

Synthesis and application as polymer electrolyte of hyperbranched copolyethers derived from cationic ring-opening polymerization of 3-{2-[2-(2-methoxyethoxy)ethoxy]ethoxy}methyl- and 3-hydroxymethyl-3'-methyloxetane

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Abstract

A kind of novel hyperbranched copolyethers intending for the solid polymer electrolyte was synthesized *via* the cationic ring-opening polymerization of 3-{2-[2-(2-methoxyethoxy)ethoxy]ethoxy}methyl-3'-methyloxetane (MEMO) and 3-hydroxymethyl-3'-methyloxetane (HMO) in the presence of $\text{BF}_3 \cdot \text{Et}_2\text{O}$ as an initiator. Herein HMO was employed to create the hyperbranched structure, whereas MEMO was responsible for the ionic transportation of the resulting copolymers. The terminal structure featured by a cyclic fragment was definitely detected by MALDI-TOF measurement. The degree of branching of the copolymers was calculated by means of ^{13}C NMR spectra. The DSC analysis implied that they hold the excellent segment motion performance and perfectly amorphous state beneficial for the ionic transportation. The ionic conductivity measurements showed that the sample HMO 30 reaches a maximum ionic conductivity of $8.0 \times 10^{-5} \text{ S/cm}$ at 30°C and $7.4 \times 10^{-4} \text{ S/cm}$ at 80°C , respectively, after doping with lithium salt LiTFSI. Moreover, the TGA assay exhibited that these hyperbranched copolymers possess the higher thermostability as compared with their liquid counterparts.

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1. Introduction

In recent years, hyperbranched polyethers made from oxetane-derived monomers *via* the cationic ring-opening polymerization (CROP) have gained widespread attention due to their one-pot synthetic protocol, unique three-dimensional architecture and potential applications in the fields of materials science as well as in homogeneous catalysis [1–12]. Alternatively, the new and improved material properties can be afforded through further modifying and functionalizing the

architecture of the hyperbranched polyethers. Until now, however, only few reports about their preparations and applications in the area of solid polymer electrolytes (SPEs) used in lithium ion battery have been disclosed [13].

Currently, SPEs have aroused much interest for their potential application in powerful batteries such as cells for electric vehicles. Due to the higher safe performance, they are considered to be the most suitable replacement of liquid electrolytes [14,15]. In our previous work [16], a hyperbranched polyether, poly(3-{2-[2-(2-hydroxyethoxy)ethoxy]ethoxy}methyl-3'-methyloxetane) (PHEMO) comprising triethylene glycol oligomer as ionic conducting group was synthesized *via* CROP and evaluated as candidate for SPEs. As is well known, the motion ability of ionic conducting group is the key to the ionic

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conducting performance of polymer electrolytes. However, the triethylene glycol group in PHEMO was directly bound to the hyperbranched backbones leading to a higher chain motion hindrance. As a consequence, its relatively lower ionic conductivity locating in 5.6×10^{-5} S/cm at 30 °C and 6.3×10^{-4} S/cm at 80 °C, respectively, still needs to be improved.

If the triethylene glycol oligomer is allowed to liberate from the hyperbranched backbones of PHEMO as a pendant group, it can impart the polymer the higher motion performance to benefit the ionic transportation significantly [17]. Accordingly, a kind of novel hyperbranched copolyethers was designed and synthesized *via* CROP of 3-[2-[2-(2-methoxyethoxy)ethoxy]ethoxy]methyl-3'-methyloxetane (MEMO) and 3-hydroxymethyl-3'-methyloxetane (HMO) in the present study. The monomer MEMO was herein incorporated to afford the pendant triethylene glycol oligomer a high flexibility to benefit the ionic conductivity, and HMO was bound to construct the hyperbranched structure to afford the resulting copolymers a lower glass transition temperature (T_g) as well as an amorphous state.

2. Experimental

2.1. Materials and measurement

All reagents used in this work were commercial products of analytical grade and available as described elsewhere [16,18,19].

FTIR spectra were measured with Shimadzu IR Prestige-21. ^1H and ^{13}C NMR spectra were recorded on Bruker ARX400 using CDCl_3 as solvent containing TMS as internal standard. All the ^{13}C NMR spectra of the copolymers were obtained by the inverted gate method with a pulse delay of 10 s. Gel permeation chromatographic (GPC) analysis was made on Waters 2414 with three Waters styragel HMW columns at 35 °C using THF as eluent. From the GPC results, the molecular weight was determined as calibrated with polystyrene standards. Netzsch PC-200 was used for an analysis of the thermal behavior of the polymers at a heating rate of 10 °C/min. The polymer samples (6–10 mg) were heated from –100 °C to 100 °C in stainless steel pans. The results of the second run were used for the glass transition temperature investigation. Thermogravimetric analysis (TGA) was conducted with TA 2000 thermogravimeter under N_2 atmosphere at a heating rate of 20 °C/min. MALDI-TOF mass measurement was carried on Bruker BIFLEX III with 2-cyano-4-hydroxycinnamic acid (CCA) as

matrix. The ionic conductivity was evaluated using CHI 660A Electrochemistry Station made in Chen Hua Instrument Company, Shanghai, China. The measurements were carried out in the range from 30 °C to 80 °C with an interval of 5 °C.

2.2. Synthesis and characterization of monomers

Synthesis of two monomers MEMO and HMO were described in our previous work [18,19]. Their characterization is as follows.

MEMO, FTIR/ cm^{-1} : 2870 (CH_2), 1112 (linear C–O–C), 969 (ring's C–O–C); ^1H NMR/ppm: 1.31 (3H, CH_3), 3.38 (3H, CH_3O), 3.54–3.56 (2H, $\text{CH}_2\text{—OCH}_3$), 3.57 (2H, C– $\text{CH}_2\text{—O}$), 3.64–3.67 (10H, O– $\text{CH}_2\text{—CH}_2$, $\text{CH}_2\text{—CH}_2\text{—O—CH}_3$), 4.34–4.53 (4H, ring's CH_2).

HMO, FTIR/ cm^{-1} : 3404 (O–H), 1047 (C–O), 969 (ring's C–O–C); ^1H NMR/ppm: 1.23 (3H, CH_3), 3.14 (1H, OH), 3.60 (2H, $\text{CH}_2\text{—O}$), 4.31–4.47 (4H, ring's CH_2).

2.3. Copolymerization

The cationic ring-opening copolymerization of the monomer HMO and MEMO proceeded at 0–5 °C (ice-water bath) in solution. The solvent CH_2Cl_2 was added into a dry round flask under N_2 atmosphere. After the two monomers were added, the initiator $\text{BF}_3 \cdot \text{Et}_2\text{O}$ at a molar feed ratio of $M/I = 50$ was introduced into the flask. The reaction was continued for 24 h and terminated by adding methanol. The obtained hyperbranched copolyethers were purified by precipitation into hexane three times, and dried *in vacuo*. The experimental results are summarized in Table 1. For the convenience, the samples were coded according to the HMO content in the molar feed ratios.

2.4. The end blockage of hyperbranched polyethers and preparation of polymer electrolytes

The aforementioned hyperbranched copolyether of 3 g was dissolved in 5 ml THF. Then, 5 g of acetic anhydride was added under N_2 atmosphere, and the resultant solution was maintained at 85 °C for 6 h. Finally, the blocked polymer was purified by precipitation into hexane three times, and dried *in vacuo*.

The end-capped copolyether was mixed with lithium trifluoromethanesulfonimide (LiTFSI) in THF solution and dried

Table 1
Experimental results of hyperbranched polyethers derived from MEMO and HMO

Sample code	M/I	Feeding ratio HMO:MEMO	Found ratio HMO:MEMO	M_n	M_w	M_w/M_n	DB/%
HMO 30	50	30:70	35.9:64.1	6800	9700	1.44	14.8
HMO 40	50	40:60	38.7:61.3	6200	8200	1.32	16.7
HMO 50	50	50:50	42.1:57.9	6200	8500	1.37	16.9
HMO 60	50	60:40	60.1:39.9	5600	7000	1.25	16.6
HMO 70	50	70:30	68.5:31.5	6100	8500	1.41	23.6

in vacuo before the ionic conductivity measurement on the Electrochemistry Station.

3. Results and discussion

3.1. Synthesis of hyperbranched copolymers

The synthetic pathway of hyperbranched copolymers is outlined in Scheme 1. The CROP of MEMO and HMO was carried out in methylene chloride solution. Since HMO itself contains a hydroxymethyl group, it was used not only as a monomer but also as a co-initiator to interact with boron trifluoride etherate to release a hydrogen proton to active other monomers to initiate CROP.

The CROP mechanism of oxetane-derived monomers had been well described in literatures [3–11,16]. It revealed that the chain propagation is actually a result of the competition or mutual conversion of two propagation mechanisms, i.e. active chain end (ACE) and activated monomer (AM) mechanisms. As depicted in Scheme 2, during the copolymerization of MEMO and HMO, the branching processes stemmed from the nucleophilic attack of the pendant hydroxymethyl of HMO on the activated HMO monomers. As the continuous branching reaction occurred, the hyperbranched structure was created.

A typical GPC trace of the resulting hyperbranched copolyethers is shown in Fig. 1, and the GPC analytical results of

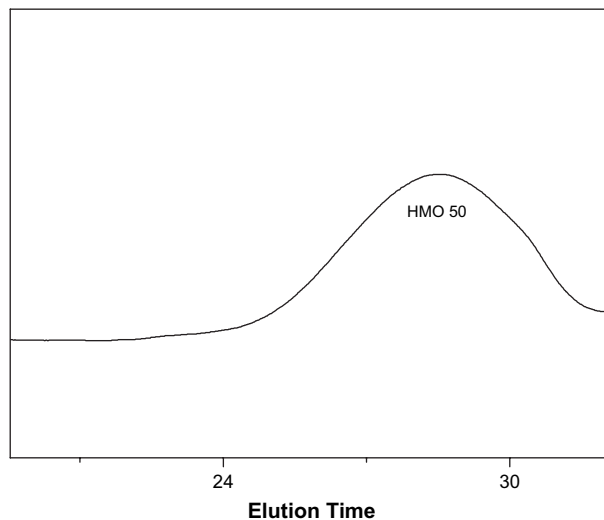
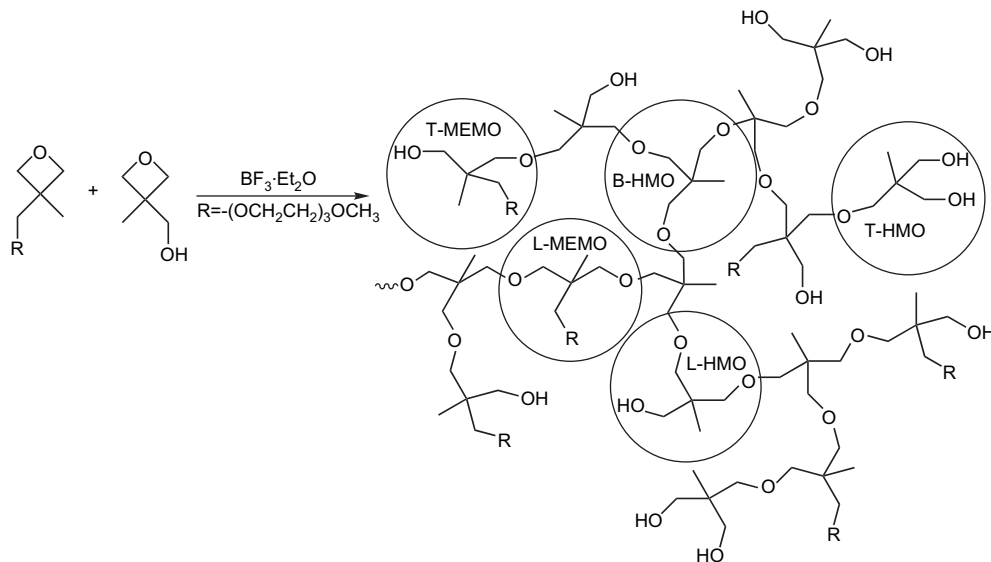
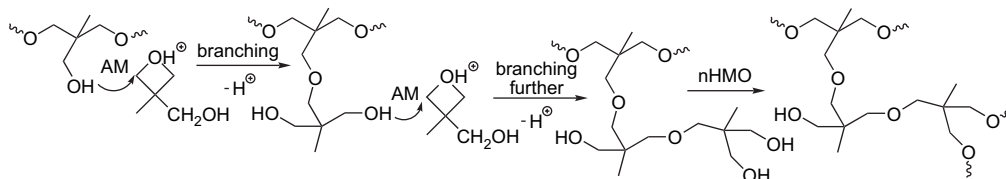


Fig. 1. GPC trace of the hyperbranched copolyether HMO 50.

all the samples are summarized in Table 1. As can be seen, a nearly symmetric and narrow molecular weight distribution revealed that the copolymerization of MEMO and HMO had taken place. Additionally, it should be addressed that the molecular weights of these copolyethers with the hyperbranched attribute are underestimated by GPC analysis when they are calibrated with linear standard PS samples.



Scheme 1. Cationic ring-opening copolymerization of MEMO and HMO.



Scheme 2. Branching process of hyperbranched polyethers according to AM mechanism.

3.2. MALDI-TOF mass spectrum

According to the CROP mechanism mentioned above, with increasing the concentration of hydroxymethyl group in the reaction system, the termination of the propagation chain involving the back-biting process of hydroxymethyl group becomes dominant and finally leads to the formation of a cyclic structure at the chain terminals. In general, this chain termination process is a result of the intramolecular reaction as illustrated in Scheme 3. The cyclic fragment structure can be clearly demonstrated by the following MALDI-TOF mass spectrum of the sample HMO 50 as showed in Fig. 2.

As shown in Fig. 2, the mass peaks can be expressed by Eq. (1).

$$M = M_{\text{Na}} + nM_{\text{HMO}} + mM_{\text{MEMO}} + M_t \quad (1)$$

where M_{Na} is the mass of Na^+ , and M_t is the mass of the end group and corresponds to a particular chemical structure at the chain end [20,21]. Since an intact macromolecule is routinely cationized by the attachment of Na^+ to form the $M + \text{Na}^+$ molecular ions during the MALDI-TOF measurement, the mass peaks obtained in the spectrum contain the mass of $M + \text{Na}^+$. Furthermore, if a cyclic structure exists at the chain end, M_t should be zero. All the observed peaks in this spectrum are calculated based on Eq. (1), and the results are summarized in Table 2. It was found that all the M_t s are equal to zero after subtracting the mass of HMO and MEMO repeating units. This clearly showed that the formation of the cyclic end structure is characteristic of the CROP of MEMO and HMO due to the inevitable intramolecular chain transfer or back-biting process as discussed above.

3.3. FTIR analysis

The IR spectra of the hyperbranched copolyethers are illustrated in Fig. 3. Compared with the monomers, the disappearing of the absorbance peak at 970 cm^{-1} indicated that the CROP had occurred. The other peaks of the resulting copolymers were assigned as follows: 3480 (O-H) , $2876 \text{ (CH}_2\text{)}$ and $1109 \text{ (C-O-C)} \text{ cm}^{-1}$. It was observed that the OH absorbance peak becomes stronger with increasing the content of HMO in the feeding composition, implying that the proportion of hydroxyl groups in the copolyethers increases with the molar concentration of HMO.

3.4. NMR identification

Fig. 4 shows the typical ^1H NMR spectrum of the hyperbranched copolymer HMO 50. Compared with the monomers, the disappearing of quartet peaks appeared at $4.3\text{--}4.5 \text{ ppm}$

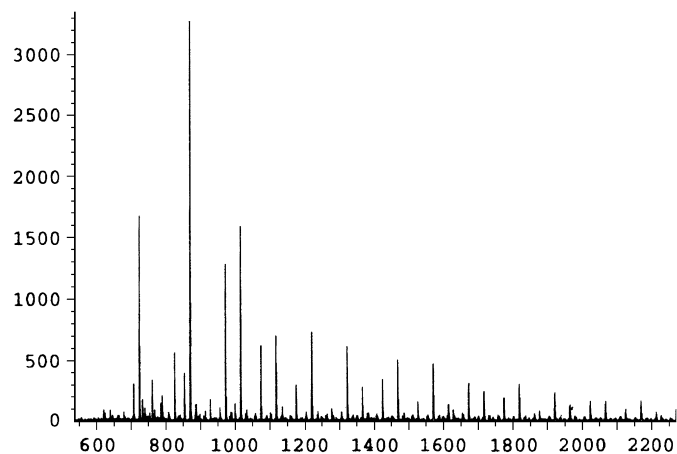


Fig. 2. Typical MALDI-TOF mass spectrum of the hyperbranched copolyether HMO 50.

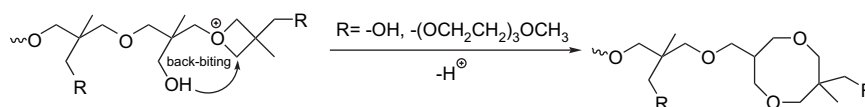
Table 2

Measured molecular masses of hyperbranched polyethers by MALDI-TOF

Measured mass	n (HMO)	M (MEMO)	M_{Na}	M_t
869.8	1×102.13	3×248.32	22.99	0
1118	1×102.13	4×248.32	22.99	0
1366.1	1×102.13	5×248.32	22.99	0
1614.2	1×102.13	6×248.32	22.99	0
723.8	2×102.13	2×248.32	22.99	0
971.9	2×102.13	3×248.32	22.99	0
1220	2×102.13	4×248.32	22.99	0
1468.1	2×102.13	5×248.32	22.99	0
1716.3	2×102.13	6×248.32	22.99	0
1964.5	2×102.13	7×248.32	22.99	0
825.8	3×102.13	2×248.32	22.99	0
1074	3×102.13	3×248.32	22.99	0
1322.1	3×102.13	4×248.32	22.99	0
1570.2	3×102.13	5×248.32	22.99	0
1818.4	3×102.13	6×248.32	22.99	0
927.9	4×102.13	2×248.32	22.99	0
1176	4×102.13	3×248.32	22.99	0
1424.1	4×102.13	4×248.32	22.99	0
1672.3	4×102.13	5×248.32	22.99	0
1920.5	4×102.13	6×248.32	22.99	0
1526.2	5×102.13	4×248.32	22.99	0
1774.4	5×102.13	5×248.32	22.99	0
2022.6	5×102.13	6×248.32	22.99	0

also clearly indicated that the CROP had taken place, which is well consistent with the result of FTIR analysis as mentioned above. The main resonance peaks were assigned as follows: $\delta = 0.83\text{--}0.91$ (CH_3 , in both HMO and MEMO), 3.38 (CH_3O , in MEMO) and 4.26 (OH) ppm. Consequently, the molar ratios of HMO to MEMO incorporated into the copolyethers can be obtained according to Eq. (2).

$$\text{HMO} : \text{MEMO} = (A_{0.83\sim 0.91}/3 - A_{3.38}/3) : (A_{3.38}/3) \quad (2)$$



Scheme 3. "Back-biting" process of the CROP of MEMO and HMO.

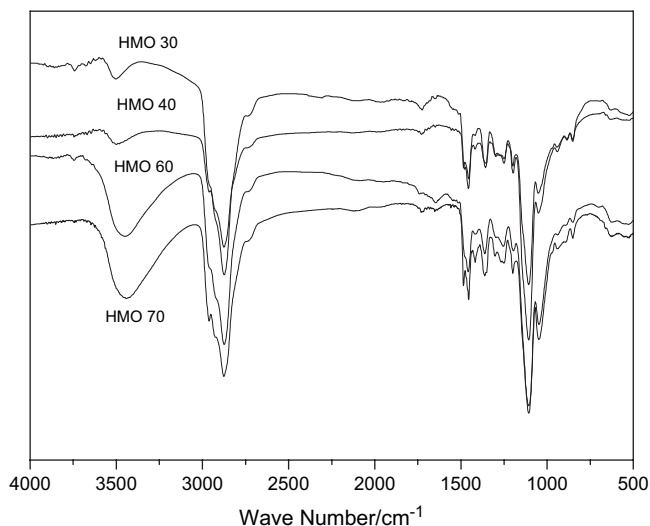


Fig. 3. FTIR spectra of the hyperbranched copolyethers.

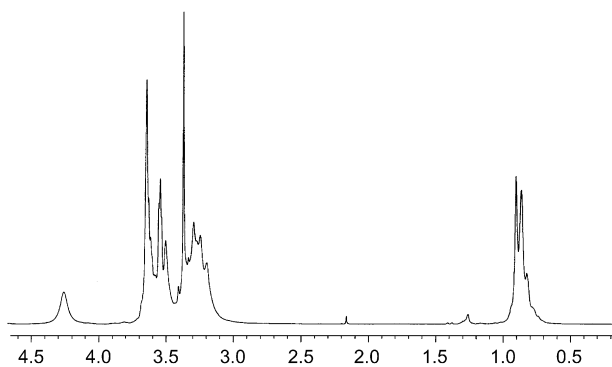


Fig. 4. ¹H NMR spectrum of the hyperbranched copolyether HMO 50.

where A_x represents the area of the corresponding ¹H NMR resonance peak at x ppm. The calculated results are summarized in Table 1. It was noticed that the proportion of the HMO repeating units in the hyperbranched copolyethers increases with the HMO content in the feeding composition. In addition, the resonance peak appeared at 4.26 ppm was ascribed to the terminal hydroxyl group. It was unshielded by the interaction of hydrogen bonding so that its resonance peak emerged at such a lower field. This phenomenon was also observed in other hyperbranched polymers containing a great number of terminal OH [16].

The degree of branching (DB) is a key parameter to characterize the hyperbranched structure of the resulting copolymers. The ¹³C NMR resonance peaks of the quaternary carbons in hyperbranched polyethers are widely utilized to determine the DB [1–12]. As is well known, the microstructure of hyperbranched polyethers is often divided into three types of which are terminal, linear and branched groups. For the hyperbranched structure derived from HMO, these three structures are all involved and outlined in Scheme 1 labeled as T-HMO, L-HMO and B-HMO, respectively. For the polyether formed from MEMO, however, because it contains no hydroxymethyl moiety in its own molecule to produce a branched

microstructure, only two microstructures represented as T-MEMO and L-MEMO are fitted to its terminal and linear structures as shown in Scheme 1. Fig. 5 exhibits the ¹³C NMR spectra of the quaternary carbons of the as-prepared hyperbranched copolyethers, where five resonance peaks clearly correspond to five types of microstructures. It was found that with increasing the HMO content in the composition, the right three peaks become stronger, while the left two peaks get weaker. Thence the right three peaks were deduced to derive from HMO, and the left two peaks to result from MEMO. From left to right, these five resonance peaks were assigned to T-MEMO, L-MEMO, T-HMO, L-HMO and B-HMO, respectively. Among them, the resonance peaks of T-HMO and L-HMO were too close to distinguish each other, and especially in the cases of the high feeding HMO contents where these two peaks became actually overlapped. Consequently, the DB of the resulting copolyethers was only calculated as follows:

$$\text{DB}\% = \text{B}/(\text{B} + \text{L} + \text{T}) \quad (3)$$

where B, L and T correspond to the integral areas of the corresponding resonance peaks of the branched, linear and terminal quaternary carbons. However, it is noteworthy that some researchers used the equation $(\text{B} + \text{T})/(\text{B} + \text{L} + \text{T})$ to calculate the DB. In the present paper, as the L and T were overlapped in the cases of the high HMO concentrations, Eq. (3) was employed to calculate the DB of the hyperbranched polyethers. As summarized in Table 1, the DB value was found to increase with the HMO content in the feeding composition. Since the hyperbranched structure was independently created by HMO, it was well understood that the more HMO was added, there were the more chances for the propagating copolyether chains to increase the DB value spontaneously.

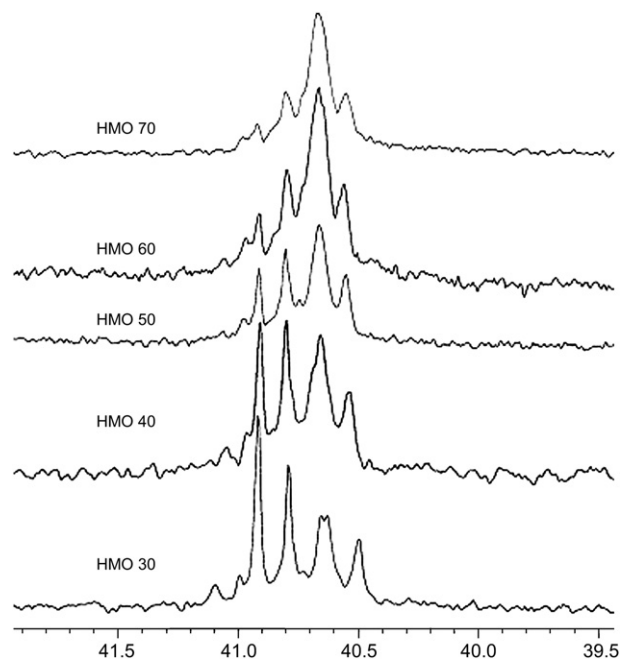


Fig. 5. ¹³C NMR spectra of the quaternary carbons of the hyperbranched copolyethers.

3.5. DSC measurement

DSC thermograms of the resulting hyperbranched copolyethers are shown in Fig. 6. No crystalline melting peak was found in all the thermograms due to the perfectly amorphous state of the hyperbranched copolyethers. Additionally, only one glass transition temperature (T_g) was observed in each curve, indicating that the product prepared by CROP is the copolyether rather than the mixture of two homopolyethers. Furthermore, the T_g decreases with the HMO content in the feeding composition. As demonstrated in our previous work [18,19], such a lower T_g as well as an amorphous state significantly benefits the improvement of the ionic conductivity. Herein, HMO 30 holding the lowest T_g was expected to possess the highest ionic conductivity among all the resulting hyperbranched copolyethers in the present study.

3.6. Ionic conductivity measurement

The lithium salt LiTFSI was found to be most favorable for improving the ionic conductivity of the solid polymer electrolytes [22]. Prior to the ionic conductivity measurement, the hydroxyl end groups of the resulting hyperbranched polymers were end-capped by acetic anhydride and then doped with LiTFSI to yield a polymer electrolyte. Fig. 7 shows the plots of the ionic conductivity changes of the hyperbranched polymers vs. the reciprocal of temperature. The sample HMO 30 indeed gave rise to a maximum ionic conductivity of 8.0×10^{-5} S/cm at 30 °C and 7.4×10^{-4} S/cm at 80 °C. Taking into account the DSC results, it was reasonable that this sample exhibits the highest ionic conductivity among all the as-prepared hyperbranched copolyethers. Besides, its ionic conductivity was also found to be higher than the hyperbranched homopolymer PHEMO, most likely due to the motion performance difference of the triethylene glycol moieties in these two kinds of the hyperbranched polymers. As mentioned above, the ionic conducting groups were grafted as the pendant chains in the former whereas they were directly bound to the polymer backbones in the latter. Evidently, the former possesses higher motion performance than the latter.

In addition, it was noticed that the molar ratio of triethylene glycol as side chain to oxetane in PHEMO is 1 whereas this

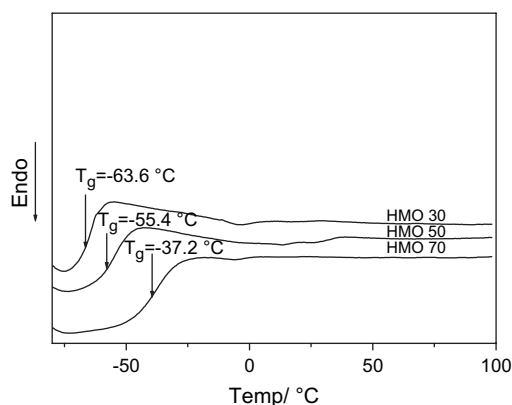


Fig. 6. DSC thermograms of the hyperbranched copolyethers.

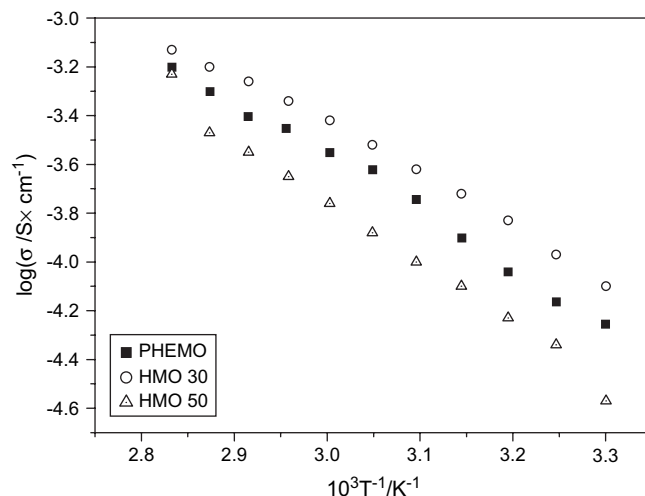


Fig. 7. Ionic conductivity plots vs. the reciprocal of temperature.

value is below 1 in the hyperbranched copolymers studied in the present work. Consequently, the ionic conductivity can be further improved by continuously increasing the MEMO content in the composition. However, an appropriate amount of HMO is also required to retain the hyperbranched structure featured by the perfectly amorphous state and lower glass transition temperature. In fact, designing and synthesizing a kind of novel oxetane-derived monomers simultaneously comprising a hydroxy group and poly(ethylene glycol) oligomers as a side chain in the same molecule remains a great challenge to further improve the ionic conductivity of the hyperbranched polyethers.

3.7. Thermal stability analysis

Fig. 8 presents the TGA analytical curves of the hyperbranched copolyether HMO 50 prior to and after doping with LiTFSI. It seemed that an addition of the lithium salt is not harmful to the thermal stability of the pure copolymers. Both curves start to lose weight at about 330 °C (10% weight loss) and ends over 400 °C. This revealed that the weight-losing

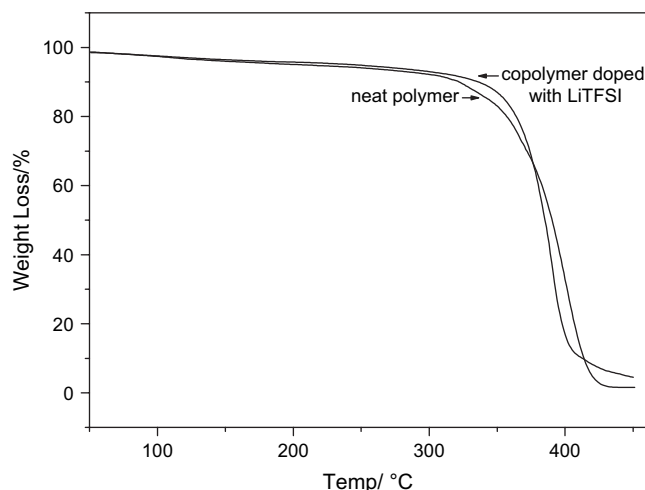


Fig. 8. TGA thermograms of the hyperbranched copolyether HMO 50 prior to and after doping with LiTFSI.

temperature of this kind of the polymer electrolytes is substantially higher than the boiling point of liquid electrolytes currently used in lithium ion battery [23]. Regarding to those liquid electrolytes, however, the weight-losing process of the resultant polymer electrolyte is gradual instead of abrupt. Thence, when used as the solid polymer electrolyte in the lithium ion battery, they certainly hold the higher thermostability over their liquid counterparts.

4. Conclusion

A kind of novel hyperbranched copolyethers was synthesized from the CROP of two oxetane-derived monomers HMO and MEMO, where HMO was used to construct the hyperbranched structure and MEMO was responsible for the ionic conductivity improvement. Their ionic conductivity was found to be higher than the hyperbranched homopolymer PHEMO. The ionic conductivity of the sample HMO 30 reached 8.0×10^{-5} S/cm at 30 °C and 7.4×10^{-4} S/cm at 80 °C, respectively. Compared with PHEMO, the substantial improvement in the ionic conductivity was attributed to the higher motion performance of the pendant triethylene glycol oligomer in the obtained hyperbranched copolyethers. The thermal decomposition temperature of above 330 °C also exhibited these polymer electrolytes having the higher thermal stability than their liquid counterparts.

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