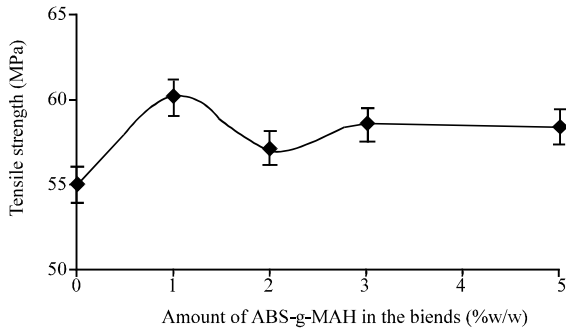


Fig. 2: Tensile strength of PA6/ABS blends versus amount of ABS-g-MAH

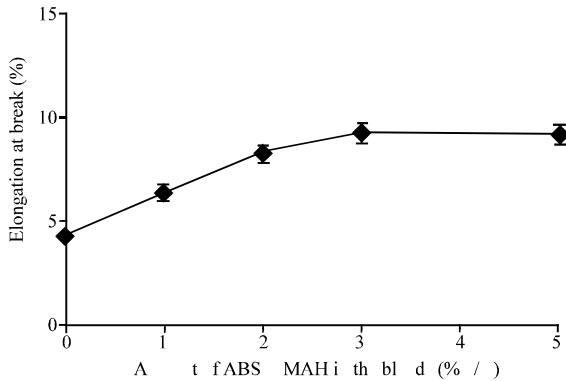


Fig. 3: Elongation at break of PA6/ABS blends versus amount of ABS-g-MAH

composites, respectively. As expected, it can be seen from figures that the tensile strength and modulus were effectively enhanced by the addition of short glass fibre. Similar result was also found by other researchers (Fu *et al.*, 2006). When the amount of SGF reached to 30 wt%, the tensile modulus increased drastically, but not in the case of tensile strength which was increased gradually. However, as shown Fig. 6, the elongation at

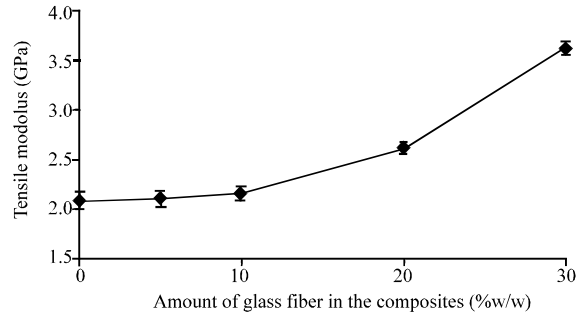


Fig. 4: Tensile modulus of PA6/ABS composites versus amount of short glass fibre

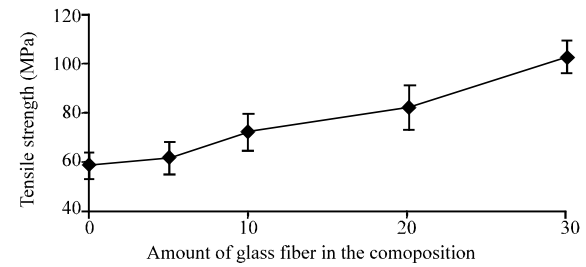


Fig. 5: Tensile strength of PA6/ABS composites versus amount of short glass fibre

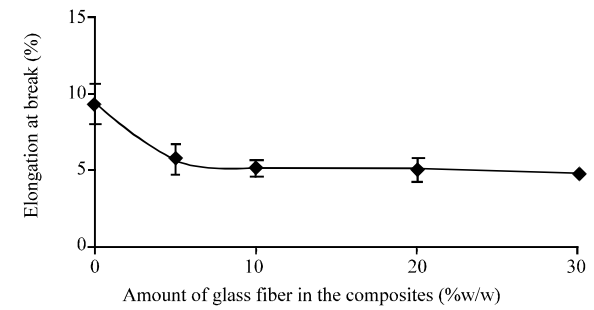


Fig. 6: Elongation at break of PA6/ABS composites versus amount of short glass fibre

break of composites decreased tremendously by increasing SGF into the composites and remained at constant value when the amount of SGF is beyond 10 wt%. Therefore, the incorporation of SGF did not improve the toughness of the composites.

Impact strength of the polymer blends: Notched Izod Impact values are plotted as a function of ABS-g-MAH content in PA6/ABS blends as shown in Fig. 7. It can be observed that the impact strength increased as the percentage of ABS-g-MAH was increased. Upon the incorporation of 2 wt.% of ABS-g-MAH in the PA6/ABS

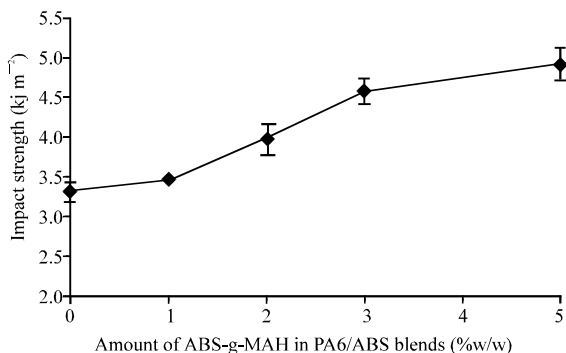


Fig. 7: Impact strength of PA6/ABS blends versus amount of ABS-g-MAH

blends, the impact strength increased drastically and reached almost a constant value when the amount of ABS-g-MAH beyond 3 wt.%. The improvement in impact properties, in the present of ABS-g-MAH was believed to be attributed by the compatibilisation of PA6 and ABS through the chemical reaction between the free terminal of amine end group of PA6 and the anhydride function in ABS-g-MAH. Such reaction has been reported in many literatures (Kudva *et al.*, 1998; Sun *et al.*, 2005).

Impact strength of polymer composites: The impact result of the composites is shown in Fig. 8 at different amount of SGF. It can be observed that when the amount of SGF was increased to about 10 wt.%, the impact strength decreased drastically. The drastically reduction could be due to matrix embrittlement (Kannan and Misra, 1994). Beyond this point, the small increment in impact strength with further addition about 20 wt.% SGF to be due to the fact that the fibres are shorter length and shorter fibres are effective energy absorbers by pull-out and debonding. The impact strength decreased with further increase of SGF to about 30 wt.% could be due to poor interfacial adhesion.

Rheological properties of polymer blends: Figure 9 and 10 show the relationship between dynamic storage modulus G' , dynamic loss modulus G'' and frequencies ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C. It can be observed that G'' is greater than the G' for all components at low frequencies. It also can be seen that within the ω range tested, curves of G' and G'' versus ω for blends were located between the virgin PA6 and ABS and almost followed a linear mixing rule. It can be found that, the virgin PA6 obeyed the linear viscoelasticity model (Feery, 1980; Bose *et al.*, 2007) but not for the blends. In addition, compatibilised PA6/ABS blends, G' and G'' increased significantly with increasing of ABS-g-

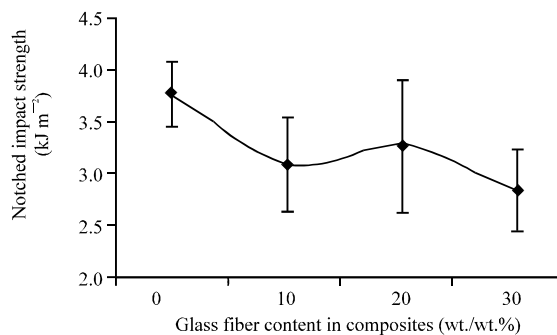


Fig. 8: Impact strength of PA6/ABS composites versus amount of short glass fibre

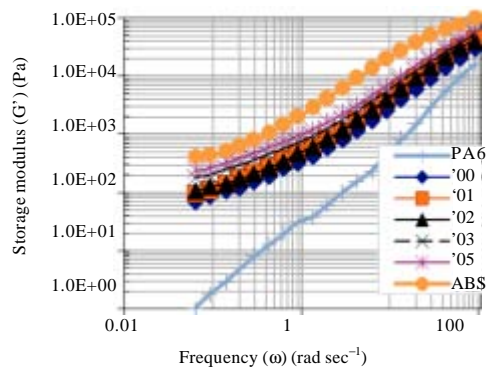


Fig. 9: Relationship between the dynamic storage modulus G' and frequencies, ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C

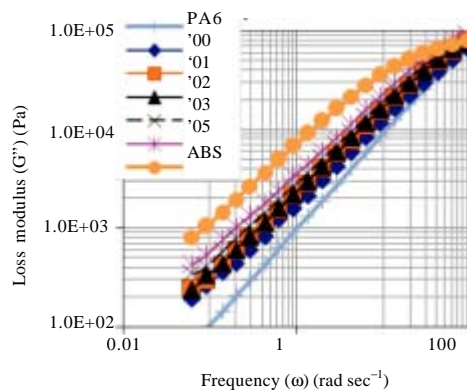


Fig. 10: Relationship between the dynamic loss modulus G'' and frequencies, ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C

MAH content. On the other hand, the slopes of the blends at low frequencies appeared to be slightly increase compared to the virgin PA6. This is believed due to the

fact that the dynamic rheological behaviour of blends mainly influenced by PA6 matrix as a continuous phase. It is also indicates the new structure formation in these blends. This result is in agreement with the previous study (Gribben *et al.*, 2005; Shi *et al.*, 2002).

It is well known that the end group of PA6 is amine and capable of reacting with maleic anhydride groups in ABS-g-MAH and forms grafted co-polymer at interface of the blends. These interactions can stabilise the interface by reducing the interfacial tension, resulting in enhancement of the interfacial viscosity and adhesion. This is the reason why those compatibilised PA6/ABS blends exhibited higher G' and G'' than uncompatibilised PA6/ABS blends. The rheological behaviour of the blends at low ω region seems to gradually deviated from that of virgin PA6 which is responsible for the existence of heterogeneous structure appeared. This is attributed to the result of increasing in complex viscosity due to enhancement of macromolecular.

Figure 11 shows the relationship between the complex viscosity η^* and frequency, ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C. It is evident that the complex viscosity increased with increasing the frequencies, indicating that the melt of these polymers belong to pseudoplastics fluids and have the flow properties of shear thinning. However the virgin of PA6 had slightly a constant complex viscosity and showed a Newtonian flow behaviour (Bose *et al.*, 2007) at low frequencies followed by a shear-thinning regime at moderate frequencies. The shear thinning of virgin PA6 was less than virgin ABS and other blends. It can be explained that the molecular chains of PA6 are rigid and hence hard to tangle each other, whereas virgin ABS with flexible chains possesses much more entanglements than PA6 (Ma *et al.*, 2007; Dreval *et al.*, 2006).

With the incorporation of ABS-g-MAH, it can be found that the complex viscosity increased with increasing of ABS-g-MAH content at low frequencies range due to improvements of phase structure and interfacial adhesion between PA6 and ABS. It is believed to be due to the graf-formation by reacting amine end-group of PA6 with maleic anhydride in ABS-g-MAH. These results are in agreement with the previous study (Khan *et al.*, 2005). These results also confirmed by the result of $\tan \omega$ as shown in Fig. 12, which is, the more amount of ABS-g-MAH introduced into the blends, the lower of $\tan \omega$ peak to be and thus less energy dissipated by the polymer systems. Therefore, by the addition of short glass fiber, the reduction of $\tan \omega$ peak can also be associated with the decreased mobility of the polymer chains. Relationship between the dynamic storage modulus G'' and frequencies, ω for PA6/ABS composites at different amount of short glass fibre at 230°C is shown in Fig. 13.

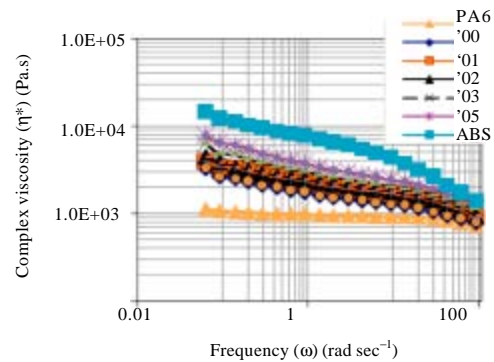


Fig. 11: Relationship between the complex viscosity η^* and frequencies, ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C

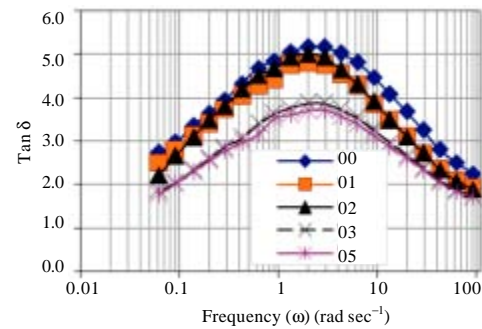


Fig. 12: Relationship between the $\tan \delta$ and frequencies, ω for PA6/ABS blends at different amount of ABS-g-MAH at 230°C

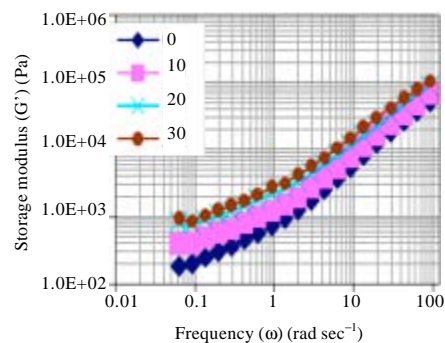


Fig. 13: Relationship between the dynamic storage modulus G'' and frequencies, ω for PA6/ABS composites at different amount of short glass fibre at 230°C

Rheological properties of polymer composites: The G' and G'' results for the polymer composites and polymer blends are shown in Fig. 14 and 15, respectively. Again, rise in storage modulus with increasing SGF concentration

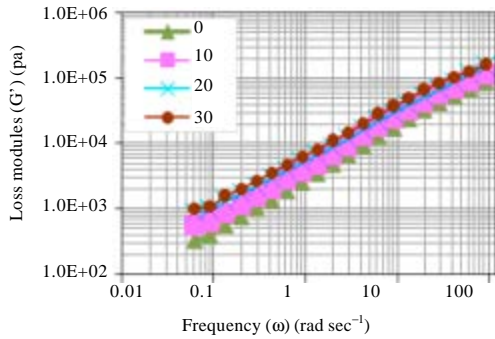


Fig. 14: Relationship between the dynamic loss modulus G'' and frequency, ω for PA6/ABS composites at different amount of short glass fibre at 230°C

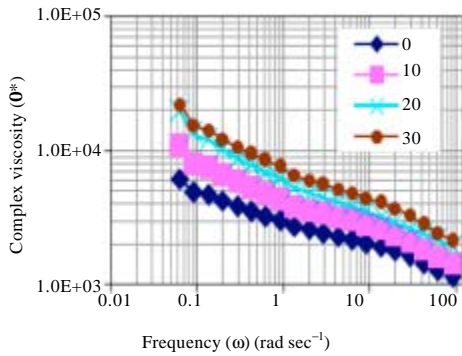


Fig. 15: Relationship between the complex viscosity and frequencies, ω for PA6/ABS composites at different amount of short glass fibre at 230°C

was observed at all frequencies. Moreover, these polymer composites can be categorised as liquid-like material, because the G'' is greater than G' at all range of frequencies study regardless the amount of SGF in the systems. This phenomenon is due to SGF acted as a ‘lubricant’ and caused the polymer phase of PA6 and ABS easily slip at interphase and interface level.

Figure 16 represents the relationship between complex viscosity and frequencies, ω for PA6/ABS composites containing different amount ranging from 0 to 30% of SGF at 230°C. It can be seen that the trend in complex viscosity growth associated with SGF loading. At all frequencies, the complex viscosity of the polymer composites was larger than unfilled PA6/ABS blends. It can also be found, that the complex viscosity decreased with the increasing of frequencies. This observation is due to the deformation of the SGF at high frequencies and its associated contribution to an increase in fluidity of the matrix phase. When more SGF added into the blends, the

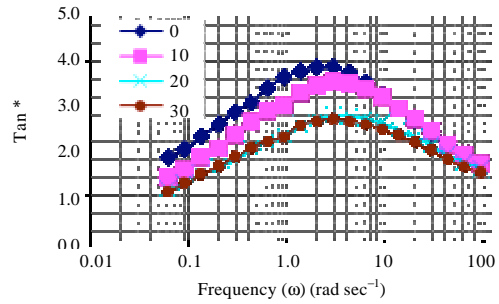


Fig. 16: Relationship between the $\tan \delta$ and frequencies, ω for PA6/ABS composites at different amount of short glass fibre at 230°C

SGF droplets began to grow and coalesce, causing an increase in the complex viscosity (Guschl and Otaigbe, 2003). Generally, all the blend systems showed a non-Newtonian behaviour at all frequencies.

CONCLUSION

The mechanical properties of PA6/ABS blends and short glass fibre reinforced PA6/ABS composites have been studied considering the effect of ABS-g-MAH and SGF content, respectively. The amount of ABS-g-MAH in the PA6/ABS blends was found to have significant effects on the mechanical and rheological properties as well as the ratio of SGF in the polymer composites. It was exhibited that the tensile and flexural properties improved upon the addition of ABS-g-MAH at its various compositions into the PA6/ABS blends, while the toughness properties decreased with introduction of SGF in the composites. The processing properties of the materials have been investigated by using dynamic rheometers. The rheological measurements confirmed the increased in interaction between the blends components, especially with ABS-g-MAH as a compatibiliser. The incorporation of SGF into the compatibilised blends increased the storage and loss modulus as well as the viscosity of the blends.

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