

# Thermal, Mechanical, and Thermo-Mechanical Properties of Thermosets Produced Using Low $M_w$ Functionalized Polyolefins and Polyamines

Tayyab Hameed,<sup>1</sup> David K. Potter,<sup>1,2</sup> Elizabeth Takacs<sup>1</sup>

<sup>1</sup>Department of Chemical Engineering, McMaster University, Hamilton, Ontario L8S 4L7, Canada

<sup>2</sup>Xerox Centre for Engineering Entrepreneurship & Innovation, School of Engineering Practice, McMaster University, Hamilton, Ontario L8S 4K1, Canada

Received 2 March 2011; accepted 21 August 2011

DOI 10.1002/app.35518

Published online 15 December 2011 in Wiley Online Library (wileyonlinelibrary.com).

**ABSTRACT:** Thermosets obtained by reacting highly functionalized maleic anhydride-grafted polyethylene and a polyetherdiamine at several  $\text{NH}_2/\text{MA}$  molar ratios were characterized for their gel content, thermal, mechanical, and thermo-mechanical behavior. Gel content varied with composition and a maximum (57%) observed when  $\text{NH}_2/\text{MA}$  molar ratio was 1.5. Two melting transitions were observed for thermosets, representing the semicrystalline polyethylene fraction in the gel and sol part of the material in contrast to a single transition for the starting polyethylene. Overall crosslinking suppressed the crystallinity of the polyethylene in the thermoset. A single  $T_g$  observed

in the DMA analysis suggested phase mixing between the polyethylene and polyether chains. A shift in the  $T_g$  observed was related to the degree of crosslinking in the thermosets. Tensile properties of the thermosets were observed to be a strong function of composition and the degree of crosslinking and the optimum mechanical performance was shown by thermosets when  $\text{NH}_2/\text{MA}$  molar ratio was 1.5 and 2.0. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 125: 46–52, 2012

**Key words:** maleic anhydride-grafted polyolefin; polyether diamines; crosslinked products; DSC; DMA; tensile properties

## INTRODUCTION

In recent studies,<sup>1–3</sup> the authors have shown the successful development of reactive prepolymers and thermosets by reacting low viscosity functionalized polyolefins and polyamines. Hameed et al.<sup>1</sup> investigated the reactions between highly functionalized low MW maleic anhydride-grafted polyethylene with several diamines. When these reactions were conducted in a solvent media, the reactions were observed to be fast. However, reactive prepolymers were obtained in each case by adjusting the concentration of the reaction mixtures. The prepolymers could be later turned into thermoset materials by heat treatment. The crosslinking reaction was followed using FTIR and titration, whereas changes in physical characteristics were examined using rheological and sintering techniques.

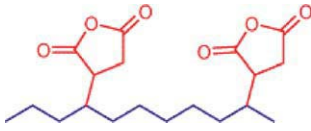
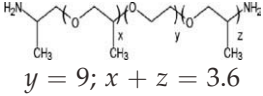
In another investigation,<sup>2,3</sup> similar reactions between a low viscosity functionalized polyethylene wax and a polyether diamine were carried out in the melt state. The products from these reactions were observed to be thermoplastic materials (prepolymers)

which crosslinked when melt processed at elevated temperatures to produce thermosets. The evolution of chemistry (FTIR) and crosslinking (gel content) with melt processing time was studied at different temperatures. Crosslinking of the prepolymers was observed to be a function of both time and temperature. These observations were supported by an increase in cyclic imide linkages as revealed by FTIR. The combination of gel content and FTIR analysis suggested that the reaction between residual maleic anhydride and amine groups continued during the melt processing of the prepolymers, resulting in formation of thermoset materials.

To the best of our knowledge, these recent studies are the first to explore such reactions using low molecular weight functionalized polyolefins and polyamines, however, several investigations exist where similar reactions were used to modify or crosslink high molecular weight polymeric systems.<sup>4–8</sup> Lu et al.<sup>4</sup> performed reactions between maleic anhydride-grafted polypropylene and several low MW diamines in an attempt to generate polyolefins with amine functionality. The stoichiometry of the reaction mixtures were manipulated toward achieving this goal. Reactions were conducted using both an extruder and a melt blender at several amine to maleic anhydride ( $\text{NH}_2/\text{MA}$ ) molar ratios. Reaction between anhydride and amine in the

Correspondence to: D. K. Potter (potterd@mcmaster.ca).

**TABLE I**  
**Characteristics of Materials [Color figure can be viewed in the online issue, which is available at**  
**wileyonlinelibrary.com.]**

		(g/mol)		$M_w/M_n$	Viscosity	Density (g/cm <sup>3</sup> )	MAH content <sup>a</sup> (%)
		$M_w$	$M_n$				
PEMA4351		3000	1200	2.5	300 mPa s <sup>b</sup>	–	5.20
ED600	 $y = 9; x + z = 3.6$	–	600	–	75 cSt <sup>c</sup>	1.035	–

<sup>a</sup> Assessed by colorimetric titrations.

<sup>b</sup> Measured at 140°C.

<sup>c</sup> Measured at 20°C.

reactor was reported to be complete within 2 min based on the mixing torque data. This was further supported by FTIR analysis of the reaction products which was suggested to show increased conversion of anhydride with increasing NH<sub>2</sub>/MA molar ratio. Dynamic viscoelastic measurements of the reaction products in the melt state were reported to suggest the reaction products were thermoplastic. However, the viscoelastic properties ( $G'$  and  $\eta^*$ ) were strongly influenced by NH<sub>2</sub>/MA molar ratio of the reaction mixtures, with maximum viscosity and storage moduli observed when the functional group stoichiometry was equimolar.

In another investigation, Colbeaux et al.<sup>5</sup> used diamines to crosslink maleic anhydride-grafted polyethylene. The degree of grafting of the polyethylenes utilized was 0.16–0.18%. Reactions were done in a polymer blender using two different diamines (aliphatic and aromatic) at several NH<sub>2</sub>/MA molar ratios. Relatively faster reactions and higher extent of reaction was reported for aliphatic diamines in comparison to aromatic amines which was suggested to be a consequence of the poor miscibility of the latter with polyethylene. The gel content and dynamic viscoelastic data suggested that the maximum crosslinking (32% gel content) occurred at NH<sub>2</sub>/MA molar ratio 2.0.

This article is a continuation of our recent work using low viscosity functionalized polyolefins and polyamines.<sup>1–3</sup> Although the formation of thermally activated reactive prepolymers and formation of thermoset materials was presented in the earlier articles, the physical properties of the thermoset materials so obtained were not fully appreciated. In this article, the thermal, mechanical, and thermo-mechanical characteristics of the thermoset materials obtained by reacting functionalized polyolefins and polyamines are presented.

## EXPERIMENTAL

### Materials

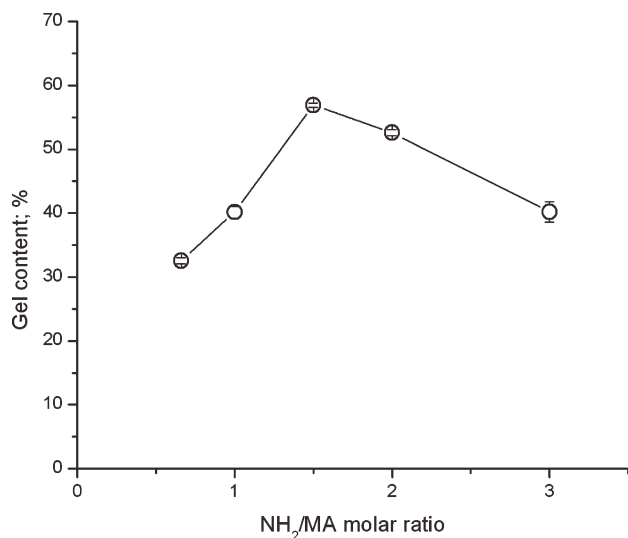
Low molecular weight maleic anhydride-grafted polyethylene was a commercial grade Licocene<sup>®</sup> PEMA4351 supplied by Clariant Canada Inc. Maleic anhydride was grafted randomly using free radical chemistry in a batch process.<sup>9</sup> Polyetherdiamine, ED600, was supplied by Huntsman Chemicals, TX, USA and used as received. The backbone of ED600 is predominantly the polyethylene oxide unit. The important characteristics of the materials are presented in Table I.

### Procedures

Reactive prepolymers were obtained by melt mixing PEMA4351 and ED600 at several NH<sub>2</sub>/MA molar ratios using either a melt blender or a resin kettle. Details of formation of these prepolymers are given elsewhere.<sup>3</sup> Thermoset materials were obtained by melt pressing these prepolymers at 180°C for 60 min using aluminum molds in a hot press. Specimens were produced as circular discs (25 mm diameter × 2 mm thick), dumbbells (2 mm thick; ASTM D638; Type V), and thin rectangular bars (25 mm × 5 mm × 0.5 mm) for different tests.

To assess the gel content, ~ 0.2–0.3 g of thermoset material were cut into small pieces and enclosed in pouches made of 120 × 120 mesh type 304 stainless steel wire cloth. These pouches were left in refluxing xylene for more than 12 h (ASTM D2765) for extraction of the noncrosslinked “sol” fraction. After extraction, samples were removed from the solvent, washed with acetone, and allowed to dry. The loss in weight was used to calculate the fraction of insoluble material or “gel content.”

Thermal properties of the thermosets and the neat PEMA4351 were measured using a Q2000



**Figure 1** Gel content as a function of NH<sub>2</sub>/MA molar ratio.

Differential Scanning Calorimeter, TA Instruments, USA. Approximately 5 mg of sample was encapsulated in aluminum pan for testing. The first heating cycle from room temperature to 140°C at 10°C/min was used to eliminate the thermal history of the material. It was followed by a controlled cooling and a second heating cycle at similar rates. Peak melting temperatures and heat of fusion data presented are from the second heating cycle.

The tensile tests were performed on an Instron 3366 (Table model), USA, tensile testing machine at room temperature. The specimen gauge length was 7.62 mm and a crosshead speed of 5 mm/s was used. All reported mechanical properties are based on an average of a minimum of four to five specimens.

Thermo-mechanical properties were measured on Dynamic Mechanical Analyzer 2980, TA Instruments, USA. Specimens were cooled down to -110°C using liquid nitrogen and then heated at 5°C/min to 200°C. All measurements were conducted using film tension clamps at a frequency of 10 Hz and amplitude strain of 10 μm.

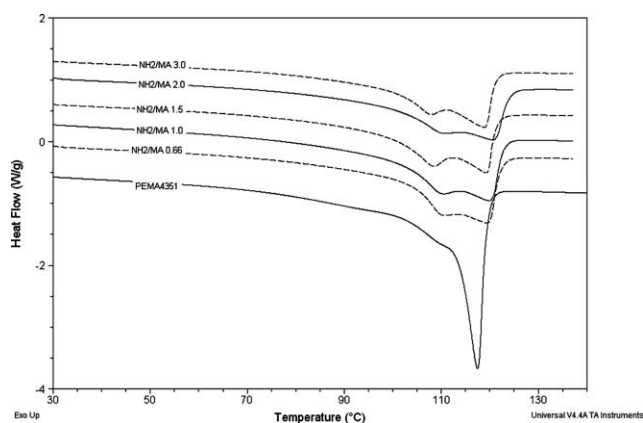
Morphology of the blends was captured by means of a Scanning Electron Microscopy (SEM) instrument (JSM 7000F). Samples were fractured in liquid nitrogen and coated with gold before testing.

## RESULTS AND DISCUSSION

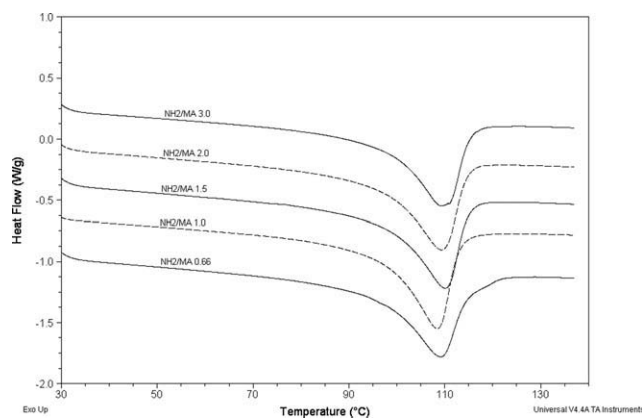
As reported in an earlier study,<sup>3</sup> all the prepolymers obtained from the melt mixing operation in a resin kettle or a melt blender were thermoplastic and showed negligible gel content. Results for gel content of the prepolymers melt pressed for 60 min at 180°C as a function of NH<sub>2</sub>/MA molar ratio of the reactive mixture are presented in Figure 1. Gel content increases with decreasing NH<sub>2</sub>/MA molar ratio,

reaches a maximum at NH<sub>2</sub>/MA molar ratio 1.5, and then starts to decline with further decrease in NH<sub>2</sub>/MA molar ratio. As the gel content corresponds to the extent of crosslinking, theoretically the maximum degree of gel content was expected at equimolar stoichiometry.<sup>4,10</sup> Lu and Macosko<sup>4</sup> while studying the reaction between hexamethylene diamine (HMDA) and maleic anhydride-grafted polypropylene at various NH<sub>2</sub>/MA molar ratios observed maximum viscosity at equimolar stoichiometry. On the other hand, Colbeaux et al.<sup>5</sup> while studying melt reactions between maleic anhydride-grafted polyethylene and several diamines reported a maximum viscosity and gel content at NH<sub>2</sub>/MA molar ratio 2.0. Although, ideally a maximum extent of reaction and thus crosslinking is expected at equimolar stoichiometry, it is dependent on the degree of mixing that could be practically achieved in the reaction mixtures, especially in polymeric systems where the viscosities are significantly high. Interestingly, the gel content obtained for the thermoset materials produced using the prepolymer technique are comparable to that of crosslinked polyolefins produced using conventional methods.<sup>11-15</sup>

The thermal characteristics of the thermoset materials were studied using Differential Scanning Calorimetry (DSC). The second heating curves for PEMA4351 and all the thermoset materials prepared using several NH<sub>2</sub>/MA molar ratios are presented in Figure 2. The melting behavior of thermoset products is different from the neat PEMA4351 starting material. Whereas, PEMA4351 shows a very sharp and a single melting peak around 118°C, the thermoset materials showed two distinct relatively broad peaks. The higher temperature melting peak in the thermoset materials closely correspond with the melting peak in PEMA4351 and are suggested to represent the melting of an unmodified/noncrosslinked PEMA4351 crystal population in the thermosets. This crystal population most probably



**Figure 2** DSC second heat curves for all thermosets.



**Figure 3** DSC second heat curves for gel fraction of thermosets.

represents the fraction of nongrafted PEMA4351 that did not become part of the network polymer. The relatively low intensity of this peak in thermoset scans compared with that of PEMA4351 suggest that most of the functionalized polyolefin did become part of the network. On the other hand, the new low temperature melting transition observed around 110°C in thermoset materials is attributed to a crystal population of modified/crosslinked PEMA4351 chains that did become part of the network polymer. The decreased melting point of this crystal population is believed to be a consequence of crystal defects introduced in chain folding because of the presence of crosslinks and branches. To confirm the hypothesis, DSC scans were performed on the gel (crosslinked) fraction obtained after solvent extraction of the thermoset materials. The overlaid second heating scans for these tests are presented in Figure 3. Unlike the scans for thermoset materials, only a single melting peak is observed in these endotherms. The temperatures for these peaks closely correspond to the lower temperature melting transition observed in Figure 2, confirming that these transitions were in fact due to the modified/crosslinked PEMA4351 crystallites that became part of the crosslinked network.

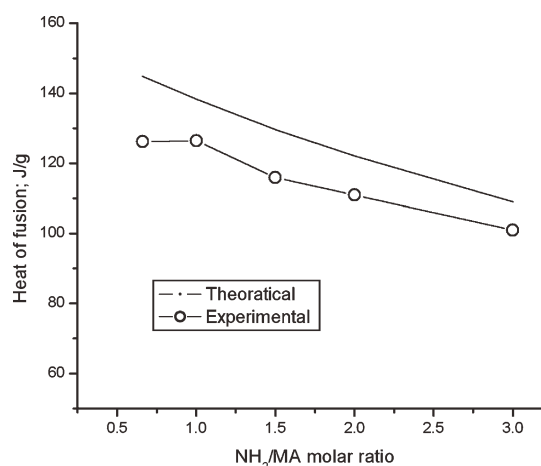
The heat of fusion assessed by integration of the two melting transitions observed for the thermoset materials are plotted in Figure 4 as a function of  $\text{NH}_2/\text{MA}$  molar ratio. The theoretical heat of fusion based on the weight fraction of PEMA4351, the semicrystalline component of the thermoset, in the thermoset materials are also plotted as a reference. The experimentally assessed heat of fusion is always less than the theoretically estimated value, suggesting the branching and crosslinking suppressed the crystallization of the PEMA4351 in the thermoset materials.

Dynamic mechanical analysis is a technique widely used to characterize the morphology of polymeric materials.<sup>14,16–19</sup> Phase separated polymeric

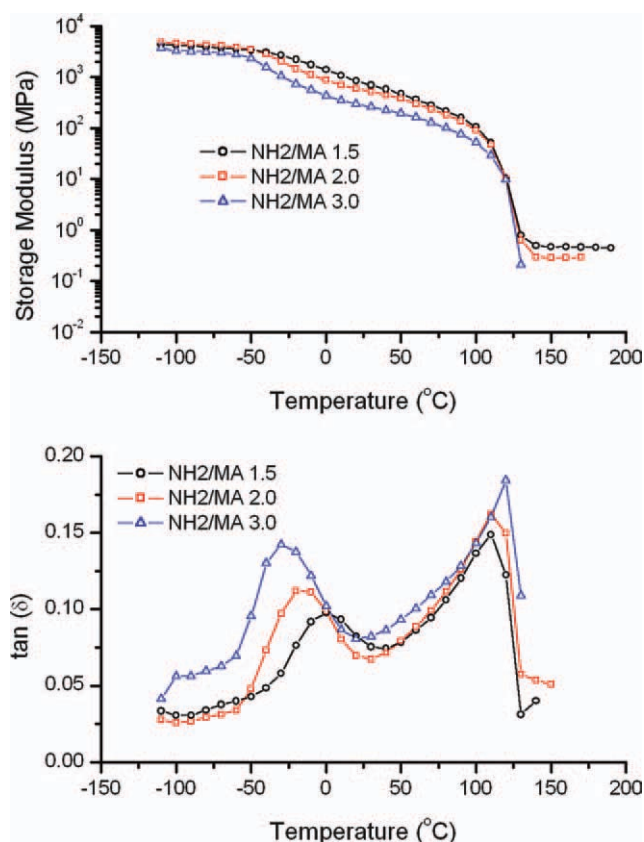
blends and elastomers<sup>20</sup> show distinct glass transition temperatures ( $T_g$ ) corresponding to the each component in the multi phase system, whereas a single  $T_g$  that is intermediate in temperature between the glass transition temperatures of the components, is taken as evidence of miscibility or partial miscibility of such polymeric systems. Temperature sweep data for the three thermoset materials representing  $\text{NH}_2/\text{MA}$  molar ratios of 1.5, 2.0, and 3.0 are presented in Figure 5. Thermosets materials formed using compositions  $\text{NH}_2/\text{MA}$  molar ratios 0.66 and 1.0 were too brittle and weak to be tested for the dynamic mechanical properties.

All thermosets materials tested showed comparable moduli below the  $T_g$ . A unique  $T_g$  was observed for each of the three thermosets that shifted toward a higher temperature with decreasing  $\text{NH}_2/\text{MA}$  molar ratio. For polymer blends and segmented polyurethanes, the presence of an intermediate glass transition temperature between those of the individual components is indicative of good phase mixing between blend components.<sup>20</sup> On the other hand, when new chemical bonds between chains of a polymer or polymer blend are formed, the linkages/bonds restrict the mobility of the polymeric chain segments which results in an increase in the observed glass transition temperature.

Thermosets characterized in this study are composed of polyethylene and polyethylene oxide chains linked by cyclic imide linkages.<sup>1–3</sup> The polyether chains of varying lengths have been reported to possess  $T_g$  values around  $-40^\circ\text{C}$  in phase segregated polyurethane systems.<sup>20</sup> The  $T_g$  observed for the system containing the highest weight fraction of polyether ( $\text{NH}_2/\text{MA}$  molar ratio 3.0) and least crosslinks (Fig. 1) is very close to this value and seems to correspond to the  $T_g$  of the polyether component in the thermoset.  $T_g$  values for polyethylene have been reported to be around  $-110^\circ\text{C}$ .<sup>14,21</sup> The DMA



**Figure 4** Heat of fusion as a function of  $\text{NH}_2/\text{MA}$  molar ratio of the thermosets.



**Figure 5** Storage moduli and  $\tan \delta$  versus temperature curves for the thermosets. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

equipment used in this study did not allow us to attain low enough temperatures to measure transitions in that range. The observed shift in the glass transition temperature in the current investigation is toward a higher temperature with decreasing  $\text{NH}_2/\text{MA}$  molar ratio and is most probably due to the increase in crosslinking of polyether component. If it were a response from a miscible or partially miscible polyethylene and polyether phase, the shift in the glass transition temperature would be in the opposite direction.

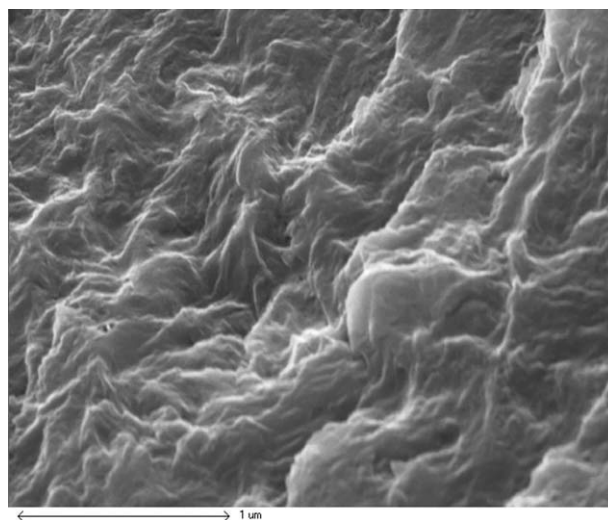
The observed trend is very much consistent with the degree of crosslinking in Figure 1. The degree of crosslinking increases with decreasing  $\text{NH}_2/\text{MA}$  molar ratio restricting the mobility of chain segments and raising the observed glass transition temperature. This analysis suggests that the polyether and polyethylene parts of the thermoset exist in discreet or partially discreet phases and that the change in the observed glass transition temperature is a consequence of the impact of crosslinking on the mobility of polyether chain segments.

The rubbery plateau region in the temperature sweeps of the three thermosets is obscured by the semicrystalline morphology of the polyethylene

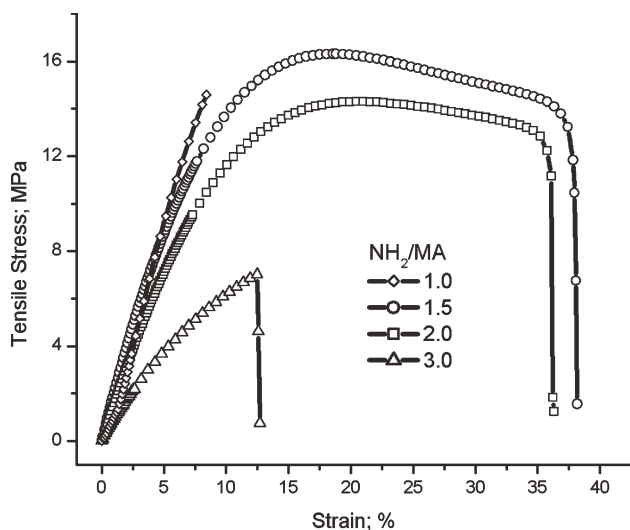
phase until melting occurs. Beyond the melting transition, distinctive rubbery crosslink plateaus are observed for the two most highly crosslinked samples. Crosslink plateaus were observed for thermoset products  $\text{NH}_2/\text{MA}$  1.5 and 2.0 which sustained the mechanical strain up to 200°C without further transition to liquid-like behavior. No such plateau was observed for the material in which the  $\text{NH}_2/\text{MA}$  molar ratio is equal to 3.0. In this case, the modulus decays rapidly and consistent with a transition to liquid-like behavior upon complete melting of the PEMA4351. However, it is interesting to note that although the thermoset with  $\text{NH}_2/\text{MA}$  molar ratio 3.0 did not show a crosslink plateau no significant deformation (necking) in the specimen shape was observed at the end of the test similar to the other two thermoset materials. The absence of a crosslink plateau in the DMA spectrum of  $\text{NH}_2/\text{MA}$  molar ratio 3.0 material indicates that the material is behaving like a thermoplastic and that the extensive gel content measured for this material (>35%) is present in localized gels.

Figure 6 shows typical SEM micrographs for the product  $\text{NH}_2/\text{MA}$  molar ratio 2.0. The micrographs for other products were similar and hence not shown. It is hard to contrast between polyethylene and polyether. A plausible explanation is the very low molecular weight and high degree of functionality of the two components results in good mixing and interlocking of the chains retarding significant phase change. In the absence of good phase contrast, it is hard to infer miscibility of the two phases.

Representative stress versus strain curves from the tensile property measurements of the thermoset materials are presented in Figure 7. The thermoset material  $\text{NH}_2/\text{MA}$  molar ratio 3.0 showed a rubber-like behavior with no yield point and failed at relatively



**Figure 6** SEM micrograph of product  $\text{NH}_2/\text{MA}$  molar ratio 2.0.

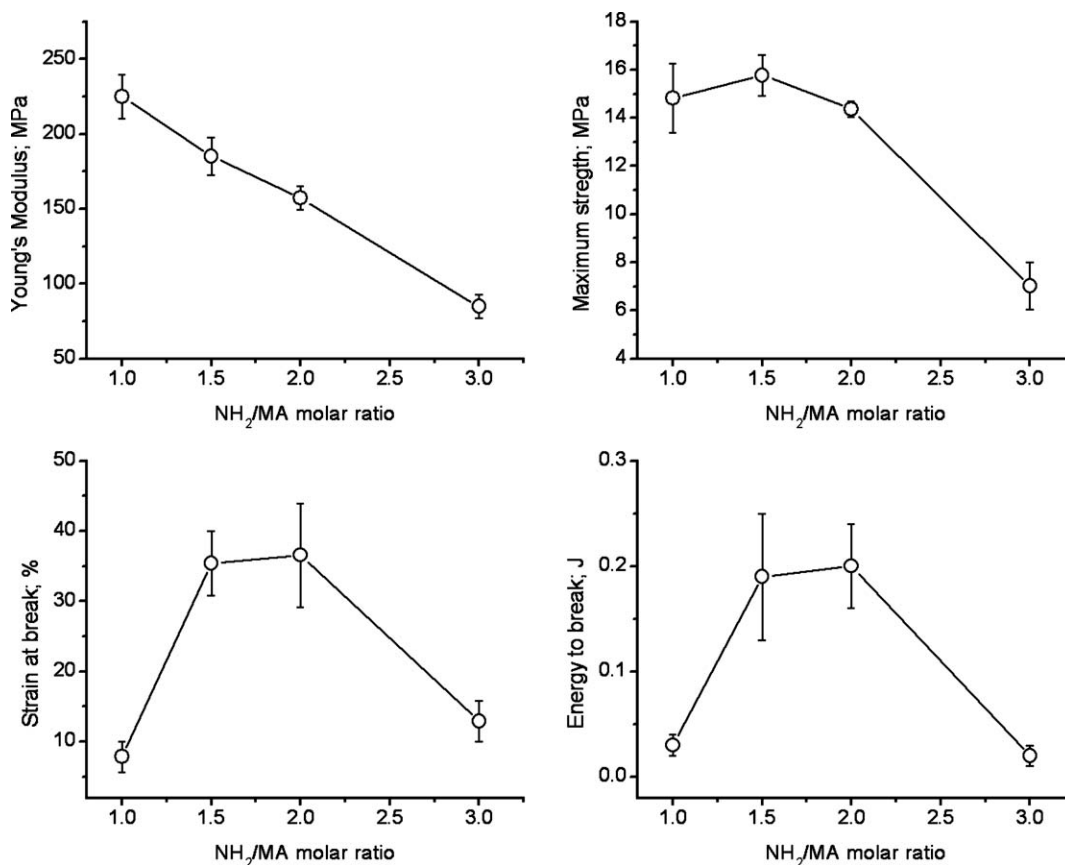


**Figure 7** Representative stress versus strain curves for the thermosets.

low elongation at break. On the other hand, the two thermosets  $\text{NH}_2/\text{MA}$  1.5 and 2.0 were much stiffer, stronger, and tougher as evident from the modulus (initial slope), strength at break, and area under the curve for these materials. They also showed yield behavior typical of semicrystalline polymers. A decrease in strength in the post yield region before

failure is typical of crosslinked polymers unlike thermoplastic materials which usually show strain hardening before failure. Finally, the curve for the thermoset  $\text{NH}_2/\text{MA}$  molar ratio 1.0, depicts the highest modulus but the strength and ductility observed was less than that shown by thermoset materials with  $\text{NH}_2/\text{MA}$  molar ratio 1.5 and 2.0. Tensile properties of the thermoset  $\text{NH}_2/\text{MA}$  0.66 could not be measured since specimens could not be produced due to the brittleness of the material.

The mechanical properties Young's modulus, maximum strength, elongation at break, and energy to break are plotted as a function of  $\text{NH}_2/\text{MA}$  molar ratio in Figure 8. Young's modulus increases with decreasing  $\text{NH}_2/\text{MA}$  molar ratio. In fact, a linear relationship with  $R^2 = 0.99$  was observed. This was expected since the weight fraction of semicrystalline material increases at lower  $\text{NH}_2/\text{MA}$  molar ratios making the material stiffer. On the other hand, the maximum strength versus  $\text{NH}_2/\text{MA}$  trend was different. The highest maximum strength was observed for  $\text{NH}_2/\text{MA}$  molar ratios 1.0, 1.5, and 2.0 showing comparable values. The maximum strength shown by thermoset  $\text{NH}_2/\text{MA}$  molar ratio 3.0 was relatively low. The trend observed in strength at break is probably a consequence of combined effect of molecular weight build up due to extensive crosslinking in these



**Figure 8** Tensile properties as a function of  $\text{NH}_2/\text{MA}$  molar ratio.

thermosets as well as the crystallinity. This relationship between the mechanical properties and the abovementioned factors is also manifest in the elongation at break trends. Elongation at break values are highest and similar for thermosets with  $\text{NH}_2/\text{MA}$  molar ratios 1.5 and 2.0. Whereas, the elongation at break values of thermosets with  $\text{NH}_2/\text{MA}$  molar ratio 1.0 and 3.0 is quite low. Comparison of strength at break and elongation at break curves highlight the combined role of crosslinking and crystallinity on the tensile properties of the thermosets. Although thermosets  $\text{NH}_2/\text{MA}$  1.0, 1.5, and 2.0 show similar strength at break, thermoset  $\text{NH}_2/\text{MA}$  1.0 shows relatively low ductility although it has a higher crystallinity than the other two materials. On the other hand, the product with the highest concentration of ED600 ( $\text{NH}_2/\text{MA}$  3.0) was expected to show the highest ductility, however, the observed elongation at break is much lower. This is likely a consequence of the relatively low extent of crosslinking in this thermoset. Finally, the energy expended before failure follows similar trends as observed for elongation at break values. These observations suggest that optimum tensile properties for these thermosets could be obtained around the  $\text{NH}_2/\text{MA}$  ratios 1.5 and 2.0. It is interesting to mention that the mechanical properties depicted by these thermosets are comparable to crosslinked polyolefins prepared using conventional techniques.<sup>22</sup>

### CONCLUSIONS

Thermosets produced by reacting highly functionalized maleic anhydride-grafted polyethylene and a polyether diamine at several  $\text{NH}_2/\text{MA}$  molar ratios were characterized for their gel content, thermal, mechanical, and thermo-mechanical behavior. Maximum gel content was observed when  $\text{NH}_2/\text{MA}$  molar ratio of the reactive mixture was 1.5. Two melting transitions were observed for thermosets, representing the semicrystalline polyethylene fraction in the gel and sol part of the material in contrast to a single transition for the starting semicrystalline polyethylene. Crosslinking overall suppressed the crystallinity of the polyethylene in the thermoset. The glass transition temperature of the polyether

component was observed to shift to a higher temperature with increasing crosslinking. Tensile properties of the thermosets were observed to be a strong function of composition and degree of crosslinking and the best mechanical performance was observed for the  $\text{NH}_2/\text{MA}$  molar ratio 1.5 and 2.0.

### References

1. Hameed, T.; Potter, D. K.; Takacs, E. *J Appl Polym Sci* 2010, 116, 2285.
2. Hameed, T.; Potter, D. K.; Takacs, E. 68th Annual Technical Conference of the Society of Plastics Engineers, ANTEC, May 2010.
3. Hameed, T.; Potter, D. K.; Takacs, E. *Polym Int*, submitted.
4. Lu, Q.; Macosko, C. W.; Horrión, J. *J Polym Sci Part A: Polym Chem* 2005, 43, 4217.
5. Colbeaux, A.; Fenouillot, F.; Gerard, J.; Taha, M.; Wautier, H. *Polym Int* 2005, 54, 692.
6. Schmidt, U.; Zschoche, S.; Werner, C. *J Appl Polym Sci* 2003, 87, 1255.
7. Song, Z.; Baker, W. E. *J Polym Sci Part A: Polym Chem* 1992, 30, 1589.
8. Vermeesch, I.; Groeninckx, G. *J Appl Polym Sci* 1994, 53, 1365.
9. Hohner, G.; Bayer, M. U.S. Pat. 7,005,224 (2006).
10. Macosko, C. W. *RIM: Fundamentals of Reaction Injection Molding*; Oxford University Press: Canada, 1989.
11. Ali Khonakdar, H.; Morshedian, J.; Eslami, H.; Shokrollahi, F. *J Appl Polym Sci* 2004, 91, 1389.
12. Chattopadhyay, S.; Chaki, T. K.; Bhowmick, A. K. *J Appl Polym Sci* 2001, 81, 1936.
13. Dadbin, S.; Frounchi, M.; Haji Saeid, M.; Gangi, F. *J Appl Polym Sci* 2002, 86, 1959.
14. Madani, M.; Sharifi-Sanjani, N.; Rezaei-Zare, E.; Faridi-Majidi, R. *J Appl Polym Sci* 2007, 104, 1873.
15. Mishra, J. K.; Chang, Y.; Lee, B. C.; Ryu, S. H. *Radiat Phys Chem* 2008, 77, 675.
16. Hu, J.; Yang, Z.; Yeung, L.; Ji, F.; Liu, Y. *Polym Int* 2005, 54, 854.
17. Lee, D.; Tsai, H. *J Appl Polym Sci* 2000, 75, 167.
18. Lee, D.; Tsai, H.; Tsai, R.; Chen, P. H. *Polym Eng Sci* 2007, 47, 695.
19. Moravek, S. J.; Hassan, M. K.; Drake, D. J.; Cooper, T. R.; Wiggins, J. S.; Mauritz, K. A. *J Appl Polym Sci* 2010, 115, 1873.
20. Stanford, J. L.; Still, R. H.; Wilkinson, A. N. *Polymer* 2003, 44, 3985.
21. Andrew, J. P. *Handbook of Polyethylene: Structures, Properties, and Applications*; Marcel Dekker: New York, 2000.
22. Barzin, J.; Azizi, H.; Morshedian, J. *Polym—Plast Technol Eng* 2007, 46, 305.