

**Design and Preparation of Bio-based Dielectric Elastomer with Polar and
Plasticized Side Chains**

Weiwei Lei,^a Runguo Wang,^a Dan Yang,^d Guanyi Hou,^a Xinxin Zhou,^a He Qiao,^a

Wencai Wang,^a Ming Tian,^a Liqun Zhang^{*abc}

^aState Key Laboratory for Organic–Inorganic Composites, Beijing University of Chemical Technology, Beijing 100029, China.

E-mail: zhanglq@mail.buct.edu.cn; Fax: +86-10-64456158; Tel: +86-10-64443413

^bKey Laboratory of Beijing City for Preparation and Processing of Novel Polymer Materials, Beijing University of Chemical Technology, Beijing 100029, China.

^cBeijing Engineering Research Center of Advanced Elastomers, Beijing University of Chemical Technology, Beijing 100029, China.

^dBeijing Institute of Petrochemical Technology, Beijing 102600, China.

Table S1. Glass transition temperature (T_g) of poly (di-*n*-alkyl itaconates) (PDAI) ^{1,2}

PDAI	$T_g/^\circ\text{C}$
Dimethyl (PDMI)	95
Diethyl (PDEI)	58
Di- <i>n</i> -propyl (PDPrI)	34
Di- <i>n</i> -butyl (PDBI)	12
Di- <i>n</i> -pentyl (PDPeI)	5
Di- <i>n</i> -hexyl (PDHxI)	-18
Di- <i>n</i> -heptyl (PDHpI)	-23
Di- <i>n</i> -octyl (PDOI)	-20
Di- <i>n</i> -nonyl (PDNI)	-11
Di- <i>n</i> -decyl (PDDI)	-3

In Table S1, the T_g of PDAI decrease first as the side chains increase in length from methyl to heptyl. PDAI bearing the flexible pendant groups are similar to the small-molecular additives by pushing the chains further apart and thereby producing a change in T_g . The T_g increasing is observed from heptyl to decyl for the developing order tendency of the long side chains. Considering the polarity of di-*n*-alkyl itaconates also, di-*n*-butyl itaconate was used to synthesize the dielectric elastomer. Furthermore, bio butanol is a promising biofuel product in the word.

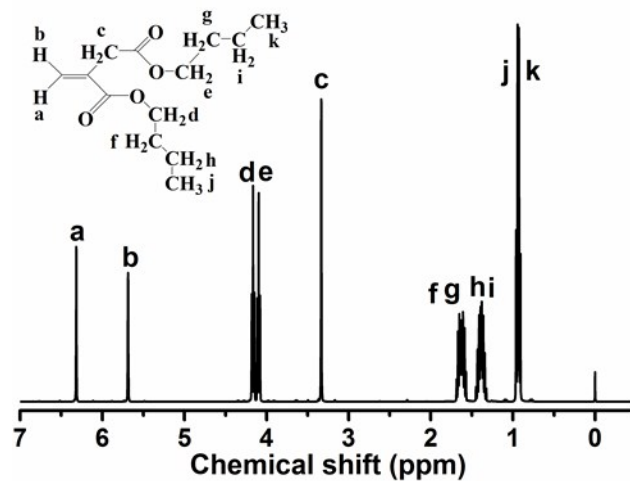


Figure S1. ¹H NMR spectrum of di-*n*-butyl itaconate (DBI)

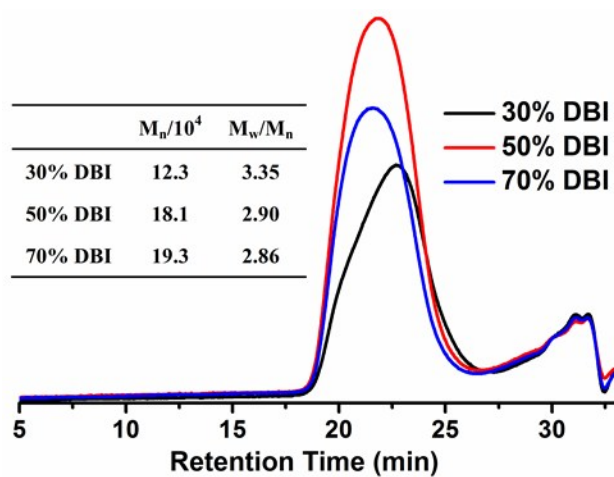


Figure S2. GPC traces for PDBII with different itaconate to isoprene feed ratios. (DBI: di-*n*-butyl itaconate, i.e., 30% DBI means 30 wt % DBI in the feed. In inserted table, M_n : number-average molecular weight, M_w/M_n : polydispersity ratio (weight average molecular weight/number-average molecular weight), by GPC (polystyrene calibration))

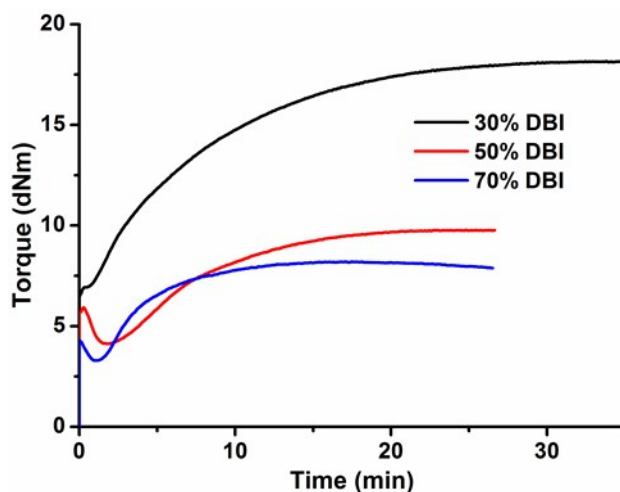


Figure S3. The curing curves of PDBII with different content of DBI in the feed. (70% DBI stands for 70% DBI in the feed.)

Table S2. The optimum cure time of PDBII with different content of DBI in the feed. (70% DBI stands for 70% DBI in the feed.)

Formulations	T90 (min:s)
70% DBI	9:36
50% DBI	14:52
30% DBI	17:39

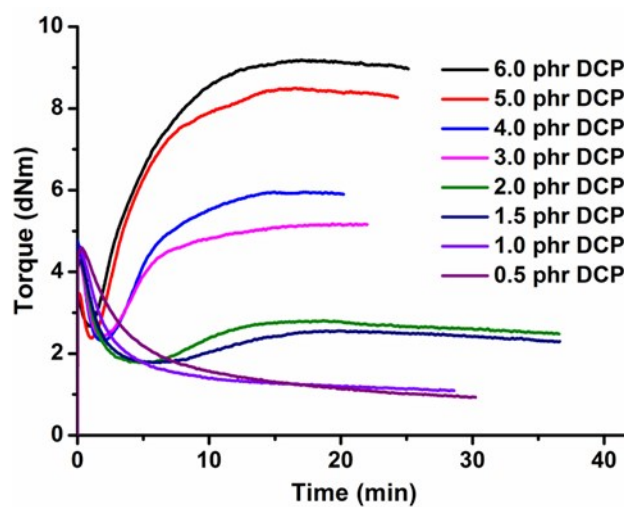


Figure S4. The curing curves of PDBII with 70% di-*n*-butyl itaconate in the feed crosslinked by

different content of DCP.

Table S3. The optimum cure time of PDBII with 70% di-*n*-butyl itaconate in the feed crosslinked by different content of DCP.

Formulations	T90 (min:s)
0.5 phr DCP	-
1.0 phr DCP	-
1.5 phr DCP	16:14
2.0 phr DCP	13:25
3.0 phr DCP	11:11
4.0 phr DCP	10:33
5.0 phr DCP	9:58
6.0 phr DCP	9:56

Table S4. Curing characteristics and elastic modulus of BaTiO₃ filled PDBII with 70% di-*n*-butyl itaconate in the feed.

Items	BaTiO ₃ /phr					
	0	10	30	50	70	90
S _{max} (dNm)	8.50	8.05	8.60	10.81	11.27	9.82
S _{min} (dNm)	2.38	1.51	1.57	1.67	1.72	1.72
ΔS(dNm)	6.12	6.54	7.03	9.14	9.55	8.10
Elastic modulus/MPa	0.420	0.146	0.180	0.214	0.290	0.158
T90 (min:s)	8:49	12:32	12:30	10:41	10:21	11:47

Table S5. Comparing actuation strain of PDBII elastomer with other dielectric elastomers.

Material	Prestrain (x,y) (%)	Area strain ^a (%)	Field Strength ^a (kV/mm)	Ref.
PANI-g-PolyCuPc-g-PU	0, 0	7	23	3
23 wt %PANI/P(VDF-TrFE-CTFE)	0, 0	1.5	9.5	4
PANI@PDVB/PDMS	0, 0	12	54	5
14PANI/15PolyCuPc/85PU	0, 0	9.3	20	3
SEBS-MA grafted PANI	0, 0	1.4	27	6
NBR/TiO ₂ /DOP	0, 0	3.04	20	6
5 wt % CNTs/PDMS	0, 0	4.4	1.5	7
P(VDF-TrFE)/40 wt %CuPc	0, 0	1.91	13	8
Polyester elastomer	0, 0	11.9	15.6	9
PDBII	0,0	14	20	
	0,0	20	30	
	0,0	25	40	

a. Estimated from graphical data in cited reference

References

- 1 J. Cowie, S. Henshall and I. McEwen, *Polymer*, 1977, **18**, 612-616.
- 2 J. Cowie, Z. Haq, I. McEwen and J. Velickovic, *Polymer*, 1981, **22**, 327-332.
- 3 C. Huang and Q. M. Zhang, *Adv. Mater.*, 2005, **17**, 1153-1158.
- 4 C. Huang and Q. M. Zhang, *Adv. Funct. Mater.*, 2004, **14**, 501-506.
- 5 M. Molberg, D. Crespy, P. Rupper, F. Nuesch, J. A. E. Manson, C. Loewe and D. M. Opris, *Adv. Funct. Mater.*, 2010, **20**, 3280-3291.
- 6 H. Stoyanov, M. Kolloosche, D. N. McCarthy and G. Kofod, *J. Mater. Chem.*, 2010, **20**, 7558-7564.
- 7 L. Z. Chen, C. H. Liu, C. H. Hu and S. S. Fan, *Appl. Phys. Lett.*, 2008, **92**, 263104-263106.
- 8 Q. M. Zhang, H. F. Li, M. Poh, F. Xia, Z. Y. Cheng, H. S. Xu and C. Huang, *Nature*, 2002, **419**, 284-287.
- 9 D. Yang, M. Tian, H. Kang, Y. Dong, H. Liu, Y. Yu and L. Zhang, *Mater. Lett.*, 2012, **76**, 229-232.