

# Dynamic Mechanical Properties of Semi-Interpenetrating Polymer Network-Based on Nitrile Rubber and Poly(methyl methacrylate-co-butyl acrylate)

Chang Su,<sup>1</sup> Dezhen Zong,<sup>1</sup> Lihuan Xu,<sup>2</sup> Cheng Zhang<sup>1</sup>

<sup>1</sup>State Key Laboratory Breeding Base of Green Chemistry-Synthesis Technology, College of Chemical Engineering and Materials Science, Zhejiang University of Technology, Hangzhou, P. R. China

<sup>2</sup>College of Chemical Engineering, Shenyang University of Chemical Technology, Shenyang P. R. China

Correspondence to: C. Zhang (E-mail: czhang@zjut.edu.cn) and L. Xu (E-mail: xulihuanss@163.com)

**ABSTRACT:** In this article, semi-interpenetrating polymer network (Semi-IPNs) based on nitrile rubber (NBR) and poly(methyl methacrylate-co-butyl acrylate) (P(MMA-BA)) were synthesized. The structure and damping properties of the prepared Semi-IPNs blends were characterized and by fourier transform infrared spectrum (FTIR), dynamic mechanical analysis (DMA), scanning electron microscopy (SEM), thermogravimetric analysis (TGA/DTG), and tensile mechanical properties. The results showed that interpenetrating network based on P(MMA-BA) and NBR was successfully obtained, which showed the improved thermal stability compared to NBR/P(MMA-BA)-based two-roll mill blends. Furthermore, Semi-IPNs showed significantly better the dynamic mechanical properties than that of the two-roll mill system. With the increasing feed ratio of BA and MMA during the preparation of Semi-IPNs, the loss peak position for P(MMA-BA) in NBR/PMMA IPNs shifted to a lower temperature from 20°C to -17°C, and when NBR in Semi-IPNs was accounted for 40 wt %, the dynamic mechanical thermal analysis showed that much more advanced damping material with wider temperature range ( $-30^{\circ}\text{C} < T < 80^{\circ}\text{C}$ ) as  $\tan \delta > 0.45$  can be achieved. Therefore, it was expected as a promising way to obtain the excellent damping materials with good oil-resisted properties according the Semi-IPNs system. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 40217.

**KEYWORDS:** composites; glass transition; properties and characterization; addition polymerization; rubber

Received 6 September 2013; accepted 21 November 2013

DOI: 10.1002/app.40217

## INTRODUCTION

Viscoelastic polymers as damping materials have attracted considerable attention both in academia and industry in recent years, due to their outstanding properties for reducing vibration and noise around the glass transition temperature ( $T_g$ ).<sup>1,2</sup> As the good damping materials, the viscoelastic polymer should have  $\tan \delta$  values of higher than 0.3 for a temperature range of at least 60°C.<sup>3</sup> However, the viscoelastic polymers usually have the characteristics of the narrow  $T_g$  peak which is far away from the room temperature, obstructing further application as damping materials. Many attempts had been made to control the damping peak position to locate within a required temperature region and to enhance the damping peak intensity of a polymer-based damping material, such as polymer blends,<sup>4,5</sup> interpenetrating networks<sup>6-8</sup> and using of plasticizers and fillers, and so on.<sup>9-11</sup>

Among the above methods, interpenetrating polymer networks (IPNs) blends is a promising approach to obtain an advanced

damping material. It is the partial or total physical interlocking of individual components in IPNs<sup>12-17</sup> results in that the blends possess excellent thermal stability and mechanical property because of a synergistic effect induced by the forced compatibility of individual components,<sup>18,19</sup> which makes IPNs blends display a broad glass-to-rubber transition due to the micro-heterogeneities in the matrix and hence exhibit a broad range of temperatures and frequencies of damping.<sup>20</sup> Qin et al. had succeeded in synthesizing a series of gradient IPNs of polyurethane/vinyl ester resin (PU/VER), resulting in the increased  $\tan \delta$  value in the middle of the two loss peaks.<sup>7</sup> Chen et al. had also researched on polyurethane/epoxy IPNs composites, in which the temperature ranges with  $\tan \delta > 0.3$  were  $\sim 50^{\circ}\text{C}$  and the  $\tan \delta$  reached a maximum value of 1.105.<sup>17</sup> Usually, the proper groups (such as acrylate and acrylonitrile group) in the matrix polymer is quite important to obtain the advanced damping materials, due to that the viscoelastic damping capability of a polymer is governed by the interaction of inherent

groups, as suggested by Sperling.<sup>21</sup> Nitrile rubber/poly(alkyl methacrylate)s blends<sup>22</sup> and nitrile rubber/poly(vinyl chloride)/poly(alkyl methacrylate)s blends,<sup>23</sup> all have the polar groups in the polymers matrix and had been reported to obtain the obviously improve damping properties. Furthermore, NBR/PMMA IPNs had been synthesized by single-step (SIPN) and multiple-step (MIPN) methods, which obtained  $\delta$  peaks of 0.3 with the temperature range ( $-10^{\circ}\text{C} < T < 80^{\circ}\text{C}$ ).<sup>20</sup>

Both NBR and poly(alkyl acrylate)s rubber, which have both acrylonitrile and acrylate groups in the polymer chain, are well-known as the high performance specialty rubber for manufacturing seals and gaskets used in high-performance automotive engines because of its outstanding oil temperature resistance, which all exhibits efficient damping properties around  $T_g$ .<sup>24</sup> Therefore, the construction of IPNs blends of NBR/poly(alkyl acrylate) will contribute to prepare a good oil resistance material with the excellent damping properties. Consequently, in this article, the Semi-IPNs blends was prepared by in-situ polymerization of poly(methyl methacrylate-*co*-butyl acrylate) in NBR solution. The effects of both the NBR content in the Semi-IPNs blends and the MMA/BA ratio in poly(methyl methacrylate-*co*-butyl acrylate) on the damping properties of these resulted Semi-IPNs were investigated comparably by dynamic mechanical thermal analysis. Furthermore, the structure, morphology and the thermal stability of the obtained the Semi-IPNs were also discussed.

## EXPERIMENTAL

### Material

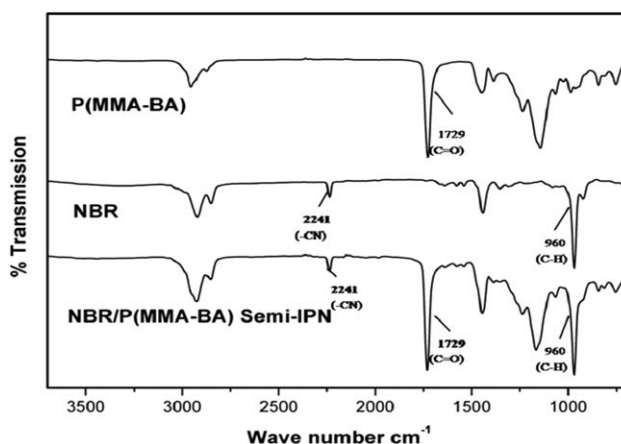
NBR in this study was Nipol DN4050 with the acrylonitrile content about 40%, purchased from Nippon Zeon Corp. Azobisisobutyronitrile (AIBN, Aladdin) was purified by recrystallization from chloroform/ethanol (80/20, v/v) mixture. Methyl methacrylate (MMA), Butyl acrylate (BA) and Acrylic acid (AA) (all from Shanghai Chemical Reagents, China) were used as received. Triethylene glycol dimethacrylate (EGDMA, Aladdin) and AA were the crosslinkers for P(MMA-BA).

### Preparation of the Semi-IPNs

For making Semi-IPNs, a certain amount of NBR was firstly added to a 250-mL four-neck round-bottom with ethyl acetate as solvent. After the rubber was completely dissolved, then MMA, BA, and crosslinking agent (2 wt % EDGMA and 2 wt % AA) were added. The mixture was heated with stirring at  $78^{\circ}\text{C}$  for 1 h. Then, 0.5 wt % AIBN of the monomer was added under nitrogen atmosphere for 3 h. The obtained Semi-IPNs were dried at  $50^{\circ}\text{C}$  in an oven to constant weight, getting Semi-IPNs damping materials-NBR/P(MMA-BA). The series of Semi-IPNs were synthesized according to the same way by varying both the feed ratio of two monomers in P(MMA-BA) and the NBR content in the NBR/P(MMA-BA) blends.

### Preparation of the Two-Roll Mill Samples

The series of two-roll mill samples were prepared by mixing of both NBR and P(MMA-BA) in a two-roll mill  $110^{\circ}\text{C}$  for 20 min. The resulting blends were preheated for 3 min, and then hot-compressed at  $110^{\circ}\text{C}$  for another 10 min under a pressure of 50 MPa. And finally, the samples were quenched in



**Figure 1.** FTIR spectra of P(MMA-BA), NBR, and NBR/P(MMA-BA) Semi-IPNs.

water-cooling to obtain the resulting samples with a thickness of about 1.5 mm.

## CHARACTERIZATION

### Fourier Transform Infrared (FTIR) Spectroscopy

Infrared measurements were conducted on a Nicolet 6700 FTIR spectrometer made by Thermo Co. in USA. The FTIR spectra were acquired by scanning the specimens in the wave number range from 400 to  $4000\text{ cm}^{-1}$  with attenuation reflection (ATR) mode at a resolution of  $1\text{ cm}^{-1}$ .

### Dynamic Mechanical Analysis (DMA)

Dynamic viscoelastic measurements were carried out by Triton 2000 dynamic mechanical analyzer (British Triton Technology) in a tension mode. The test frequency was fixed at 1 Hz and the measurements temperatures were ranged from  $-30$  to  $100^{\circ}\text{C}$  at a heat rate of  $3^{\circ}\text{C}/\text{min}$ .

### Scanning Electron Microscopy (SEM)

The compressing molded sheet samples were fractured in liquid nitrogen, coated with Pt-Pd, and then examined with a HITACHI X-650 scanning electron microscopy (Japan).

### Thermogravimetric Analysis (TGA/DTG)

TGA(DTG) techniques (Pyris Diamond TG/DTA) were used. The samples were heated from  $25^{\circ}\text{C}$  to  $600^{\circ}\text{C}$  at a heating rate of  $10^{\circ}\text{C}/\text{min}$  in a nitrogen atmosphere.

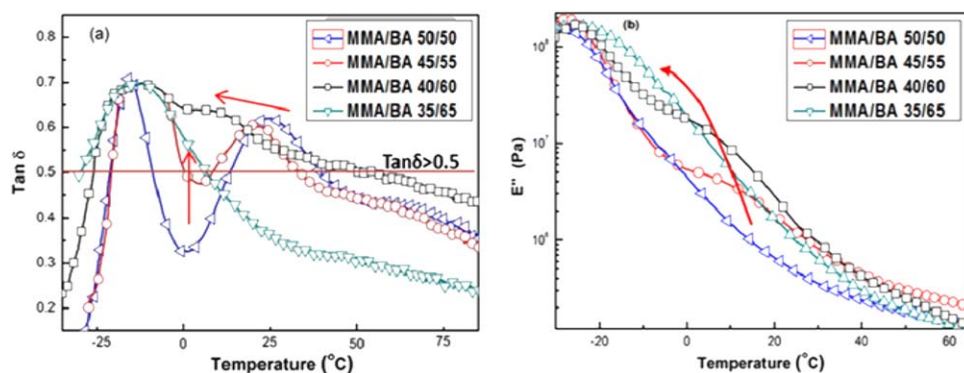
### Tensile Mechanical Properties

Tensile strength and elongation at break were measured by using a Universal testing machine (American Instron 5996). The samples were measured at room temperature at a crosshead speed of 500 mm/min, with dumbbell-shaped specimens, according to GB/T528-2009.

## RESULTS AND DISCUSSION

### FTIR Spectra

FTIR spectra were used to characterize the structure of Semi-IPNs. Figure 1 showed the FTIR spectra of P(MMA-BA), NBR, and of NBR/P(MMA-BA) Semi-IPNs, respectively. The pure NBR shows a characteristic absorption peak at around  $2241\text{ cm}^{-1}$ , which are assigned to the stretching vibration of



**Figure 2.** Temperature dependence of  $\tan \delta$  (a) and  $E'$  (b) at 1 Hz for NBR/P(MMA-BA) Semi-IPNs (40 : 60, by weight) with different feed ratio of BA and MMA during the preparation of Semi-IPNs. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

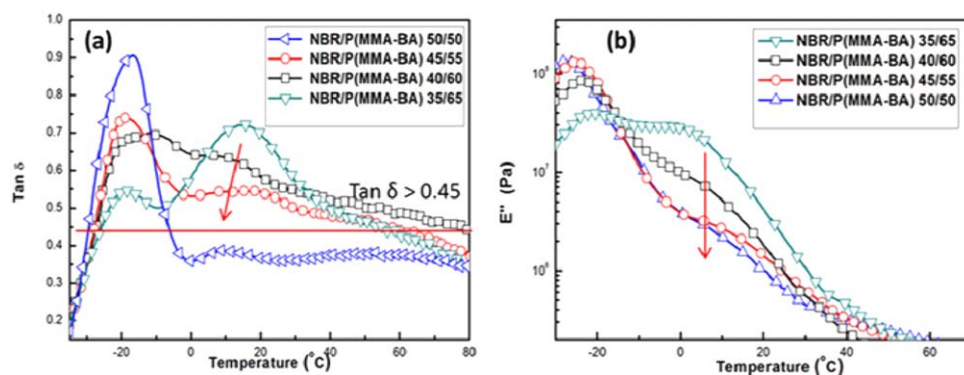
the  $C\equiv N$  group of NBR. And the crosslinked P(MMA-BA) displays the strong absorption peak appearing at  $960\text{ cm}^{-1}$  and  $1729\text{ cm}^{-1}$ , which can be assigned to C—H of trans-1, 4 structure and the free C=O in COOH group of the polymer, respectively. Comparatively, the spectrum of the Semi-IPNs includes the all characteristic peaks of both NBR and P(MMA-BA) at around  $960\text{ cm}^{-1}$ ,  $1729\text{ cm}^{-1}$ , and  $2241\text{ cm}^{-1}$ , indicating that NBR and P(MMA-BA) segments have been successfully included in the obtained Semi-IPNs blends. Furthermore, it can be observed that there are not any shift of the characteristic peaks to occur in Semi-IPNs blends compared to the corresponding pure polymer segments, indicating that no chemical interaction occurs during the formation of Semi-IPNs.

### Dynamic Mechanical Thermal Analysis

The DMA technique was employed to investigate the thermal properties and damping properties of NBR/P(MMA-BA) Semi-IPNs. Figure 2 showed the temperature dependence of  $\tan \delta$  (a) and  $E'$  (b) for NBR/P(MMA-BA) (40/60 (wt/wt)) with various feed ratio of BA and MMA during the preparation of NBR/P(MMA-BA) Semi-IPNs. As shown in Figure 2(a), the Semi-IPNs sample with the feed ratio of BA and MMA at 50/50 (wt/wt) exhibits two relaxation peaks, in which the relaxation peak at about  $-20^\circ\text{C}$  is attribute to the NBR segment in Semi-IPNs and another one is due to the existence of PMMA. With the increase of the feed ratio of BA and MMA to 55/45 (wt/wt), the

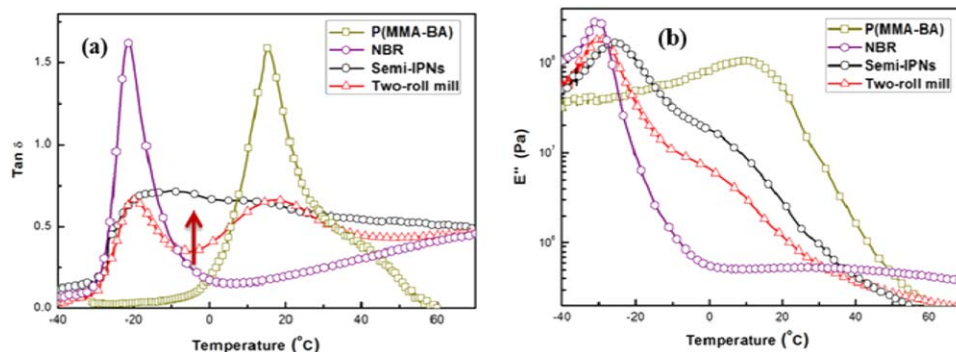
loss peak position of NBR keeps unchanged, while the peak position for P(MMA-BA) shifts to a lower temperature from  $20^\circ\text{C}$  to  $-17^\circ\text{C}$ , due to the more BA component as the inner plasticizer increasing the flexibility of P(MMA-BA) chains. When the feed ratio of BA and MMA is at 60/40 (wt/wt), the valley between two relaxation peaks nearly disappears, and the curve shows a broad damping platform with wide damping temperature range ( $-25^\circ\text{C} < T < 75^\circ\text{C}$ ) as  $\tan \delta > 0.5$ . However, with further increasing the feed ratio of BA and MMA to 65/35 (wt/wt), the sample exhibits only one overlapped relaxation peak with the narrow damping temperature range, due to the forced compatibility of IPNs and the adjacent glass temperature ( $T_g$ ) of both NBR and P(MMA-BA). Therefore, the adjusting BA/MMA ratio of P(MMA-BA) in the NBR/P(MMA-BA) Semi-IPNs is a valid means for constructing the broad temperature range damping materials.

Figure 2(b) showed the temperature dependence of loss modulus ( $E''$ ) for NBR/P(MMA-BA) Semi-IPNs. When the feed ratio of BA and MMA increases from 50/50 (wt/wt) to 60/40 (wt/wt), the obtained NBR/P(MMA-BA) Semi-IPNs displays a higher  $E''$  value at the higher temperature part compared to that at 50/50 (wt/wt), while the  $E''$  value at near  $-20^\circ\text{C}$  is almost unchanged. Also the peak positions of the loss modulus peaks near  $20^\circ\text{C}$  tend to shift toward the lower temperature with the increase of the BA content, and, meanwhile, the corresponding peak height increase gradually. This trend is in accord



**Figure 3.** Temperature dependence of  $\tan \delta$  (a) and  $E'$  (b) at 1 Hz for NBR/P(MMA-BA) Semi-IPNs with different contents of NBR. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]





**Figure 4.** Temperature dependence of  $\tan \delta$  (a) and  $E''$  (b) at 1 Hz for Semi-IPNs and two-roll mill blends. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

with the result of Figure 2(a), which can be explained as: For the P(MMA-BA) copolymer, the value of  $E''$  affected by the structure of chain segments and pendant groups. With the increase of BA ratio in the feeding, the larger butyl pendant groups in BA increase intermolecular friction and the more energy consumption, results in the increased  $E''$  value at the higher feed ratio of BA and MMA.

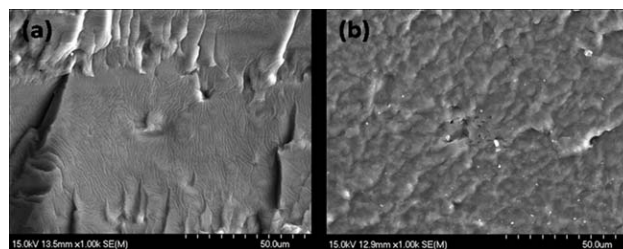
Figure 3 showed the  $\tan \delta$  (a) and  $E''$  (b) for NBR/P(MMA-BA) Semi-IPNs with various content of NBR [The feed ratio of BA and MMA at 60/40 (wt/wt)]. As shown in Figure 3(a), as the NBR content is added from 35 to 50 wt %, the peak values of  $\tan \delta$  near  $-20^\circ\text{C}$  for NBR increase from 0.55 to 0.9. However, the  $\tan \delta$  peak values of P(MMA-BA) at  $25^\circ\text{C}$  decrease correspondingly from 0.75 to 0.38, due to that the corresponding content of P(MMA-BA) in blends is reduced. Specially, with the ratio of NBR and P(MMA-BA) at 40/60 (wt/wt), a widely continuous damping platform temperature range ( $-30^\circ\text{C} < T < 80^\circ\text{C}$ ) with the promising  $\tan \delta$  peak height ( $\tan \delta > 0.45$ ) is formed, which has been obviously improved as compared with previous relating reports.<sup>13,20</sup> Furthermore, it can be aware that the glass transition temperature ( $T_g$ ) for both NBR and P(MMA-BA), specially for P(MMA-BA), shift inward gradually as the NBR content increases. The possible reason can be attributed to the forced compatibility of interpenetrating network structure, and with the suitable NBR/P(MMA-BA) ratio, NBR and P(MMA-BA) in blends can form a fully interpenetration network structure, resulting in respective  $T_g$  of polymers shifting inward.

Figure 3(b) further shows the temperature dependence of the  $E''$  for the Semi-IPNs system with various content of NBR. With the increase of NBR, the peak position of  $E''$  near  $-25^\circ\text{C}$  is almost unchanged, while the corresponding peak value increases obviously due to that NBR is dominating in the Semi-IPNs system. Comparatively, the height of the second peak which is assigned to the loss modulus peak of P(MMA-BA) is reduced, correspondingly.

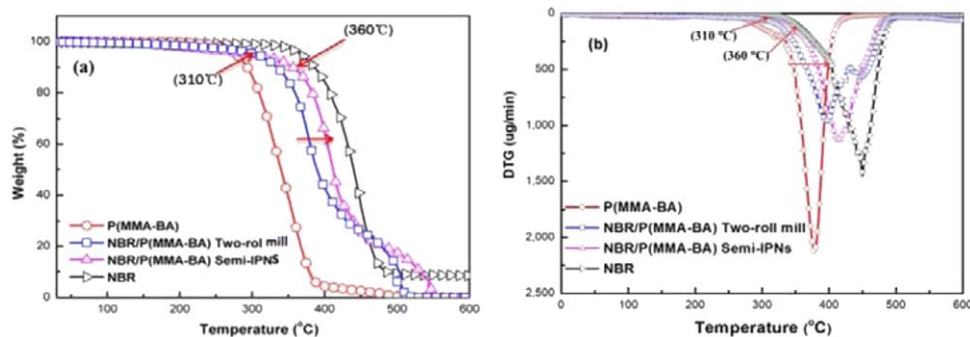
For further prove that Semi-IPNs of NBR/P(MMA-BA) had been prepared, the  $\tan \delta$  and  $E''$  for both the Semi-IPNs and the two-roll mill blends were studied comparatively. Figure 4(a) showed the  $\tan \delta$  for both Semi-IPNs and two-roll mill blends with the ratio of NBR/P(MMA-BA) at 40/60 (wt/wt) and the feed ratio of BA and MMA at 40/60 (wt/wt). As shown in the

figure, the two-roll mill blends of NBR/P(MMA-BA) shows a typical characteristic of the immiscible blends with two separate glass transition temperature ( $T_g$ ) at the respective positions of component polymers, and between the “two peaks,” a pronounced “valley” is obviously showed. Compared to the two-roll mill blends, the  $T_g$ s of NBR and P(MMA-BA) for the Semi-IPNs shift inward, and meanwhile, a broad and continued damping platform with wider temperature range ( $-30^\circ\text{C} < T < 80^\circ\text{C}$ ) as  $\tan \delta > 0.45$ , all of which indicate the formation of the typical interpenetrating network.<sup>27,28</sup> It can be deduced that the Semi-IPNs forces the miscibility between component polymers, producing microheterogeneous phases rather than macroheterogeneous phases in the blends, resulting in the  $T_g$  of component polymer to shift inward.<sup>13,27</sup> On the other hand, the acrylic esters in the Semi-IPNs are well-distributed, which have even greater friction force during the movement of chain segments than that of the two-roll mill blends system, which can also attributed to the improved damping properties.

Figure 4(b) showed the difference on the temperature dependence of  $E''$  for NBR, P(MMA-BA), Semi-IPNs and two-roll mill blends. As shown, NBR has a high value  $E''$  at a low temperature, while P(MMA-BA) presents one  $E''$  peaks at  $16^\circ\text{C}$ . For the Semi-IPNs and the two-roll mill blends, both present two  $E''$  peaks, which can be assigned to the contribution of NBR and P(MMA-BA), respectively. The peak position of  $E''$  for NBR and P(MMA-BA) in Semi-IPNs shift inward in contrast to in the two-roll mill blends. Still, the Semi-IPNs display a higher dynamic loss modulus than the two-roll mill blends in a wide temperature range. The improved damping properties can still be ascribed to the stronger friction between molecular chains produced by the interpenetrating network system.



**Figure 5.** SEM micrographs of NBR/P(MMA-BA): (a) Semi-IPNs and (b) two-roll mill.



**Figure 6.** TGA and DTG thermogram of P(MMA-BA), NBR, two-roll mill blends, and Semi-IPNs. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

### Scanning Electron Microscopy

The microstructure of the Semi-IPNs and the two-roll mill blends were analyzed by SEM. Figure 5 showed the morphology of NBR/P(MMA-BA) Semi-IPNs samples and NBR/P(MMA-BA)-based two-roll mill blends samples [NBR/P(MMA-BA) at 40/60 (wt/wt)]. Figure 5(a) shows that surface of the fracture pattern of Semi-IPNs is relatively smooth, while the fracture surface of the two-roll mill blends samples is rugged and many small particles can be observed on them [shown in Figure 5(b)]. The above results indicate clearly that two-roll mill process leads to poor dispersion for both P(MMA-BA) phase and NBR phase. However, the compatibility of NBR and P(MMA-BA) is improved evidently through the semi-interpenetrating polymer network, which will result in the formation of microheterogeneous morphology rather than macroheterogeneous phases and the improved damping properties.

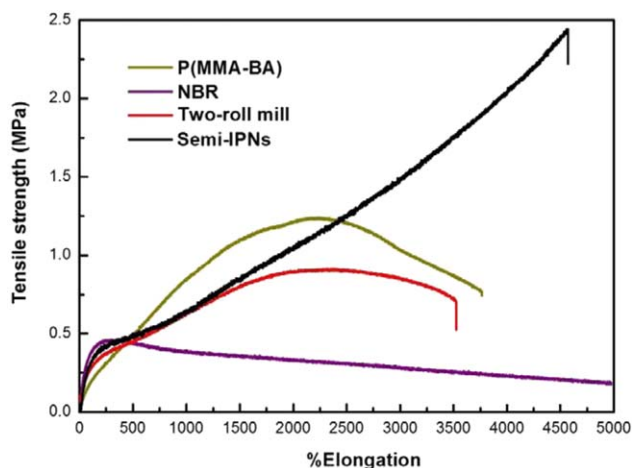
### Thermo Gravimetric Analysis (TGA)/Derivative Thermogravimetric Analysis (DTG) Studies

Figure 6 showed TGA and DTG of P(MMA-BA), NBR, NBR/P(MMA-BA) Semi-IPNs (40/60(wt/wt)) and the two-roll mill blends. As shown in Figure 6(a), P(MMA-BA) and NBR both have the decomposition temperature at around 240°C and 380°C, respectively. And, the decomposition temperature of

NBR/P(MMA-BA) blends by two-roll mill method is around 310°C with two obvious decomposition process. However, the Semi-IPNs show an improved thermal stability and its decomposition temperature is at around 360°C under the same testing conditions. DTG has also been provided in Figure 6(b), which present the change of weight loss rate of the measured samples. For the two-roll mill blend, it shows a typical characteristic of the immiscible blends with two peaks of weight loss rate, whereas the Semi-IPNs blend exhibits only one peak during the measurement. This can be attributed that the interpenetrating network structure of NBR/P(MMA-BA) which makes the incompatible two-phase be tangled together, resulting in the improved thermal stability of NBR.

### Tensile Mechanical Properties

Figure 7 showed the tensile mechanical properties of the P(MMA-BA), NBR, NBR/P(MMA-BA) Semi-IPNs (40/60(wt/wt)) and the two-roll mill blends, respectively, and the measured results are listed in Table I. As can be seen, the tensile strength and the elongation at break for Semi-IPNs are improved evidently in contrast to two-roll mill blends, and specially, Semi-IPNs exhibits the largest tensile strength of 2.48 MPa compared with measured samples. This increase is the indication of reinforcement by interpenetrating network structure. While, the lower tensile strength of two-roll mill blends than that of Semi-IPNs can be attributed to serious phase separation among NBR/P(MMA-BA) blends.



**Figure 7.** Tensile properties of NBR, P(MMA-BA), two-roll mill blends, and Semi-IPNs. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

### CONCLUSIONS

Semi-Interpenetrating Polymer Network (Semi-IPNs) based on nitrile rubber (NBR) and P(MMA-BA) were successfully synthesized by *in-situ* technology. DMA showed that with increasing the feed ratio of BA and MMA, the loss peak position of NBR

**Table I.** The Measured Results of Tensile Properties

Sample	Tensile strength (MPa)	% Elongation
NBR	0.46	>5000
P(MMA-BA)	1.24	3763
NBR/P(MMA-BA) two-roll mill	0.91	3521
NBR/P(MMA-BA) Semi-IPNs	2.48	4568

kept unchanged, while the peak position for P(MMA-BA) shifted to a lower temperature from 20 to  $-17^{\circ}\text{C}$ , and when the NBR content was added, the  $\tan \delta$  peak values for P(MMA-BA) increased and the  $\tan \delta$  peak values for NBR decreased. Specially, when the feed ratio of BA and MMA was 60/40 (wt/wt) and NBR/P(MMA-BA) ratio was 40/60 (wt/wt), the curve showed a broad damping platform with wider temperature range ( $-30^{\circ}\text{C} < T < 80^{\circ}\text{C}$ ) as  $\tan \delta > 0.45$ , which was superior to that of the NBR/P(MMA-BA)-based two-roll mill system. The results could be attributed to that the forced miscibility by Semi-IPNs, which could improve the distribution of component polymers in Semi-IPNs and increase the friction force during the movement of chain segments, resulting in the improved damping properties. SEM showed that NBR/P(MMA-BA) Semi-IPNs improved the component polymer's dispersion. Also, the tensile mechanical properties showed that the tensile strength and elongation at break for Semi-IPNs were improved evidently and the result of TGA proved that Semi-IPNs exhibited the better thermal stability. All the results showed that Semi-IPNs could be expected to be a good candidate for damping materials with good oil-resisted properties.

#### ACKNOWLEDGMENTS

The authors gratefully thank the National Natural Science Foundation of China (NSFC, No. 51103132, No. 51003095) and Research on Public Welfare Technology Application Projects of Zhejiang Province, China (2010C31121) for financial support. This work also was supported by the analysis and testing foundation of Zhejiang University of Technology.

#### REFERENCES

1. Buravalla, V. R.; Remillat, C.; Rongong, J. A.; Tomlinson, G. R. *Smart Mater. Bull.* **2001**, *8*, 10.
2. Zhu, Y.; Zhou, W.; Wang, J.; Wang, B.; Wu, J. R.; Huang, G. S. *J. Phys. Chem. B.* **2007**, *111*, 11388.
3. Yang, Y.Q.; Du, M.; Zheng, Q. *J. Found. Mater.* **2002**, *33*, 234.
4. Järvelä, P. T.; Li, S. C.; Järvelä, P. K. *J. Appl. Polym. Sci.* **1998**, *62*, 813.
5. Chiu, H. T.; Chiu, S. H.; Wu, J. H. *J. Appl. Polym. Sci.* **2003**, *89*, 959.
6. Fay, J. J.; Murphy, C. J.; Thomas, D.A.; Sperling, L. H. *Polym. Eng. Sci.* **1991**, *31*, 1731.
7. Qin, C. L.; Cai, W. M.; Cai, J.; Tang, D.Y.; Zhang, J. S.; Qin, M. *Mater. Chem. Phys.* **2004**, *85*, 402.
8. Trakulsujaritchok, T.; Hourston, D. J. *Eur. Polym. J.* **2006**, *42*, 2968.
9. Liu, Z.Y.; Wang, Y.Y.; Huang, G. S.; Wu, J. R. *J. Appl. Polym. Sci.* **2008**, *108*, 3670.
10. Ajayan, P. M.; Suhr, J.; Koratkar, N. *J. Mater. Sci.* **2006**, *41*, 7824.
11. Zhang, C.; Wang, P.; Ma, C. A.; Sumita, M. *J. Appl. Polym. Sci.* **2006**, *100*, 466.
12. Sperling, L. H. *J. Polym. Sci.* **1977**, *12*, 141.
13. Manoj, N. R.; Chandrasekhar, L.; Patri, M. B.; Chakraborty, C.; Deb P. C. *Polym. Adv. Technol.* **2002**, *13*, 644.
14. Manoj, N. R.; Ratna, D.; Dalvi, V.; Chandrasekhar, L.; Patri, M. B.; Chakraborty, C. P.; Deb C. *Polym. Eng. Sci.* **2002**, *42*, 1748.
15. Yu, W.W.; Zhang D. Z. M. *Eur. Polym. J.* **2013**, *49*, 1731.
16. Amiri Omarae, I.; Mohammadi, N. *Polym. Eng. Sci.* **2002**, *42*, 2328.
17. Chen, S. B.; Wang, Q. H.; Wang, T. M. *Polym. Test.* **2011**, *30*, 726.
18. Lipatov, Y. S.; Karabanova, L. V.; Mater, J. *J. Mater. Sci.* **1995**, *30*, 1095.
19. Hsieh, K. H.; Han, J. L.; Yu, C. T.; Fu, S. C. *Polymers* **2001**, *42*, 2491.
20. Samui, A. B.; Dalvi, V. G.; Patri, M.; Chakraborty, B. C.; Deb, P. C. *J. Appl. Polym. Sci.* **2004**, *91*, 354.
21. Sperling, L. H. *Polym. Sci.* **1989**, *6*.
22. Patri, M.; Samui, A. B.; Chakraborty, B. C.; Deb, P. C. *J. Mater. Sci.* **1994**, *65*, 549.
23. Mathew, A.; Deb, P. C. *J. Mater. Sci.* **1994**, *53*, 1104.
24. Omran, A. M.; Youssef, A. M.; Ahmed, M.; Abdel-Bary, E. M. *J. Elastom. Plast.* **2010**, *63*, 197.
25. Patri, M.; Reddy, C. V.; Narasimhan, C.; Samui, A. B. *J. Appl. Polym. Sci.* **2007**, *103*, 1120.
26. Gómez Jiménez, M. A.; Rivera Armenta, J. L.; Mendoza Martínez, A. M.; Robledo Muñoz, J. G.; Rangel Vazquez, N. A.; Terres Rojas, E. *Lat. Am. Appl. Res.* **2009**, *39*, 2.
27. Manoj, N. R.; Raut, R. D.; Sivaraman, P.; Ratna, D.; Chakraborty, B. C. *J. Appl. Polym. Sci.* **2005**, *96*, 1487.