FLEXURAL PROPERTIES AND THERMAL CONDUCTIVITY OF ALUMINIUM NITRIDE FILLED POLYPROPYLENE COMPOSITE

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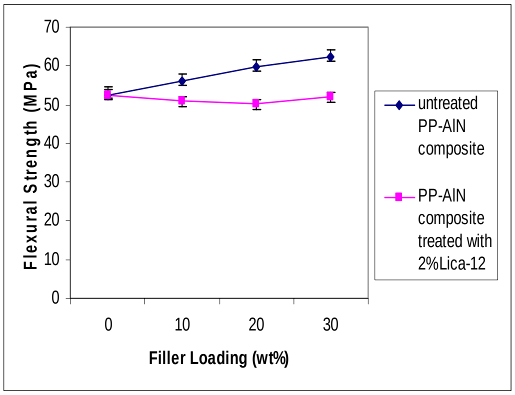
Abstract
Aluminium Nitride (AlN) filled Polypropylene (PP) composites are fabricated, where the compounding is carried out by using Z- blade mixer with filler loadings of 0wt%, 10wt%, 20wt% and 30wt%. Composite sheets produced from hot press molding process are prepared for testing samples. The effect of filler loadings and incorporation of titanate coupling agent on flexural properties and thermal conductivity are studied. The results demonstrate that, an increase in filler loading is found to increase the flexural modulus, flexural strength and thermal conductivity of AlN filled PP composites. By adding 2% titanate coupling agent (Lica 12), the decrease of flexural strength and flexural modulus with increasing filler loadings can be observed in 10%AlN-90%PP composite. However, the addition of 5% titanate coupling agent in 10%AlN-wt%PP composite allowed the increment of flexural strength and flexural modulus and assisted in greater increment of thermal conductivity of the composites.

Introduction
Thermally conducting but electrically insulating polymer-matrix composites is increasingly important for electronic packaging because the heat dissipation ability limits the reliability, performance and miniaturization of electronics [4]. In order to produce this kind of materials, ceramic fillers such as aluminum nitride, silicon carbide and alumina is used. That explained why ceramic powder reinforced polymer materials have been used extensively as electronic packaging materials [1, 2]. This work is focused on AlN as filler in PP matrix due to its combination of high thermal conductivity, low dielectric constant and low cost. Nowadays, a large and steadily growing body of published research is available demonstrating the ability of titanate coupling agents to enhance the properties of composites combining a wide range of fillers and polymers [8]. Thus, the research studied the effect of titanate coupling agent (Lica-12) addition on the flexural properties and thermal conductivity of PP-AlN composite. In addition, the possibility to produce effective thermal conductivity at low filler concentrations is also investigated.

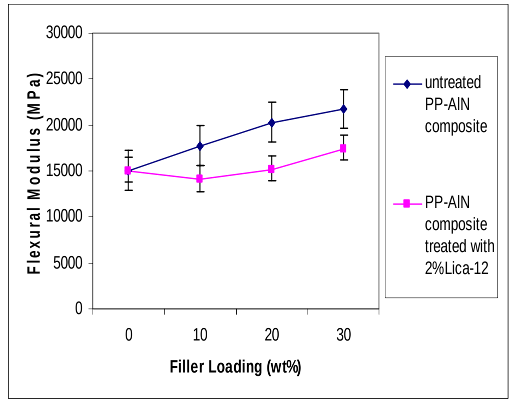
Experimental Methodology
Composite polypropylene filled with aluminum nitride (AlN-PP) was prepared by using melt mix method. AlN-PP composite was prepared by using Z-blade mixer with the rotation speed of 35 rpm at 190°C and then compressed by using hot press molding with the pressure of 165 kg/cm^2 at 190°C. Composites with different weight percentage of filler contents varying from 0wt% to 30wt% were prepared. In order to improve properties of composites consisting of PP filled with AlN particles, the particles surface was treated with a titanate coupling agent. The effect of titanate coupling agent on flexural properties and thermal conductivity of AlN-PP composite were measured as a function of filler loading and coupling agent content.

Results and Discussion
Flexural Properties
Figure 1a and 1b present the effect of filler content in flexural strength and flexural modulus for aluminum filled polypropylene (AlN-PP) composite respectively. Both flexural strength and flexural modulus of the composite exhibited an increase with increasing filler loading. The overall increase in the flexural modulus with increasing filler content is expected since the addition of filler in particle form increases the stiffness of the composites [9]. PP-AlN composite treated with 2%Lica-12 showed the lowering of flexural strength and flexural modulus especially at 10%AlN-90%PP. The factor that might be contributed to this is because titanate couplers also act as plasticizer in addition to improving filler dispersion [8]. The plasticizing effect of titanate might have reduced the intermolecular forces in the matrix, and consequently produces the reductions in stiffness [9]. However, both flexural strength and flexural modulus exhibit and increment at 30%AlN-70%PP. Apparently, higher filler content would tend to reduce the plasticizing effect of the coupling agent.

Figure 2a and Figure 2b shows the effect of amount percent of titanate coupling agent (Lica-12) on 10%AlN-90%PP composite. As compared with AlN-PP composite untreated with coupling agent, the AlN-PP treated with Lica-12 showed lower value of flexural strength. However, by increasing the coupling agent content up to 5%, the trend can be seen as increasing. From Figure 2b, we can conclude that by increasing the amount of coupling agent content up to 5% can slightly increase the flexural modulus of 10%-AlN-90%PP composite.

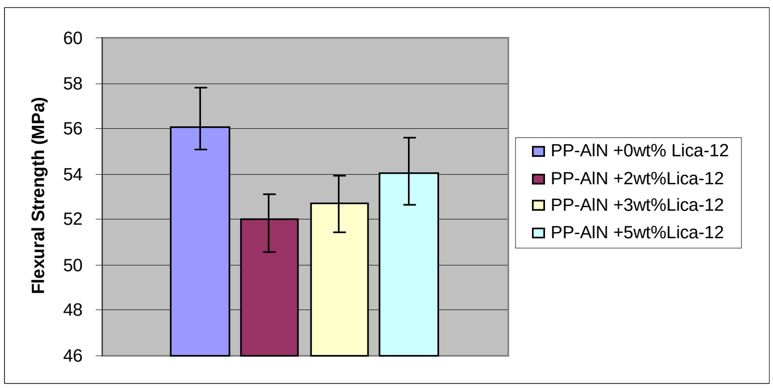


(a)

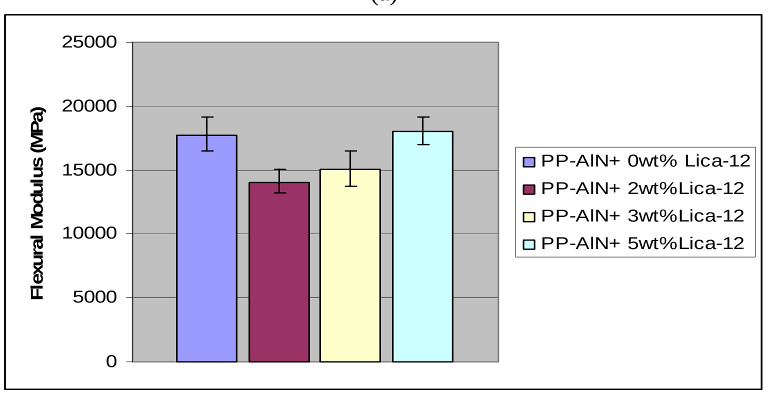


(b)

Figure 1 a) Flexural strength and b) flexural modulus of PP-AlN composite in different filler loadings.



(a)



(b)

Figure 2 a) Flexural strength and b) flexural modulus of 10%AlN-90%PP composite with different Lica-12 content.

Thermal conductivity

Figure 3 shows the effect of filler loadings and the addition of 2% titanate coupling agent (Lica-12) on thermal conductivity of PP-AlN composites. As expected, the thermal conductivity for AlN filled PP composites increase with increasing filler loading. Previous study [1] on polystyrene-AlN composites also showed the same trend of thermal conductivity with increasing of filler loading. The thermal conductivity of AlN is much larger than that of PP, so the addition of AlN filler to the PP matrix will result in an increased in the thermal conductivity of the composite. For the increase of thermal conductivity as increasing filler loadings, the reason may be because at higher particle content, the filler tends to form agglomerates and conductive chains resulting in a rapid increase in thermal conductivity [7]. The increase in thermal conductivity is also due to the decrease in the filler-matrix thermal contact resistance through the improvement of the interface between matrix and particle [4]. From SEM micrograph study, it can be seen that the filler distribution and AlN particle contact is improved by increasing the filler loading. The condition of the filler distribution is gradually enhancing as the filler loading increase as shown in Figure 4. This explains why sample 30%AlN-70%PP showed the highest thermal conductivity value among the other samples. Good filler distribution in the matrix improves the contact between filler and filler thus providing good channel for heat to transfer [10].

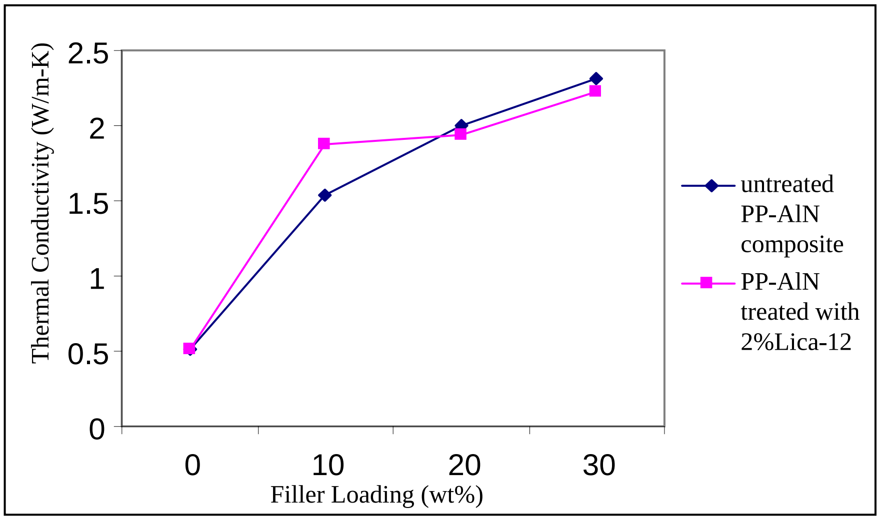
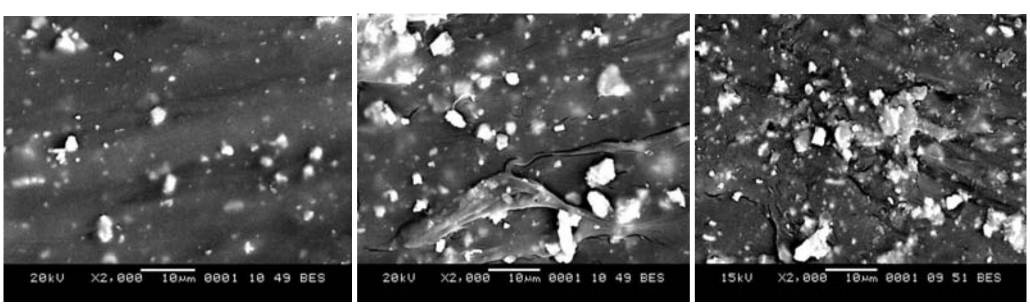


Figure 3: Effect of filler loadings on thermal conductivity of PP-AlN composites.

PP-AlN composite treated with Lica-12 demonstrate an increasing in thermal conductivity with increasing filler loadings (same trend as untreated one). However for the treated composites, the significant increments of thermal conductivity can be clearly seen to PP added with 10% AlN while for PP added with 20% and 30% AlN filler showed a slight decrease in thermal conductivity compared to untreated one. Apparently, the titanate coupling agent is only assist for the low weight fraction filler (10%AlN) composite. By considering only for 10%AlN-90%PP composite, the experiment continued for determining the best amount fraction of coupling agent (Lica-12), which can give the highest value of thermal conductivity for the composite. Figure 5 shows the result.



(a) (b) (c)

Figure 4: SEM micrograph for a) 90%PP-10%AlN b) 80%PP-20%AlN and c) 70%PP-30%AlN with magnification 2000x

It can be seen that the best composition of Lica-12 that produce the highest thermal conductivity in 10%AlN-90%PP composite is 5%. This result can be related with the effect of titanate coupling agent (Lica-12). The addition of coupling agent enables the matrix to form stronger bonding with the filler forming a 'molecular bridge' between the two, preventing gap where air can trapped at the interface and enhanced heat transfer between the matrix and the filler at the interface [6]. Coupling agent can enhance the compatibility between the resin and the filler thus enhance the dispersion of the fillers, improved wet-out between resin matrix and filler which in turn improve the thermal conductivity ability of the polymer composites [10]. It has been postulated that titanate coupling agent improves the compatibility of inorganic filler with polymer, by enhancing their interfacial adhesion [8]. Comparison between morphology of treated and untreated PP-AlN composite is shown in Figure 6. Figure 6a shows the poor dispersion of filler due to weak filler-matrix interaction while Figure 6b shows better filler dispersion in matrix and better wetting by matrix caused by the addition of Lica-12.

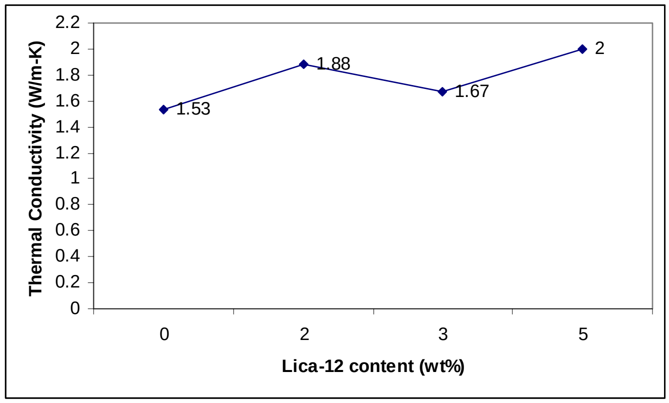
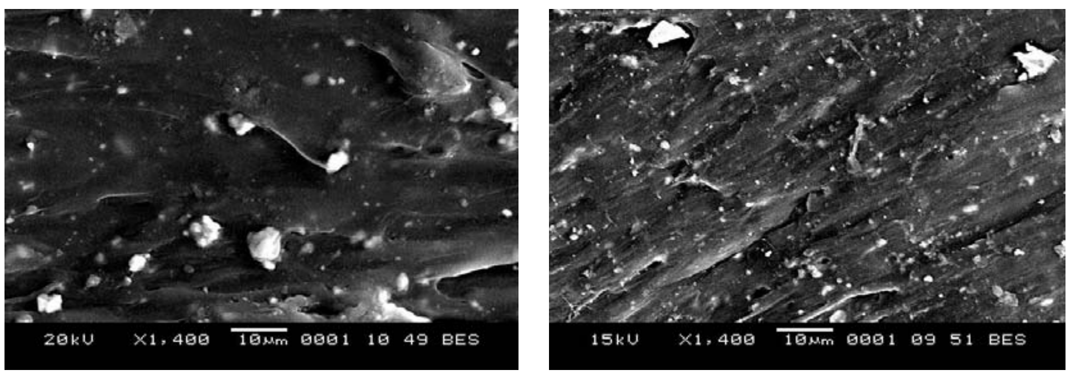


Figure 5: Effect of coupling agent (Lica-12) content on thermal conductivity of PP-AlN composites



(a) (b)

Figure 6: SEM micrograph of 90%PP-10% AlN composite (a) untreated (b) treated with 5% Lica-12 at 1400x magnification showed better dispersion of filler and improved wet-out between PP (matrix) and AlN (filler)

Conclusion

Flexural properties and thermal conductivity of AlN filled PP composites increased with increasing filler loading**.** Appropriately used, titanate coupling agents create significant improvements in the properties of AlN filled polypropylene composite. 0.5% of titanate (Lica-12) is able to produce good dispersion of filler in the 10%AlN-90%PP composite system, increase the thermal conductivity, while maintaining reasonable flexural properties. This research proved that there is possible to increase thermal conductivity of polymer composite even at low ceramic filler concentrations.

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