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Melt rheology of aliphatic hyperbranched polyesters with various molecular weights

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Abstract

Rheological behaviour of a series of aliphatic hyperbranched polyesters, Boltorn polymers, with different molecular weights (generations) is studied in the molten state. The onset of a nonlinear region occurs at much lower strains for Boltorn polymers of lower generations (generation 2 and 3) compared with those of higher generations (generation 4 and 5) in the dynamic strain sweep experiments. Relaxation times calculated from dynamic rheological parameters using an empirical equation indicate that the generation 2 and 3 polymers relax much slower than their higher generation counterparts. The generation 2 and 3 polymers show shear-thinning behaviour, whilst higher generation materials are Newtonian, in both oscillatory and steady shears within the deformation rates investigated. The Cox–Merz rule is found to not be obeyed by all the polymers studied. With increasing temperature, steady shear viscosity of the Boltorn polymers decreases but the features of shear rate dependence, shear thinning or Newtonian behaviour, remain the same. Flow activation energies obtained from the temperature dependence of steady shear viscosity show that Boltorn polymers have higher activation energies than most linear polymers, indicating a stronger temperature dependence of flow. The generation 2 and 3 polymers exhibit greater flow activation energies than the generation 4 and 5 materials due to stronger polar interactions. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Rheology; Hyperbranched polymer; Activation energy

1. Introduction

Dendritic polymers, including dendrimers and hyperbranched polymers, represent an important class of polymeric materials that can be contrasted to conventional linear thermoplastic. Their molecular architecture is highly branched and thus the molecules are intrinsically globular (at sufficient molecular weight) with a high surface functionality. They are one of the most rapidly expanding fields of synthetic polymer science in recent years [1–4], despite theoretical work being reported as early as 1952 [5].

The building blocks of dendritic polymers are AB_x type monomers which contain one functional group A and x functional groups B. Note that functional groups A and B react with each other. Each layer in the dendritic structure is called a generation [4]. When a B_y core (a compound with y functional groups B) is used and the synthetic procedure is controlled with extreme care, dendrimers without unreacted functional group B inside the molecules can be produced. The requirements of highly purified chemicals and complicated polymerization steps mean that dendrimers are expensive to produce [2]. Although hyperbranched polymers can be synthesized from similar reactants as dendrimers, the production of hyperbranched polymers is less costly as they are produced in a less controlled polymerization process and thus contain linear sequences due to unreacted B groups inside the molecules.

It is the “molecular ball bearing” nature of the dendrimers which leads to their advantageous properties such as high solubility in solvents and low melt viscosity compared with the linear polymers [6]. Due to their imperfect structures, it could be expected that hyperbranched polymers would show behaviour intermediate to that of linear and dendritic polymers, yet many favourable properties, namely high solubility and low viscosity, are retained.

Although many dendritic materials have been successfully synthesized and characterized, which indicates the maturity of such materials in terms of polymer chemistry and physics [7,8], certain properties such as melt rheology have been less frequently explored. There are only limited reports even for solution rheology of dendrimers [1,8,9]. Hawker and coworkers reported melt rheology of dendrimers that are aromatic dendritic polyethers [6]. In this brief

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Table 1
Material characteristics of Boltorn hyperbranched polymers

	Hydroxyl number (mg KOH/g)	Molecular weight (g/mol)	Polydispersity (M_w/M_n)
Boltorn H20	490–520	2100	1.3
Boltorn H30	480–510	3500	1.5
Boltorn H40	470–500	5100	1.8
Boltorn H50	465–490	7500	2.5

study, the dendrimers were found to be Newtonian and obey the Cox–Merz rule that predicts equal values of complex and steady shear viscosities at same deformation rate (frequency or shear rate). The same conclusion also has been made for hyperbranched polymers [7]. Due to their low melt viscosities, dendritic polymers have been used in the preparation of polymer blends as flow modifiers to reduce the blend viscosity [10,11] and as processing aids to reduce the occurrence of extrudate irregularities [12]. The great potential of dendritic polymers to be used in pharmaceutics and biochemistry has also been suggested [2,7]

The aim of the present study is to rheologically characterize a series of commercially available aliphatic hyperbranched polyesters, Boltorn polymers, in the molten state. Both oscillatory and steady shear experiments are carried out and the applicability of Cox–Merz rule verified. Flow activation energies are evaluated from the temperature dependence of steady shear viscosities of the polymers.

2. Experimental

Hyperbranched polymers studied in this work are aliphatic polyesters using ethoxylated pentaerythritol as central cores and 2,2-bis(methylol)propionic acid (bis-MPA) as dendritic units. They are supplied by Perstorp, Sweden, under the tradename as Boltorn H series dendritic polymers. Boltorn H20 indicates that it statistically contains two generations of bis-MPA, whilst Boltorn H50 is the hyperbranched polymer with five generations of MPA. The majority of hydroxyl groups are believed to be present in the peripheral regions of the Boltorn polymers. Hydroxyl number, molecular weight and polydispersity of the Boltorn polymers from the data sheet provided by Perstorp are listed in Table 1.

Because Boltorn polymers are hygroscopic, they were dried in a vacuum oven at ambient temperature for 24 h before processing and measurements. Compression-moulded sample platens of 2 mm thickness were made using a laboratory pneumatic press using a pressure of 20 MPa for 5 min at 110°C. The formed platens were then quenched to room temperature quickly by removing the mould assembly to another press cooled by a running water jacket.

The dynamic and steady shear rheological properties of the samples were determined on a Rheometrics Dynamic

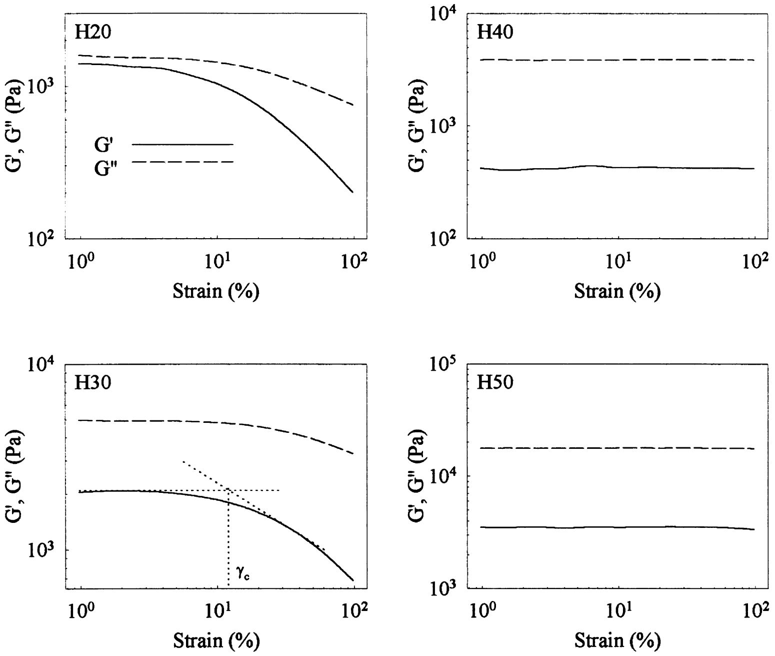

Fig. 1. Strain dependence of storage modulus and loss modulus of polymers at 100 Hz.

Analyser II (RDAII) rotational rheometer using 25 mm parallel-plate fixtures with a constant gap setting of 0.9 mm. The fixtures were mounted between the instrument actuator that applied the test strain, and the transducer that sensed the sample response. The sample was placed between the preheated fixture plates located inside of an environmental chamber purged with dry nitrogen. The temperature control was achieved by monitoring sample temperature with a thermocouple and chamber temperature with a platinum resistance thermometer (PRT). Calibration of the thermocouple and PRT was performed regularly with a thermocouple calibrator, Micormite II portable temperature laboratory. Ten minutes were allowed to removed excessive melt and to reach thermal equilibrium before the test started. Dynamic measurements were performed within the linear viscoelastic (LVE) region obtained by strain sweep tests. No preshear was applied to the sample.

3. Results and discussion

3.1. Dynamic rheological properties

An important first step in performing dynamic rheological characterization is to determine the LVE region of materials in which dynamic rheological parameters are independent of applied strains. Fig. 1 shows the strain dependence of storage (G') and loss moduli (G'') of the Boltorn materials at 110°C with a frequency of 100 Hz by strain sweep experiments. H20 and H30 have short LVE regions with critical strains (γ_c) about 10%, whilst H40 and H50 have much wider LVE regions which are clearly greater than 100% of strain. LVE regions are found to be small ($\gamma_c < 10\%$) for materials with specific microstructures such as filled polymers [13] and liquid crystalline polymers [14,15]. By contrast, dilute polymer solutions and isotropic polymers have very wide LVE regions ($\gamma_c > 100\%$) due to lack of interactions and delicate microstructures which are susceptible to large strains [16]. The different LVE regions shown by the Boltorn polymers is apparently due to their different molecular characteristics, i.e. various molecular sizes, number of hydroxyl groups and distribution of the hydroxyl groups, which lead to different microstructures.

Fig. 2 shows molecular weight dependence of G' and G'' of the Boltorn polymers at arbitrary frequencies of 1 and 10 Hz from dynamic frequency sweep within LVE range at 110°C. At 1 Hz, G' of H20 and H30 is much greater than those of H40 and H50, whilst G'' shows an increasing trend with molecular weight. Except for H20, G'' is larger than G'. The similar behaviour is also observed at 10 Hz but G'' is greater than G' for all samples. These results indicate than the loss moduli are more dominant than the storage moduli for the Boltorn polymers with higher generation as G'' is a few orders of magnitude higher than G' for H40 and H50.

Frequency dependence of the complex viscosity of the Boltorn polymers at 110°C measured by dynamic frequency sweep experiment within LVE range is depicted in Fig. 3. Although the literature regarding to melt viscosity of dendritic polymers to date suggests that they behave like Newtonian fluids, showing a constant viscosity plateau with changing deformation rate (shear rate or frequency), H20 and H30 clearly exhibit shear-thinning behaviour. By contrast, H40 and H50 are very much Newtonian within the frequency range studied. It has been reported that the lack of entanglement of the dendritic polymers leads to their Newtonian behaviour [6,17]. It is apparently not the case here for H20 and H30 where some sort of interaction (if not an entanglement effect) occurs. Since they are smaller molecules compared with H40 and H50 and thus less likely to have more entanglements (if any) than H40 and H50, the most likely cause for these different frequency dependences of complex viscosity could be the degree of polar interaction

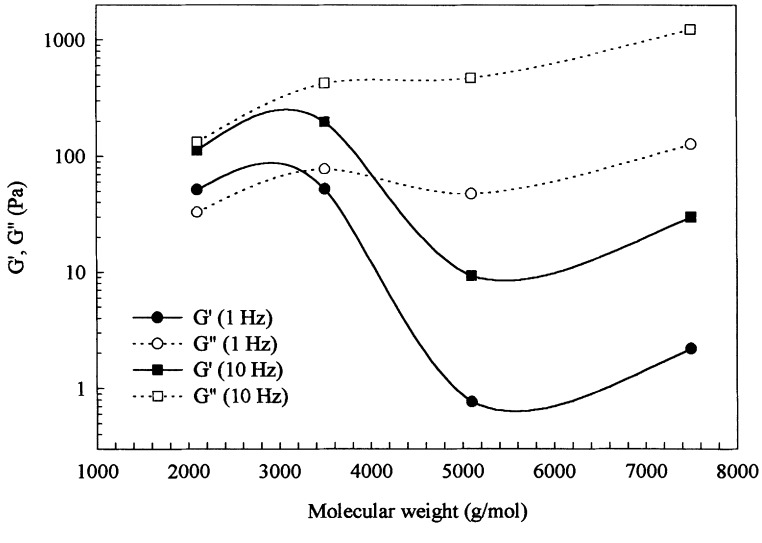

Fig. 2. Molecular weight dependence of storage moduli and loss moduli of polymers at 1 and 10 Hz.

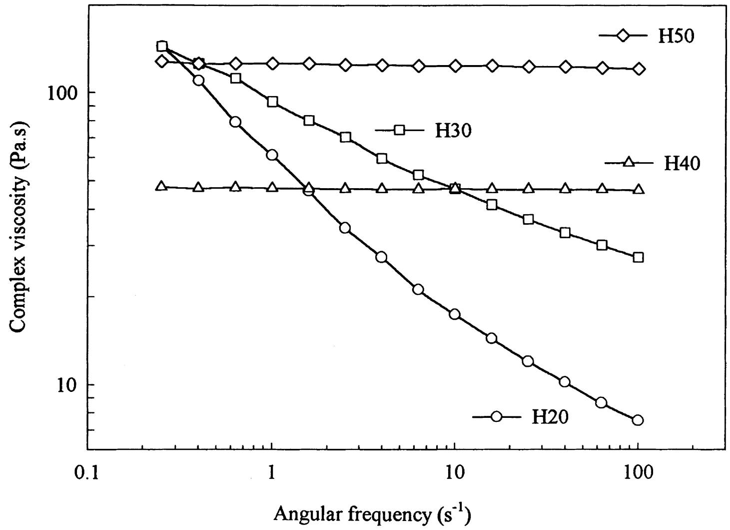

Fig. 3. Complex viscosity (η^*) of Boltorn polymers as a function of angular frequency at 110°C.

between molecules due to the numerous peripheral hydroxyl groups. Table 1 indicates that the low generation materials have the higher concentration of hydroxyl units. Furthermore, it is likely that as the generation number increases, the dendritic arms become more flexible and fold back into the hyperbranched molecules and thus less polar end groups will be exposed to other molecules. It is known that longer emanating chains from the molecular cores are more flexible and able to facilitate chain folding compared with shorter ones [7,18].

As mentioned in the introduction, melt rheology of dendritic polymers has not yet been extensively investigated and thus there is little evidence or discussion in the literature to support our contention with regards the rheology of dendritic polymers, that is, different complex viscosity behaviour results from the polar interaction of peripheral hydroxyl groups. However, solution rheology of amidoamine hyperbranched polymers was found to show classic polyelectrolyte behaviour that solution viscosities can be significantly reduced with the addition of lithium chloride [19]. Furthermore, it has been reported that the addition of ionic groups in linear polymers (ionomers) considerably alters the rheological characteristics of the polymers [20–22]. In general, viscosity of polymers can increase strikingly if the ionic interaction is incorporated and shear-thinning behaviour occurs at much lower deformation rates [20]. A thermolabile network formed due to the strong self-association of ionic groups has been proposed to explain this viscosity enhancement phenomenon [21]. The idea that H20 and H30 may have stronger intermolecular interaction (even in the melt) due to more

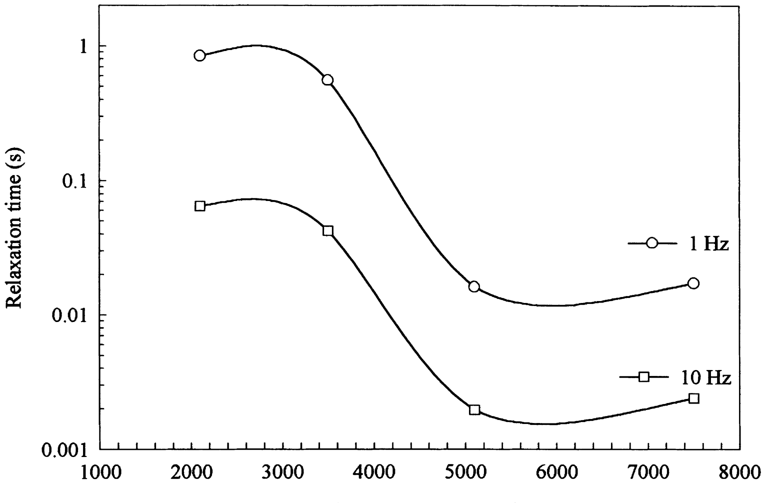

Fig. 4. Relaxation times of Boltorn polymers calculated by Eq. (1) at 1 and 10 Hz.

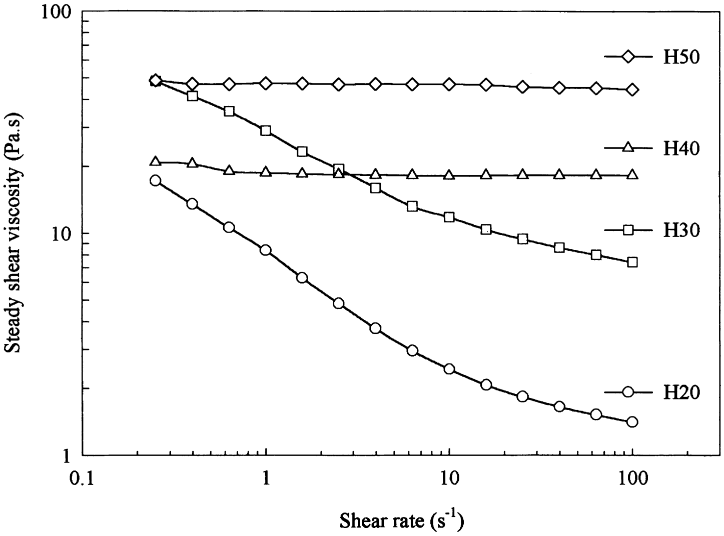

Fig. 5. Steady shear viscosity (η) of Boltorn polymers as a function of shear rate at 110°C.

peripheral hydroxyl groups compared with H40 and H50 is along these lines.

Characteristic relaxation times of polymeric materials at specific frequencies have been calculated using an empirical equation substituted by dynamic rheological parameters [23–26],

$$\lambda = G'/(|\eta^*| \times \omega^2) \tag{1}$$

where G' is the storage modulus, η^*, the complex viscosity and ω, the applied frequency. Fig. 4 shows relaxation times from Eq. (1) of Boltorn polymers at 110°C. At both frequencies, 1 and 10 Hz, relaxation times of H20 and H30 are much longer than H40 and H50. This deviates from what would be expected from linear flexible polymers where larger macromolecules have longer relaxation times due to more entanglements constraining molecular mobility [26]. A similar molecular weight dependence of relaxation times as for linear polymers has been observed for arborescent graft polystyrenes that have many long branched chains and contain no polar moieties [27]. Since the entanglement effect in dendritic materials is believed to be less dominant compared with conventional linear flexible polymers [6], the most likely cause of this behaviour may also be the polar interaction between molecules due to their different molecular sizes and different hydroxyl numbers. H20 and H30 are thought to have more extended molecular conformation as far as their radiating chains are concerned, whilst the longer chains of H40 and H50 are more flexible [18]. Rigid macromolecules such as LCPs have been reported to have relaxation times of the same order as H20 and H30 at same frequency [23–25]. By contrast, flexible linear polymers show relaxation times a few orders smaller than LCPs, which resembles the behaviour of H40 and H50 [24,25].

3.2. Steady shear viscosity

Steady shear viscosity of the Boltorn polymers as a function of shear rate at 110°C is shown in Fig. 5. As also found in their oscillatory shear analogues, H20 and H30 are shear thinning, while H40 and H50 are Newtonian. H20 is less viscous than its higher generation counterparts within the shear rate range studied. H30 has almost the same viscosity as H50 at 0.1 s^{-1} and becomes less viscous than both H40 and H50 once the shear rate exceeds 2.5 s^{-1}.

Another commonly reported rheological feature of dendritic polymers is the applicability (or otherwise) of the Cox–Merz rule which predicts that complex viscosity, $\eta^*(\omega)$, and steady shear viscosity, $\eta(\dot{\gamma})$, are the same value at the same deformation rate.

$$|\eta^*(\omega)| = \eta(\dot{\gamma}) \qquad \text{when } \omega = \dot{\gamma} \tag{2}$$

If the rule holds, it is of much use to industrial rheologists to obtain steady shear viscosity from oscillatory experiments as the former directly relates to polymer processing and the latter is easier to perform. It has been reported that the Cox–Merz rule is obeyed by dendrimers [6,28] and hyperbranched polymers [7] in the molten state, although no graphs of comparison of η and η^* of dendritic polymers have been given to date. Fig. 6 gives the comparison of η and η^* of Boltorn polymers at 110°C (from Figs. 3 and 5). η is all smaller than η^* for all Boltorn polymers and thus the Cox–Merz rule does not hold for the Boltorn polymers.

In general, isotropic linear polymers have been found to follow the rule [29–32]. However, rheology of materials susceptible to changes in microstructures such as suspensions [33] and LCPs [32,34,35] may not obey the rule. In addition to neat polymers, immiscible polymer blends often show deviations from the rule due to morphological effects,

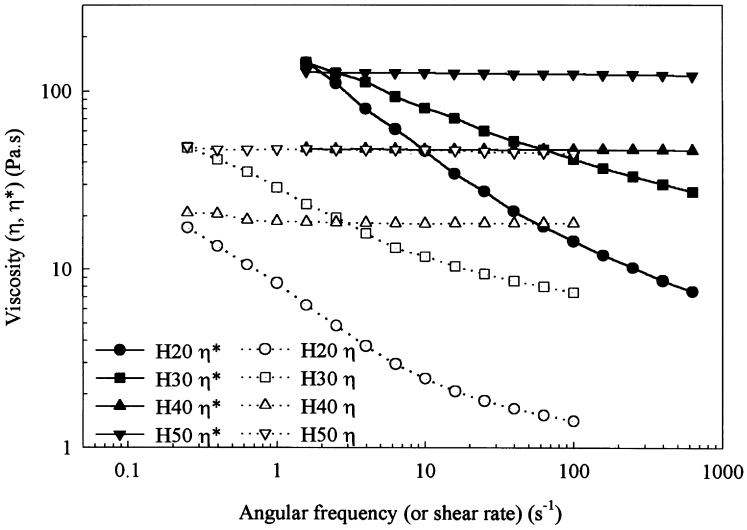

Fig. 6. Comparison of η and η^* of polymers at 110°C.

even though the constituent polymers of the blends do obey the rule [36]. Boltorn polymer melts are likely to have certain microstructures that respond differently upon the application of steady and oscillatory shears. Note that oscillatory shear may be able to reveal the shear-thinning potential of materials but it does not alter the structure of the materials if applied within the LVE region. By contrast, steady shear can effectively rearrange molecular packing of materials along the shear direction. Lin and Winter [37] reported that η of a LCP is much lower than its η^* due to the better molecular alignment achieved by steady shear.

3.3. Flow activation energy

Fig. 7 depicts that steady shear viscosity of H20 at various temperatures (80–110°C). H20 shows shear-thinning behaviour at all temperatures with shear thinning being most prominent at 80°C. This demonstrates that the shear thinning shown by H20 is a general feature rather than due to the specific temperature (110°C) used in Fig. 5. At any given shear rate, steady shear viscosity of H20 monotonically decreases with temperature. Note that H30 also shows similar behaviour as H20 and its graph is not shown to avoid repetition.

Fig. 8 depicts the steady shear viscosity of H50 at various temperatures (100–130°C). H50 exhibits Newtonian behaviour at all temperatures and its steady shear viscosity monotonically decreases with temperature. H40 also behaves in a similar fashion as H50.

As steady shear viscosity of the Boltorn polymers all decrease with temperature, it suggests that the temperature dependence of the steady shear viscosity may

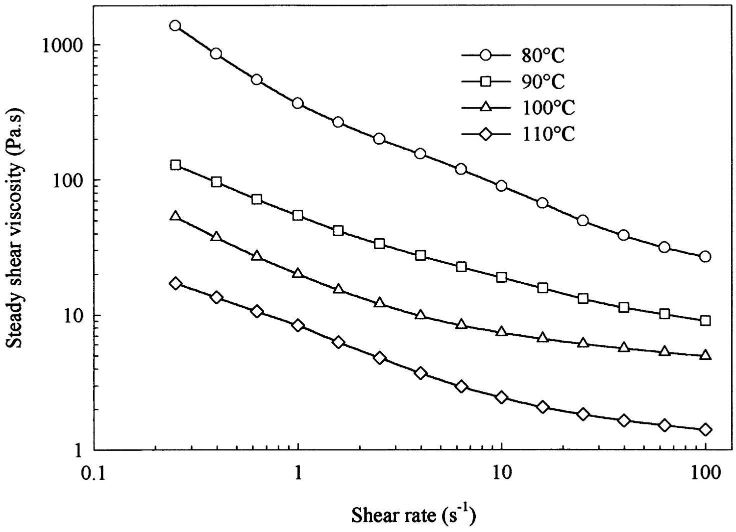

Fig. 7. Steady shear viscosity of Boltorn H20 at various temperatures.

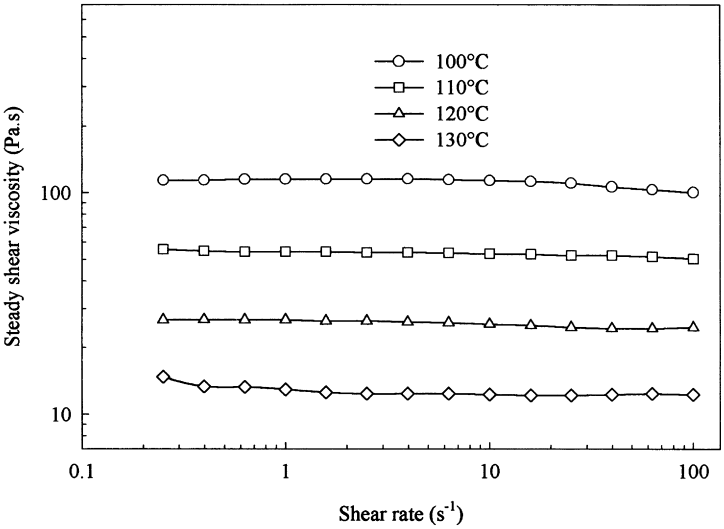

Fig. 8. Steady shear viscosity of Boltorn H50 at various temperatures.

follow an Arrhenius type equation [38],

$$\eta = A \times e^{E_a/RT}$$

where A is a constant, R, the gas constant, E_a, the activation energy and T, the temperature.

Fig. 9 shows the plot of ln η versus $1/T$ of the Boltorn polymers at an arbitrary shear rate of 1 s^{-1}. H20 and H30 do not show a satisfactory linear relationship between ln η and $1/T$, whilst H40 and H50 exhibit good linear fits. This indicates that the stronger polar interaction present in H20 and H30 molecules complicates the temperature dependence of their steady shear viscosity. Unlike H40 and H50 whose behaviour is less affected by this consideration, the increase in chain mobility of H20 and H30 may be further promoted due to the weakening of hydrogen bonding.

It is expected that greater shear rates may help to reduce the polar influence on the temperature dependence of steady shear viscosity, as molecules have to undergo greater scale of realignment to cope with the more vigorous deformation applied. Fig. 10 shows the plot of ln η versus $1/T$ of the Boltorn polymers at an arbitrary shear rate of 10 s^{-1}. Although H20 and H30 still exhibit inferior linear fits compared with H40 and H50, their linear relationship between ln η and $1/T$ improves compared with that at lower shear rate (1 s^{-1}).

Table 2 gives the correlation factors and flow activation energies of the Boltorn polymers evaluated from the linear fits of Figs. 9 and 10. A value of unity for the correlation factors indicates a perfect linear fit and a smaller value means less correlation within the data. The improvement of the linear fit can be achieved by increasing molecular

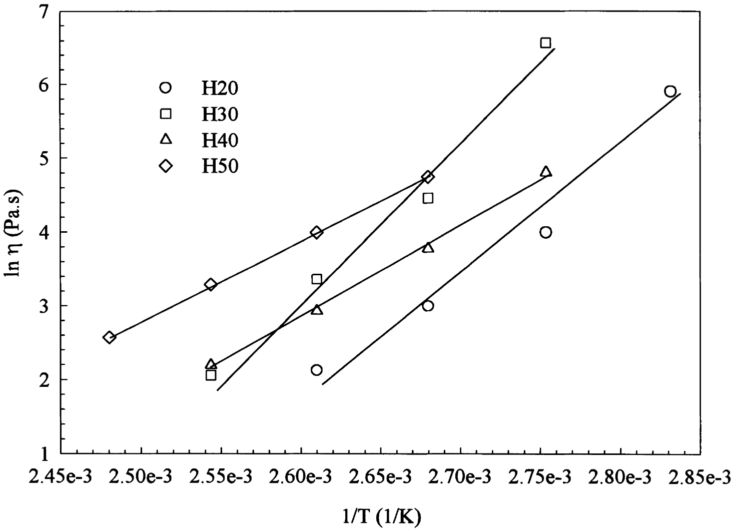

Fig. 9. Arrhenius plot of steady shear viscosity of Boltorn polymers at 1 s^{-1}.

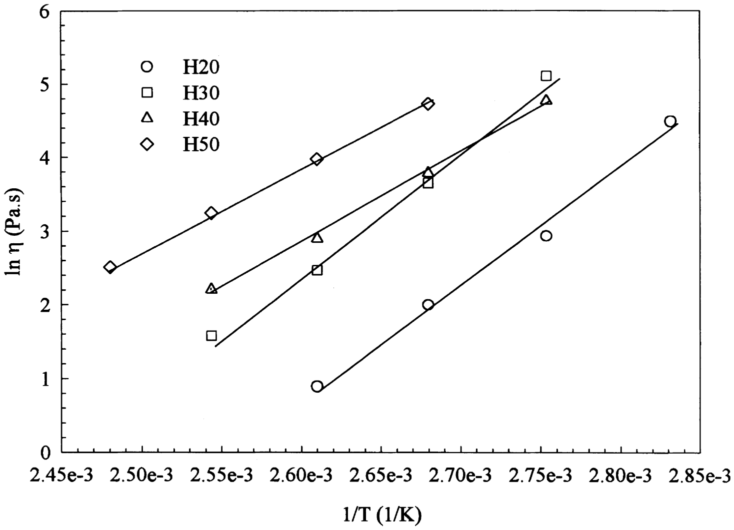

Fig. 10. Arrhenius plot of steady shear viscosity of Boltorn polymers at 10 s^{-1}.

weight at a given shear rate or higher shear rates for a specific polymer. At 10 s^{-1}, H20 and H30 have similar flow activation energies, 132 and 140 kJ/mol, respectively, which are higher than H40 and H50. The flow activation energies of the Boltorn polymer appear to be higher than most of the linear polymers whose flow activation energies are lower than 70 kJ/mol in general [39–43]. Higher flow activation energies (>100 kJ/mol) have been found for poly-4-methyl-1-pentene and poly-α-methylstyrene [39]. Since there is no such data available for dendritic polymers to date, discussion is made possible by comparing with branched polymer [42,43] and ionomers [21,44]. Both long and short chain branches in polyethylene have been known to lead to higher flow activation energies [42,43]. The incorporation of ionic groups into polymers not only increases the melt viscosity and encourages the occurrence of shear thinning as stated previously, but also results in greater flow activation energies [21,44,45]. The latter has been attributed to the presence of ionic domains in the melts enhancing flow resistance, and a greater activation energy [44]. A linear relationship between flow activation energy and content of ionic group has been found in sulfonated PET [45], sulfonated poly(1,4-butylene isophthalate) and [21] sulfonated styrene-butadiene rubber [44]. Therefore, the much higher activation energies of H20 and H30 is likely to result from their stronger polar interaction compared with H40 and H50, although the strength of hydrogen bonding is much weaker than ionic attraction.

4. Conclusions

Melt rheology of a series of aliphatic hyperbranched polymers, Boltorn polymers, with different molecular weight (generations) is investigated. Boltorn polymers of lower generations (H20 and H30) exhibit much shorter LVE regions compared with Boltorn polymers of higher generations (H40 and H50). Short LVE regions are commonly observed for materials possessing specific microstructures, which are easily altered by deformation, such as liquid crystalline polymers and suspensions. Storage moduli of H20 and H30 are very close to their loss moduli, whilst loss moduli of H40 and H50 are much greater than their storage moduli. H20 and H30 show shear-thinning behaviour in both oscillatory and steady shear. This is opposite to what has been reported that dendritic polymers are Newtonian fluids. By contrast, H40 and H50 are Newtonian in both deformation conditions. The Cox–Merz rule is also found not to hold for all the Boltorn polymers studied, despite limited literature available suggesting otherwise. Flow activation energies of Boltorn polymers are higher than that of most of linear polymers. H20 and H30 have flow activation energies and relaxation times much greater

Table 2
Flow activation energies of the Boltorn polymers

	E_a (kJ/mol) at 1 s^{-1}	Correlation factor χ^2	E$_a$ (kJ/mol) at 10 s^{-1}	Correlation factor χ^2
H20	139	0.971	132	0.992
H30	174	0.984	140	0.993
H40	103	0.997	102	0.997
H50	91	0.999	92	0.999

than H40 and H50, which may also result from the stronger polar interactions present in H20 and H30.

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