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Elastic behavior of flexible polyether(urethane–urea) foam materials

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Abstract

Polyether(urethane–urea) foams (PEUU) with varying urea contents and different polyether segments (PPO and PPO-*co*-PEO (93/7 w/w)) were compacted to transparent solid plaques via compression molding. The thermal, mechanical and elastic properties of the compacted PEUU materials were studied. With increasing urea content, the shear modulus was increased, while the glass transition temperature (T_g) remained low and unaffected. The T_g's of PEEU's with PPO segments were, however, lower than with PPO-*co*-PEO segments, indicating more mixing of urea segments with PPO-*co*-PEO. The flow temperatures of both PEUU's were high (~300 °C) for all compositions. The compression sets, tensile sets and hysteresis energy of the PEUU's were low and increased with urea content. The use of PPO-*co*-PEO segments resulted in PEEU's with higher compression sets and tensile sets and also more hysteresis. The recovery of the PEUU's showed two relaxation regimes: a fast (elastic) recovery and a slower (viscoelastic) recovery. The recovery of these PEUU's is almost complete giving time.

Keywords: Poly(propylene oxide); Viscoelastic; Recovery

1. Introduction

Water-blown slabstock flexible polyether(urethane–urea) (PEUU) foams are extensively used in applications such as cushioning and mattresses [1]. These segmented PEUU's consist of alternating sequences of mobile polyether, mostly poly(propylene oxide) (PPO), and rigid urea segments linked by urethane groups. Due to thermodynamic incompatibility between the polyether and urea segments phase separation occurs through liquid–liquid demixing [2], often in combination with crystallization of the urea segments [3]. The PEUU foams also contain a high amount of chemical crosslinks as most of the polyether segments in the foam formulation has a functionality higher than 2.

Phase separation results in a morphology of urea hard domains dispersed in a soft polyether rich matrix. Several authors reported that the PEUU copolymers contained a spherulitic and fibrous crystalline urea phase and spherical urea globules, both dispersed in a soft polyether matrix as demonstrated by AFM [4,5]. In other literature, a complex morphology was suggested [6–8], consisting of a soft matrix rich in polyether, with a dispersed crystalline urea phase and amorphous urea aggregates. The hard domains act as physical crosslinks and provide together with the chemical crosslinks, the elastic behavior of the PEUU's. Furthermore, the hard domains function as fillers reinforcing the blockcopolymer.

For these (foam) materials the elastic properties are important and depend both on the elastic behavior of the solid material as well as the structure of the foam. The properties of the solid material, thus excluding the foam structure, can be studied by compacting the foams into solid plaques [7]. Armistead et al. [7] studied compacted PEUU materials with increasing urea content and found a high degree of phase separation in these materials. These authors reported that increasing the urea content of these materials resulted in higher moduli. Abouzhar et al. [9] found for similar PEUU's with increasing the urea contents also higher moduli, but simultaneously poorer elastic properties (higher hysteresis and higher tensile sets).

In our study two types of PEUU foam materials have been investigated. One series is based on PPO, while the other is based on a random copolymer of PPO with 7 wt% ethylene oxide (PPO-*co*-PEO). The 'elastic' properties

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(compression set, tensile set and hysteresis) on compacted PEUU foams were studied as a function of the urea content. Particular attention is given to the (visco)elastic behavior of these materials.

2. Experimental

2.1. Materials

Samples of conventional slabstock flexible polyether (urethane–urea) foams (PEUU) were a gift from Shell. In one series poly(propylene oxide) (PPO, $M_n \approx 3000$ g/mol) was used and the other contained a random copolymer of PPO with 7 wt% ethylene oxide (PPO-*co*-PEO). Both series contained a mixture of 80 mol% tri-functional PPO and 20 mol% mono-functional PPO (the functionality of the polyether segments is 2.6 [10]). The rigid urea segment content was varied from 25.8 to 34.6 wt% by changing the toluene diisocyanate (TDI) to water ratio in the feed. A 80:20 molar mixture of 2,4-TDI:2,6-TDI was used. The foam samples were dried for 24 h at 70 °C in a vacuum oven before processing.

2.2. Compression molding

Foam samples ($120 \times 120 \times 60$ mm^3) were compacted by compression molding in two steps [7] on a Lauffer OPS 40 press At first the samples were compacted using a pressure of 100 bar at 150 °C for 3 min, after which the samples were allowed to cool whilst maintaining the pressure. The compacted samples were subsequently cut to fit in the mold. Thereafter, compression molding of these performed samples was continued at 200 °C for 3 min at 100 bar. The plaques ($120 \times 120 \times 2$ mm^3) were homogeneous and transparent and contained no air bubbles. Test specimens were cut from the plaques to the desired shape and subsequently dried at 70 °C in vacuum for 24 h before testing

2.3. Dynamical mechanical thermal analysis (DMTA)

Compression molded samples ($70 \times 9 \times 2$ mm^3) were used for dynamical mechanical thermal analysis. The storage modulus (G') and loss modulus (G'') as a function of temperature were measured using a Myrenne ATM3 torsion pendulum at a frequency of 1 Hz and 0.1% strain. The samples were first cooled to −100 °C and then subsequently heated at a rate of 1 °C/min. The glass transition temperature is taken as the maximum of the loss modulus. The flow or softening temperature (T_{flow}) is defined as the temperature, where the storage modulus reaches 1.0 MPa. The flex temperature (T_{flex}) is defined as the temperature at the start of the rubber plateau region. The decrease in storage modulus of the plateau region with increasing temperature is quantified by $\Delta G'$, which is calculated from:

$$\Delta G' = \frac{G'_{(T_g+50\ ^\circ\mathrm{C})} - G'_{(T_{\text{flow}}-50\ ^\circ\mathrm{C})}}{G'_{(25\ ^\circ\mathrm{C})}} \frac{1}{\Delta T} \quad \text{in } [^\circ\mathrm{C}^{-1}] \tag{1}$$

ΔT is described as the temperature range: $(T_{\text{flow}} - 50\ ^\circ\mathrm{C}) - (T_g + 50\ ^\circ\mathrm{C})$.

2.4. Compression set

The samples for compression set measurements were cut from the compacted plaques The compression set is measured according to the ASTM 395 B standard. After 24 h compression of 25% at room temperature the stress was released and half an hour later the thickness of the samples was measured. The compression set is defined as:

$$\mathrm{CS} = \frac{d_0 - d_2}{d_0 - d_1} \times 100\% \tag{2}$$

with:

d_0, thickness before compression (mm); d_1, compressed thickness (mm); d_2, thickness 30 min after release of compression (mm)

The compression sets of some compacted PEUU samples were also determined 24 h after compression at 70 °C (also ASTM 395B) and during relaxation at room temperature. The compression set was taken as the average of three measurements.

2.5. Tensile set and hysteresis

Cyclic stress–strain experiments were conducted on samples from compacted plaques, which were cut to dumbbells (ISO 37 s3). A Zwick Z020 universal tensile machine equipped with 500 N load cell was used to measure the stress as a function of strain for each loading and unloading cycle at a strain rate of $3.33 \times 10^{-2}\ \mathrm{s}^{-1}$ (test speed of 50 mm/min). The strain of each loading–unloading cycle was increased (stair-case loading) and the tensile set was determined as function of the applied strain. The tensile set was calculated from the following relation:

$$\text{Tensile set} = \frac{\Delta\varepsilon_{\text{remaining}}}{\Delta\varepsilon_{\text{cycle}}} = \frac{\varepsilon_{r,\text{cycle}(i)} - \varepsilon_{r,\text{cycle}(i-1)}}{\Delta\varepsilon_{\text{cycle}}} \times 100\% \tag{3}$$

With $\varepsilon_{r,\text{cycle}(i)}$ the remaining strain at the end of cycle i and with $\varepsilon_{r,\text{cycle}(i-1)}$ the remaining strain at the end of the preceding cycle $i-1$. Of each loading–unloading cycle the relative hysteresis was determined as the area under the loading cycle (W_{loading}) minus the area under the unloading cycle ($W_{\text{unloading}}$), and divided by the area under the loading cycle as described by Eq. (4):

$$\text{Hysteresis} = \frac{W_{\text{loading}} - W_{\text{unloading}}}{W_{\text{loading}}} \times 100\% \tag{4}$$

3. Results and discussion

3.1. Introduction

Samples of water-blown flexible polyether(urethane–urea) foams (PEUU) were compacted by compression molding to homogeneous transparent solid plaques, as described previously [7]. The samples differ in urea content (varying form 25.8–34.6 wt%) but also in the type of polyether segment. The urea segment is made from a 80:20 molar mixture of 2,4-TDI:2,6-TDI and water. Physical, (visco)elastic and mechanical properties of these compacted PEUU were studied. The structure of these blockcopolymers is presented in Fig. 1, in which *x* indicates the urea segment length and *n* is the degree of polymerization.

One series contains poly(propylene oxide) (PPO, $M_n \approx 3000$ g/mol) as the polyether segment and the other a new polyether, which is a random copolymer of PPO with 7 wt% ethylene oxide (PPO-*co*-PEO, $M_n \approx 3000$ g/mol). In both series, the polyether consists of a mixture of 80:20 triol:monol and has an average functionality of 2.6 [10]. These segmented blockcopolymers contain therefore a large amount of chemical crosslinks. The composition and properties of the compacted PEUU materials are given in Table 1.

3.2. Dynamical mechanical thermal analysis

The storage and loss moduli of compacted PEUU samples, as function of temperature, with increasing urea content are presented in Figs. 2 and 3. The maximum in the loss modulus is the alpha transition. For the blockcopolymers studied this transition is also known as the glass transition. All blockcopolymers have a low glass transition temperature (T_g), a high flow temperature (T_{flow}) and a temperature independent modulus in the plateau region as indicated by the relatively low values of $\Delta G'$ (Table 1). The modulus of the rubbery plateau increases with increasing urea content of the polymers. In a previous study, Armistead et al. [7] also found temperature independent rubber moduli for compacted PEUU materials and increasing moduli with increasing the urea content.

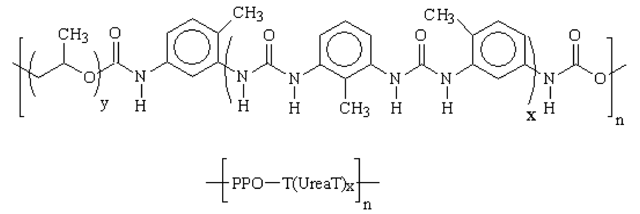

Fig. 1. Structure of TDI (2,4-TDI:2,6-TDI, 80:20 mol/mol) and water based PEUU's with urea segment length *x* and PPO segments. (The structure only indicates the linear parts).

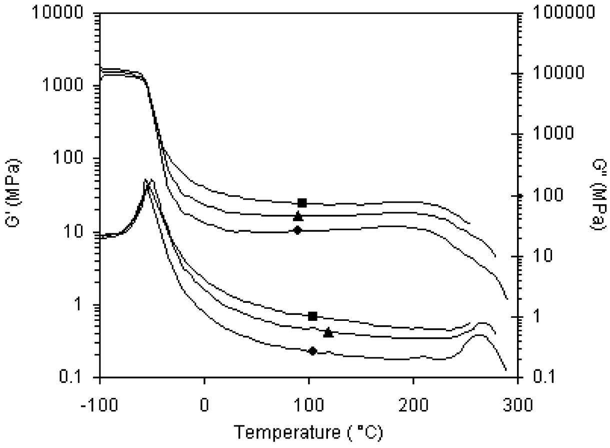

Fig. 2. Storage modulus (G') and loss modulus (G'') of compacted PEUU's with PPO as function of temperature. The urea contents are: (●) 25.8 wt%; (▲) 30.4 wt%; (■) 34.6 wt%.

The low temperature glass transitions of all PEUU's are sharp and shift to higher temperatures when PPO-*co*-PEO instead of PPO is used (Fig. 4). The flex temperatures of the PEUU's are low and lie about 20 °C above the T_g. This indicates a good low-temperature flexibility of these PEUU's and is due to the use of amorphous PPO as the polyether segment. A second maximum in the loss modulus (G'') is observed at 270 °C for all PEUU materials and may be assigned to an amorphous phase that contains the urea segments. In earlier studies, it was found that these PEUU's are multiphase systems containing amorphous and crystalline hard domains in a soft amorphous matrix [11,12]. The second maximum in the loss modulus may also be explained by changes in the dimensions of the test sample, possibly caused by degradation of the sample. The modulus, as measured by DMTA, depends strongly on the dimensions of the test sample.

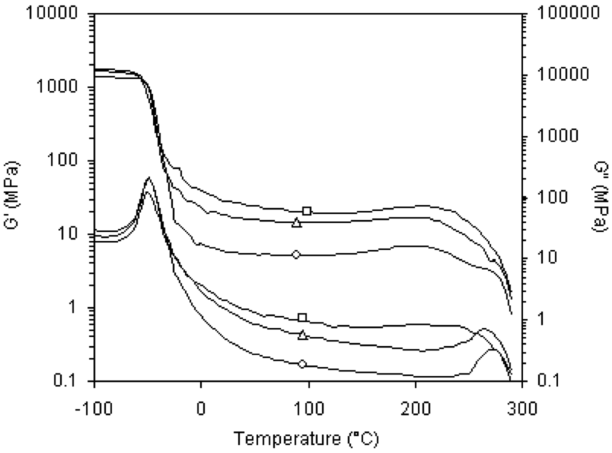

Fig. 3. Storage modulus (G') and loss modulus (G'') of compacted PEUU's with PPO-*co*-PEO as function of temperature: The urea content are: (○), 25.8 wt%; (△), 30.4 wt%; (□), 34.6 wt%.

Table 1
Properties of compacted TDI-based PEUU series (2,4-:2,6-TDI, 80:20 mol/mol) and with different polyether segments ($M_n = 3000$ g/mol): PPO and PPO-*co*-PEO (7 wt% ethylene oxide)

Urea segment length (x)[a]:	PUU[b] (wt%)	$T_{g,soft}$ (°C)	T_{flex} (°C)	T_{flow} (°C)	G' (25 °C) (MPa)	$\Delta G' \times 10^{-3}$ (°C^{-1})	CS[c] (20 °C) (%)	CS[d] (70 °C) (%)	TS[e] (at 50%) (%)	Hys.[e] (at 50%) (%)
PPO										
2.8	25.8	−51	−30	~300	10.5	2.8	2.2	10	16	32
3.5	29.2	−52	−30	~300	14.5	2.6	4.7	14	18	38
3.7	30.4	−53	−30	~300	18.5	2.0	7.3	17	17	40
4.0	31.5	−52	−30	~300	22.4	2.7	3.6	10	20	41
4.8	34.6	−50	−30	~300	31.2	3.0	8.1	15	22	49
PPO-*co*-PEO										
2.8	25.8	−47	−21	290	6.00	2.0	9.3	–	25	33
3.5	29.2	−46	−25	290	14.8	2.4	7.4	–	25	40
3.7	30.4	−47	−25	~290	17.5	2.8	7.2	–	27	41
3.7	30.4	−46	−29	~290	17.9	2.7	8.9	–	–	–
4.0	31.5	−46	−25	~300	31.3	3.0	8.6	–	29	43
4.8	34.6	−49	−26	~300	28.4	2.9	12	–	30	49

[a] Calculated from weight content and polyether molecular weight ($M_n = 3000$ g/mol).
[b] Combined urethane–urea segment content [10].
[c] CS, compression set measured at room temperature (20 °C).
[d] CS, compression set measured at 70 °C.
[e] TS, tensile set; Hys., hysteresis (both measured at 50% strain).

The flow temperatures (T_{flow}) of the PEUU's are high and well above the thermal decomposition temperature. PEUU materials are thermally instable at temperatures above 200 °C. However, although the samples were dark and brittle after being heated to 300 °C, it is expected that the DMTA values are still of use as the deformation strain in this experiment is small (0.1%). The effect of molecular weight on polymer properties becomes significant when the material is subjected to higher strains than 0.1%.

The glass transition temperature of the soft phase is sensitive towards the degree of intersegmental mixing of urea segments with polyether segments. Less mixing, i.e. a higher degree of phase separation, leads to a lower T_g of the soft phase as described by the relation of Fox [13]. For the compositions studied the molecular weight between crosslinks (M_c) of both PEEU's is the same and is in these series constant. The number of crosslinks in the blockcopolymers (crosslink density) is therefore the same for each composition.

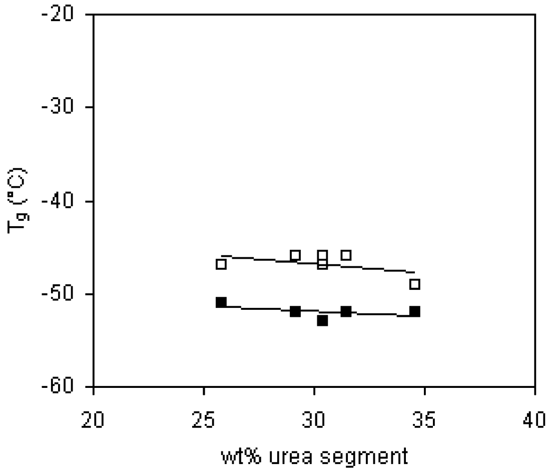

Fig. 4. Glass transition temperature as function of urea content (incl. urethane groups): (■) PPO; (□) PPO-*co*-PEO (7 wt% EO).

In Fig. 4, the glass transition is presented as function of the urea segment content of the two PEUU's with different polyether phases. The T_g of the soft phase of the PEUU's remains unaffected with increasing urea content. This may indicate no change in the soft phase composition. With the new PPO-*co*-PEO segments higher T_g's are observed as compared to the T_g's of the blockcopolymers with PPO segments. Poly(ethylene oxide) (PEO) has a slightly higher T_g than PPO and it is therefore expected that the T_g of the PPO-*co*-PEO phase will be somewhat higher than the T_g of PPO [13]. However, this latter effect is probably too small to explain the differences shown in Fig. 4. The higher affinity of the urea segments towards the PPO-*co*-PEO segments is therefore expected to cause more intersegmental mixing of the segments, which result in the higher T_g's of the PEEU's containing these polyether segments.

The shear modulus (G' (25 °C)) of PEUU's with different polyether segments increases with increasing urea content (semi-logarithmic plot, Fig. 5). Similar results were obtained for PEUU copolymers containing either the PPO or the PPO-*co*-PEO segments, which may suggest that the morphologies of both PEUU materials are comparable.

The increase in modulus of the PEUU's with increasing urea contents is higher than can be expected for spherical particle filled polymers, commonly predicted by the Guth–Smallwood relation [14,15]. The moduli are, however, lower than for fiber-reinforced systems, modeled by Cox

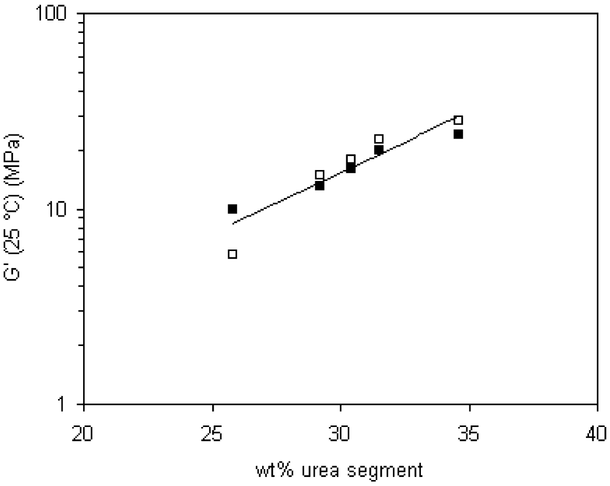

Fig. 5. Influence of hard segment concentration on shear modulus (G' at 25 °C) of compacted PEUU having different PPO segments: (■) PPO; (□) PPO-*co*-PEO (7 wt% EO).

[16] or Halpin–Tsai [17,18]. In the calculations used to model the experimental modulus of the spherical particle filled polymers to the Guth–Smallwood and the modulus of fiber-reinforced polymers to the Halpin–Tsai relation, the authors used a modulus of the soft phase of 0.4 MPa, modulus of the hard phase of 5000 MPa. A fiber aspect ratio of 1000 was used in the case fiber structures are present as reinforcements. Furthermore it was assumed that the fibers are oriented randomly. The urea content was taken as the filler content.

The aforementioned trends in modulus-composition behavior (Fig. 5) may suggest that the semi-crystalline PEUU copolymers contain both spherical and fiber-like hard domains. As demonstrated by AFM before, PEUU copolymers were found to contain urea globules and a crystalline lamellar phase [4,5].

3.3. Compression set

The compression set is a measure of the elastic behavior of the material. Low compression sets indicate a high elasticity. The compression set at room temperature, as function of the shear modulus of polyether(urethane–urea)s, is presented in Fig. 6 and Table 1.

Non-elastic deformation is mainly due to deformation of the hard phase. PEUU's with higher moduli and thus higher urea content have higher compression sets. This is probably due to more deformation of the hard phase, of which more is present. The compression set of PEUU's with PPO-*co*-PEO is also higher than with PPO segments. The incorporated 7 wt% PEO in the soft phase (PPO-*co*-PEO) resulted in higher amounts of dissolved urea segments as compared to the amount of urea segments dissolved in PPO. This may cause a slower recovery and may explain the higher compression sets, observed after 30 min relaxation.

The compression sets of PEUU with PPO segments were

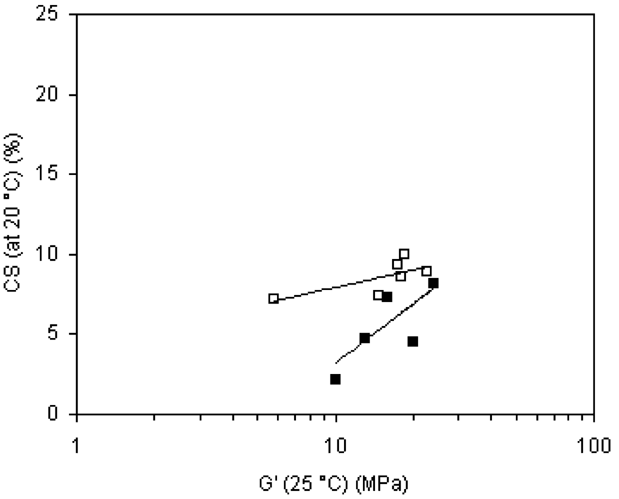

Fig. 6. Compression set at room temperature as function of the shear modulus (G' at 25 °C) of both PEUU series: (■) PPO; (□) PPO-*co*-PEO (7 wt% EO).

also measured at 70 °C (Table 1) and also increased with modulus. The CS-values were higher than the compression sets measured at 20 °C.

The compression set, according to the ASTM standard, is measured 30 min after releasing the compressive strain. If the compression set is measured as function of time, information can be obtained about possible (visco)elastic processes. Samples were compressed to 25% of the original thickness and the compression set was therefore measured as function of the relaxation time (Fig. 7). The studied PEUU's contain 30.4 wt% urea segments and have a shear modulus of approximately 18 MPa.

The compression set of both PEUU's decreases linearly with the logarithm of time. The first 80% of the compressive strain is lost in less than 10 s and is associated with relaxation of the soft polyether phase (elastic recovery). PEUU's with a PPO-*co*-PEO soft phase recover less during

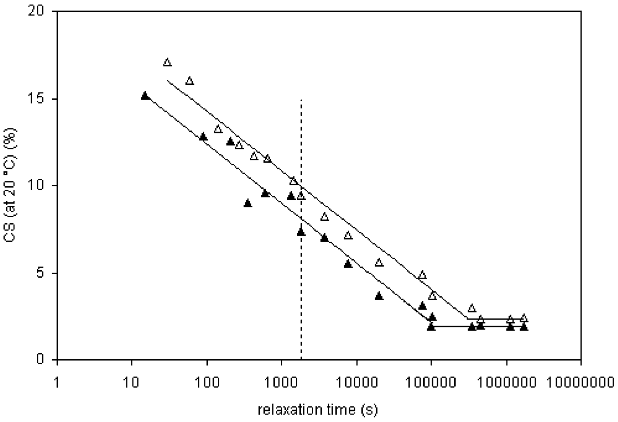

Fig. 7. The compression set as function of relaxation time (recovery) of compacted PEUU polymers having 30.4 wt% urea segments (G' (25 °C) = 18 MPa) and different polyether segments. (▲) PPO; (△) PPO-*co*-PEO. (Dashed line indicates the CS after 30 min according to ASTM 395 B standard).

the first 10 s than PEUU's with PPO segments, which may be due to the higher fraction of rigid segments dissolved in the PPO-*co*-PEO phase; limiting the recovery of the polyether phase.

From Fig. 8, a slow recovery of the PEEU materials can be observed in the interval of $10-1\times10^5$ s. This recovery may be ascribed to viscoelastic relaxation of the deformed urea phase. The rate of this slower recovery is similar for both PEUU samples. Given time, the samples recover almost completely (to 2–3%). Also, the set observed in the compression set experiments is largely viscoelastic.

3.4. Tensile set and hysteresis

Cyclic tensile tests are useful to describe the deformation of polymers as function of strain The compacted PEUU copolymers were subjected to a stair-case loading and the tensile set and hysteresis as function of strain were determined (Fig. 8a and b).

The tensile set of the PEUU's increases with increasing strain and levels off at higher strains. Higher tensile sets were observed for PEUU's with higher urea contents and may be explained by the fact that at higher urea content, more of the deformable urea phase undergoes (plastic) deformation. Up to 25% strain, similar tensile sets were found for both PEUU series. However, at higher strains (>25%) the PEUU's with PPO-*co*-PEO segments exhibit relatively higher tensile sets than PEUU's with PPO.

The hysteresis for each cycle was plotted as function of the applied strain for PEUU's with PPO or PPO-*co*-PEO as the polyether phase (Fig. 9a and b, respectively). A low hysteresis indicates little energy loss and the corresponding materials are low damping. Above 10% strain the percentage hysteresis remains constant with strain. A similar behavior was found before in hysteresis studies of thermoplastic polyether(urethane) elastomers [19]. However, for these polymers a continuous strong increase in hysteresis up to strains of 300% or higher was reported before the hysteresis leveled off with higher strains. The hysteresis of the studied PEUU materials increases with increasing urea contents and can also be explained by the higher fraction of deformable urea phase in these materials (Fig. 9a and b).

A polyether phase containing PPO-*co*-PEO segments probably has more interaction with the urea segments as

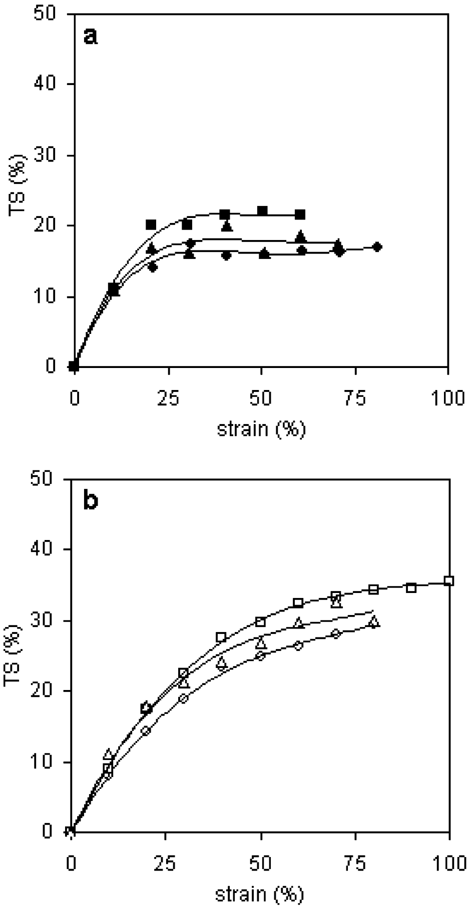

Fig. 8. Tensile set as function of the applied strain during cyclic tensile testing of PEUU series with varying urea content, (a) PPO: (●) 25.8 wt%; (▲) 30.4 wt%; (■) 34.6 wt%. (b) PPO-*co*-PEO: (○) 25.8 wt%; (△) 30.4 wt%; (□) 34.6 wt%.

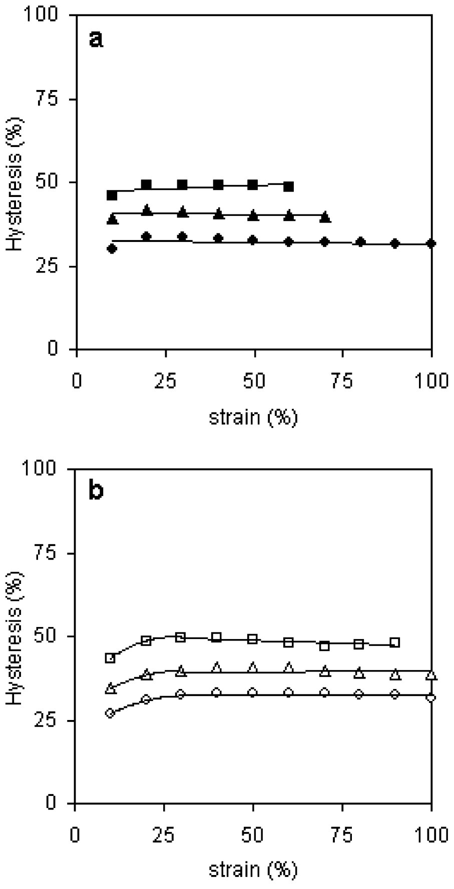

Fig. 9. Percentage of hysteresis energy as function of the applied strain during cyclic tensile tests of compacted PEUU's having different polyether phase and varying urea content. (a) PPO: (●) 25.8 wt%; (▲) 30.4 wt%; (■) 34.6 wt%. (b) PPO-*co*-PEO: (○) 25.8 wt%; (△) 30.4 wt%; (□) 34.6 wt%.

compared PPO segments, which is suggested from the T_g data of these materials (Fig. 4). This may result in relatively limited segmental mobility. The recovery of PEUU copolymers with PPO-*co*-PEO segments may therefore be slower as compared to the recovery of PEUU's with PPO segments. This probably leads to higher compression and tensile sets observed for the former polymers. The hysteresis of the two PEUU series is, however, little dependent on the polyether phase as studied.

4. Conclusions

The elastic properties of compacted flexible polyether (urethane–urea) foams (PEUU) with either a PPO or a PPO-*co*-PEO (PPO random copolymer with 7 wt% ethylene oxide) polyether phase were studied. These blockcopolymers also contain a tri-functional polyether and are thus chemically crosslinked.

The compression sets (2–12% at 25% compression), tensile sets (16–30% at 50% strain) and hysteresis (32–49% hysteresis at 50% strain) of the compacted PEUU's were low indicating that these materials are highly elastic. The segmented blockcopolymers based on PPO-*co*-PEO have higher T_g's and higher compression and tensile sets as compared to polymers with PPO segments. The higher amount of dissolved urea segments in the PPO-*co*-PEO phase may cause the increase in T_g and is expected to lead to slower relaxation of the soft phase, hence increasing the compression and tensile set of these PEEU materials.

Following the compression set as function of the relaxation time, i.e. the recovery, reveals two regions of relaxation: a fast and a slow (viscoelastic) recovery. The fast relaxation within the first 10 s is ascribed to elastic recovery of the soft phase. For PEUU's containing PPO-*co*-PEO this recovery is slower as compared to the recovery of the PEUU's with PPO segments. After 10 s there is a viscoelastic recovery associated with relaxation of the deformed urea phase and is similar for the two PEEU series. After 1 day the materials have recovered almost completely. The observed viscoelastic behavior of these PEUU's may be explained by rearrangements of the deformed urea phase.

Both PEUU materials exhibit a similar hysteresis and modulus-composition behavior. The increase in shear modulus with urea content for the PEUU's is stronger than can be predicted for particle filled systems [13,14] but less than expected for fiber-reinforced material [15,16] (Fig. 5). These results suggest that the PEUU's contain both spherical and fiberlike domains as has been reported before by several authors [4–8]. It is expected that these domains are the result of phase separation by liquid–liquid demixing and by crystallization of the urea segments.

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