Polyimide resins and their carbon fiber reinforced composites

Biao Liu, Mian Ji, Aijun Hu, Lin-Fan, Shi-yong Yang*

Laboratory of Advanced Polymer Materials, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, P. R. China, E-mail:

Carbon fiber reinforced polyimide composites, due to their unique combination of thermal and mechanical properties, have been widely used in recent years.[1] The method to obtain polyimides composites with good processability and toughness has always been a scientific and technical challenge. PETI-5 composite is the most outstanding one which has a CAI value > 300MPa.[2] There are still some improvements needed for this material. In order to fabricate composites with better quality, a resin with better melt flow is required. Moreover, a precursor solution with high solid content, low viscosity, and solvents which can be easily removed is also needed.[3] These requirements call for more challenges for the fabrication of high performance polyimide composites.

In present work, a series of biphenyl-type imide oligomers end-capped by 4-phenylethynyl phthalic anhydride (4-PEPA) were synthesized through a modified PMR route. Stable resin solutions with solid content of 50% and viscosity of 40-50 cP were obtained. The solvents and volatiles of the resins were easily removed at low temperature as shown in **Fig.1**. B-stage resins with different copolymerization ratios and molecular weights were discussed by TGA, DSC and FTIR.

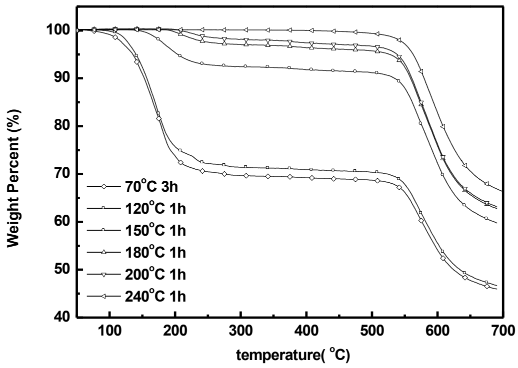

Fig.1 TGA curves of PI-1 after different thermal treatments

Different mole ratios of a-BPDA were incorporated into the oligomer backbones, and the processibility was significantly improved when the a-BPDA content reached 20% (PI-3) or higher (**Fig. 2**). This result should be attributed to the asymmetric structure of a-BPDA. After cured, PI-3 and PI-4, which had a molecular weight of 5000 g/mol, exhibited excellent mechanical properties as shown in **Table 1**. PI-3, which had the best overall properties with tensile strength >120MPa, flexural strength >150MPa and elongation at break >18%, was fabricated into carbon fiber laminates.

Table 1 Characterizations and properties of cured polyimide resins

Samples	TGA			DMA		Tensile properties			Flexural properties	
	T_d (°C)	T_5 (°C)	R_{700} (%)	E' (℃)	tan δ (℃)	Strength (MPa)	Modulus (GPa)	Elongation (%)	Strength (MPa)	Modulus (GPa)
PI-3	546	548	69.3	273	283	124.3	2.0	18.8	154.8	3.1
PI-4	538	540	69.8	270	286	124.8	2.0	16.2	157.0	3.2

Through an optimized curing procedure, unidirectional and quasi-isotropic laminates were fabricated with good quality. After impacted at an energy of 6.7 KJ/m, only a small damage could be observed in the center of the laminate indicated by C-scan (**Fig.3**). High retention of mechanical properties at elevated temperature (~250□) and high damage tolerance were obtained on the composites laminates as shown in **Table 2**. These properties indicated a promising potential for future aerospace application.

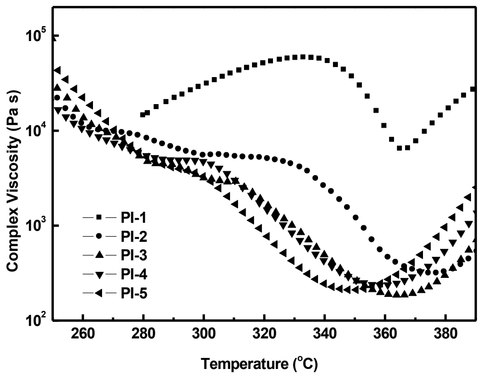

Fig.2 Rheology behaviors

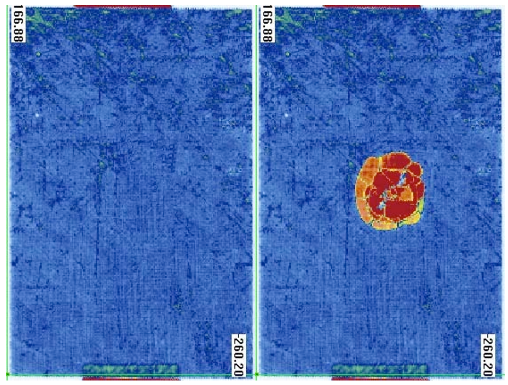

Fig.3 C-scan images before and after impact

Table 2 Mechanical properties of the laminates

	Lay-up	Properties
0° Flexural strength, MPa		
RT	$[0°]_{12}$	1536.8±86.7
177 □		1137.9±71.8
250 □		819.1±36.2
0° Flexural modulus, GPa		
RT	$[0°]_{12}$	143.8±4.6
177 □		144.4±3.3
250 □		140.0±0.9
CAI strength, MPa	$[-45°,0°,45°,90°]_{3s}$	229.2±13.2
OHC strength, MPa	$[-45°,0°,45°,90°]_{2s}$	303.2±17.5
OHT strength, MPa	$[-45°,0°,45°,90°]_{2s}$	366.9±49.9

Acknowledgment

Funding from the National Natural Science Foundation of China (NSFC) for distributed Young Scholars (No5090308.) is gratefully acknowledged.

References

1. Pater, R.H. and P.A. Curto, *Advanced materials for space applications. Acta Astronautica.*, **61**(11-12), 1121-1129(2007).
2. Smith Jr, J., J. Connell, and P. Hergenrother, *The effect of phenylethynyl terminated imide oligomer molecular weight on the properties of composites. Journal of Composite Materials.*, **34**(7), 614(2000).
3. Hou, T.H., B.J. Jensen, and P.M. Hergenrother, *Processing and properties of IM7/PETI composites. Journal of Composite Materials.*, **30**(1), 109-122(1996).