

Polyols from recycled poly(ethylene terephthalate) flakes and rapeseed oil for polyurethane foams^{*)}

Aiga Paberza^{1), **)}, Anda Fridrihsone-Girone¹⁾, Arnis Abolins¹⁾, Ugis Cabulis¹⁾

DOI: dx.doi.org/10.14314/polimery.2015.572

Abstract: Bio/recycled polyols from rapeseed oil (RO) and poly(ethylene terephthalate) (PET) were prepared using a continuous, two-step synthesis method. The bio/recycled polyol properties were compared to RO and commercially available PET based polyols. Rigid polyurethane foams with a unified apparent density of 40–45 kg/m³ were prepared from these polyols. The thermal conductivity, thermal properties, water absorption and mechanical properties of foams were tested. The results indicated that the RO/PET polyols can be successfully used for preparing polyurethane foams as energy efficient thermal insulation material.

Keywords: rigid polyurethane foams, rapeseed oil, PET waste, renewable raw materials.

Poliole z recyklowanych płatków poli(tereftalanu etylenu) i oleju rzepakowego przeznaczone do wytwarzania pianek poliuretanowych

Streszczenie: Otrzymano biorecyklowane poliole z oleju rzepakowego i poli(tereftalanu etylenu), na drodze ciągłej dwuetapowej syntezy. Porównano ich właściwości z właściwościami polioliu uzyskanego z oleju rzepakowego i dostępnego w handlu polioliu na bazie poli(tereftalanu etylenu). Z syntetyzowanych polioli wytworzono sztywne pianki poliuretanowe o gęstości pozornej w zakresie 40–45 kg/m³. Oznaczono przewodność cieplną, właściwości termiczne, chłonność wody oraz właściwości mechaniczne otrzymanych pianek. Uzyskane wyniki wskazują, że poliole zsyntetyzowane z oleju rzepakowego i poli(tereftalanu etylenu) mogą być stosowane do produkcji pianek poliuretanowych przeznaczonych do celów termoizolacyjnych.

Słowa kluczowe: sztywne pianki poliuretanowe, olej rzepakowy, odpady PET, surowce odnawialne.

Poly(ethylene terephthalate) (PET) is one of the most widely used packaging materials in Europe and also in the world. In Europe, 65 billion postconsumer PET bottles were recycled in 2013 according to Petcore Europe, in this way PET is the most recycled polymer in Europe [1]. PET recycling can be classified in four stages: primary recycling (preconsumer industrial scrap), secondary recycling (mechanical recycling), tertiary recycling (chemical recycling) and quaternary recycling (energy recovery). From the environmental and sustainability standpoint, chemical recycling is the most favorable process because, in chemical recycling, new raw materials are produced that in turn can be used again in refineries or petrochemical and chemical production. It also conserves petrochemical resources, energy and is a partial solution for the solid PET waste problem [2].

Chemical recycling of PET can be carried using different reactions: hydrolysis, glycolysis, methanolysis and

others. The most studied method is glycolysis using different glycols — ethylene glycol, diethylene glycol, propylene glycol and others [3]. Glycolysis offers the following advantages over other recycling methods — lower reagent amounts (it is not necessary to employ a large excess of reactant), lower temperatures and pressure [4]. PET glycolyzate can be used as a polyol for polyurethane (PUR) foam or polyisocyanurate (PIR) foam production. The use of these polyols improves the mechanical and thermal properties of the resulting PUR foams. Better thermal properties allow a reduction in the consumption of flame retardants in the final PUR formulation. However, these PET polyols also have major drawbacks — high viscosity polyols and incompatibility with the physical blowing agents. PET glycolyzate is not compatible with the physical blowing agents (pentanes and hydrofluorocarbons), which are currently allowed foaming agents under the Montreal Protocol. This results in unstable polyol systems that separate during storage and if such a system is used for the production of PUR/PIR foams, the dimensional stability and other properties of the resulting PUR/PIR foams are affected negatively [5–7].

Several authors have reported that vegetable oil based polyols show good compatibility with physical blowing

¹⁾ Polymer Laboratory, Latvian State Institute of Wood Chemistry, 27 Dzerbenes St., LV-1006, Riga, Latvia.

^{*)} Material presented at the conference „Polyurethanes 2015 — cooperation for innovation”, September 9–11, 2015, Cracow, Poland.

^{**)} Author for correspondence; e-mail: aiga.paberza@gmail.com

agents. Yakushin *et al.* reported that polyols synthesized from rapeseed oil (RO) or sunflower oil amidated with diethanolamine show good compatibility with components conventionally used in polyol systems [8]. Guo *et al.* reported that a soy based polyol showed greater compatibility with cyclopentane than a commercially available petroleum derived polypropylene based polyol [9].

To decrease the viscosity and increase the compatibility of PET containing polyols with physical blowing agents, new type of polyols should be synthesized that would offer advantages from “both worlds” — aromatic moieties from PET and lipophilic nature of the vegetable oils. However, in the existing literature, there are not many papers that describe polyols from both renewable

bio/recycled polyols. The properties of the final bio/recycled polyols were investigated, as well as bio/recycled based PUR foams obtained, tested and compared to bio based foams from RO polyol and foams from PET based polyol.

EXPERIMENTAL PART

Materials

A short description and sources of the reagents used for the synthesis of RO/TEA, RO/PET polyols and PUR foams in this study are given in Table 1. All the reagents were used without any prior treatment.

Table 1. Reagents used for the synthesis of RO/PET polyols

Materials	Supplier	Description
PET flakes	PET Baltija, Latvia	Clear PET flakes
Rapeseed oil (RO)	Iecavnieks&Co, Latvia	I value = 117 I ₂ mg/100 g
Triethanolamine (TEA)	BASF, Germany	99.5 %
Zinc acetate dehydrate	Sigma Aldrich, Germany	≥ 98 %
Diethylene glycol (DEG)	Sigma Aldrich, Germany	99 %
Lupranol 3422	BASF, Germany	OH value = 490 mg KOH/g
Neopolyol 380 (NEO)	NEO GROUP, Lithuania	OH value = 370–390 mg KOH/g
Solkane 365/227	Solvay, Belgium	Pentafluorobutane / Heptafluoropropane (87/13)
PC CAT NP-10	Performance Chemicals Handels, Germany	
NIAX Silicone L6915	Momentive Performance Materials, Germany	
Trichlorpropylphosphate	Lanxess, Germany	
Polymeric diphenylmethane diisocyanate IsoPMDI 92140 (pMDI)	BASF, Germany	NCO group content = 31.5 %

and recycled resources. Cakić *et al.* synthesized biorenewable polyols in a two stage synthesis: the first, glycolysis of PET using different molar ratios of the PET repeating unit to poly(ethylene glycol) PEG400 at different molar ratios; the second, transesterification of the oligoester polyols with different weight ratios of oligoester polyol to castor oil [10]. Beneš *et al.* carried out the depolymerization of PET using castor oil as a reagent and microwaves. They found that using castor oil as a reagent for PET depolymerization offers the following advantage — presence of long acylester chain in castor oil prevents the formation of cyclic oligomers that form when conventional PET depolymerization is carried out using different glycols [7].

The present paper describes polyols containing both renewable and recycled raw materials and synthesis using a continuous two-step method: (1) synthesis of rapeseed oil using transesterification with triethanolamine (TEA); (2) PET depolymerization with DEG at different molar ratios in the synthesized RO polyol media. The synthesized polyol from a combination of both, bio based raw materials and recycled PET, were named

Synthesis of RO/PET polyols

RO/PET polyols were synthesized using a two-step, continuous synthesis method. The reaction was carried out in a three neck 1.0 dm³ reaction flask equipped with a mechanical stirrer, a thermometer, a condenser and an argon inlet.

The first stage of the synthesis was to synthesize RO polyols using transesterification with triethanolamine (TEA), which was carried out at 170 ± 5 °C for 1 hour, 0.15 wt % zinc acetate was used as catalyst. The molar ratio was 1 M of RO to 2.9 M of TEA. A more detailed description of RO polyol synthesis is given in our previous work, Stirna *et al.* [11]. The end of the reaction was tested by the solubility of the obtained product in ethanol at the volume ratio (resin:ethanol = 1:1). When the product — RO/TEA polyol — was obtained, the temperature was increased to 230 °C and the second synthesis step — PET depolymerization — was carried out. To the mixture, PET flakes and DEG were added. The PET flakes were depolymerized at different molar ratios of PET to DEG using 0.5 wt % (based on the weight of PET) zinc acetate

as a catalyst. The molar ratios of PET to DEG were 1:2; 1:4 and 1:6. The reaction was carried out at 220–230 °C to conduct the glycolysis for 4 hours after which the flask was allowed to cool. Samples were named RO/PET-2, RO/PET-4 and RO/PET-6 where the number indicates the molar ratio of DEG.

Overall, the molar ratios of RO:TEA:PET:DEG were 1:2.9:3.2:6.5 for RO/PET-2 polyol, 1:2.9:2.1:8.5 for RO/PET-4 polyol and 1:2.9:1.6:9.5 for RO/PET-6 polyol.

To compare RO/PET polyols with vegetable oil based polyol, RO/TEA polyol was also synthesized as described above (first step of synthesis) and in our previous paper by Stirna *et al.* [11]. Also, commercial polyol NEO based on industrial PET scrap was chosen. NEO is synthesized using PET scraps and DEG. NEO was used without any prior treatment.

Preparation of polyurethane foams

The foams were prepared by the free-rising method in a mold, at room temperature. At first, the component was mixed in a 500 cm³ plastic beaker. Afterwards isocyanate was added and mixed for 15 s at a speed rate of 2000 rpm. Then, the mixture was quickly poured into an open mold (30×30×10 cm). The height of these foams was ~10 cm. The foams were conditioned for 24 h at room temperature.

Samples for different tests from prepared PUR foam blocks were cut using a band saw and afterwards they were conditioned for at least 24 h at room temperature.

Methods of testing

– The OH value was determined using the conventionally used acetic acetylation method. The acid value was determined according to DIN 53402. The hydroxyl and acid value of NEO was also tested in the laboratory.

– Viscosity measurements were carried out using a Haake Viscotester 6L/R plus at 20 °C.

– The moisture content in the polyol was tested using a conventionally used Karl Fischer method.

– The Fourier transform infra-red spectra measurements (FTIR-ATR) were made on a Perkin-Elmer spectrometer Spectrum One FTIR Spectrometer. The FTIR-ATR spectra of polyols were collected at a resolution of 4 cm⁻¹.

– The polyol compatibility with blowing agents was tested in plastic centrifuge tubes where 10 g of polyol was weighed and then 20 % of blowing agent was added. Then, the blends were centrifuged for 15 minutes at 55 rpm. The blends were then kept at ambient conditions for regular visual inspection.

– The compression strength and modulus of elasticity parallel and perpendicular to foaming direction of PUR foams were tested according to the requirements of ISO 844:2009 standard on testing machine Zwick/Roell Z100.

– The bulk density of obtained PUR foams was tested according to the ISO 845 standard.

– The thermal conductivity coefficient (λ , W/m·K) was tested using a Linseis Heat Flow Meter 200 according to ISO 8301 standard.

– The TGA and its derivative thermograms (DTGs) were studied using a Mettler Toledo TGA/SDTA 851e instrument. The heating rate was set to 10 °C/min under a flow of 20 cm³/min nitrogen and a temperature range was set from 25 to 1000 °C. The weight of the sample was ~8 mg.

– Water absorption was tested according to ISO 2896:2001 standard by immersing PUR samples in water for 7 days.

– Closed cell content was tested according to ISO 4590:2003, PUR samples with dimension (10×3.5×3.5 cm) were cut.

RESULTS AND DISCUSSION

Polyol characterization

The acid numbers, hydroxyl value, water content and viscosity of synthesized polyols are given in Table 2.

Table 2. Characteristics of polyols from RO and PET

Polyol	OH value mg KOH/g	Acid value mg KOH/g	Water content, %	Viscosity at 20 °C mPa·s
RO/TEA	363	2.5	0.12	160
RO/PET-2	233	3.2	0.23	1250
RO/PET-4	340	2.8	0.19	540
RO/PET-6	368	2.3	0.27	250
NEO	366	2.9	0.12	9640

All polyols used in this study have low acid numbers, which are less than 5 mg KOH/g, and thus all polyols are favorable for the production of rigid PUR foams. With an increase of the molar ratio of PET to DEG from 1:2 M to 1:6 M, the hydroxyl number of the derived polyols increased by 58 % and their viscosity decreased by 80 %. The results indicate that with increasing ratio of glycol to PET, the extent of depolymerization was greater. Aromatic polyol NEO showed the highest viscosity of all polyols. Except for RO/PET-2 polyol, the hydroxyl value of all polyols was in a comparable range. The water content of all polyols is admissible.

The polyols were also characterized by bio/recycled content (Table 3). The highest total bio/recycled content was for RO/TEA polyol – 67 % but this polyol does not contain PET. The total bio/recycled content of RO/PET polyols increased up to 57 % if the PET depolymerization was carried out at lower molar ratio of PET to DEG.

Depolymerization of PET by a two step continuous synthesis method with RO/TEA polyol and DEG was

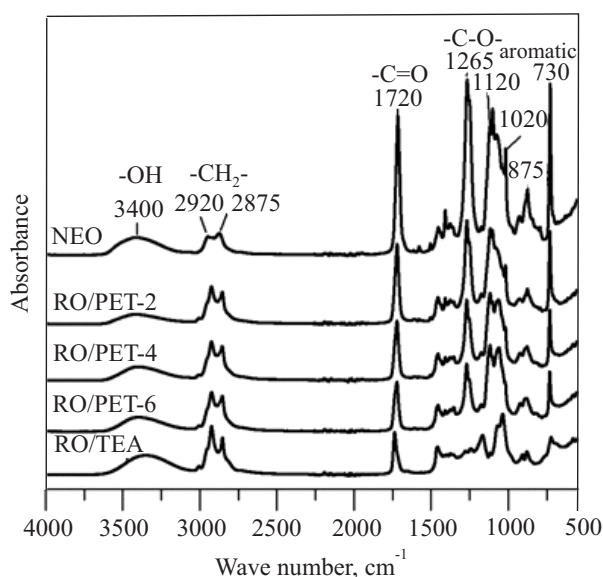


Fig. 1. FTIR-ATR spectra of polyols

confirmed using FTIR-ATR. The FTIR-ATR results of the RO/PET polyols obtained at different molar ratios are presented in Fig. 1. To compare the polyols, spectra of RO/TEA and NEO polyol were added.

Table 3. Renewable and recycled content in RO/TEA polyols

Polyol	Renewable content, %	Recyclate content, %	Total renewable + recyclate content, %
RO/TEA	67.0	-	67.0
RO/PET-2	33.5	23.8	57.3
RO/PET-4	33.5	15.6	49.1
RO/PET-6	33.5	11.6	45.1
NEO	-	45–48*	45–48

* Information obtained from the producers.

The band around 3400 cm^{-1} is due to the free hydroxyl groups present in all polyols. The band at 3400 cm^{-1} is stronger for RO/TEA, NEO and RO/PET-6 polyol. These results correlate with the higher hydroxyl values of these polyols in comparison to RO/PET-2 and RO/PET-4 polyols synthesized at lower glycol molar ratios. There is no absorption of aromatic groups for RO/TEA polyol. In the $3000\text{--}2850\text{ cm}^{-1}$ range, two bands can be observed due to symmetric and asymmetric stretching of CH_2 groups in all polyols. For polyols containing RO, the two bands in this region are more pronounced due to the presence of long fatty acid radicals.

For all polyols, there is a sharp band at 1720 cm^{-1} , which confirms that the bio/recycled polyols also contain an ester bond in their structure. It can also be seen that the peak around 1720 cm^{-1} becomes sharper and more intense with an increase in the amount of PET in the RO/PET polyols (Table 3), which is due to an increase in the number of ester linkages in the polyols.

In future, more experiments should be carried out to study the chemical structure of RO/PET polyols.

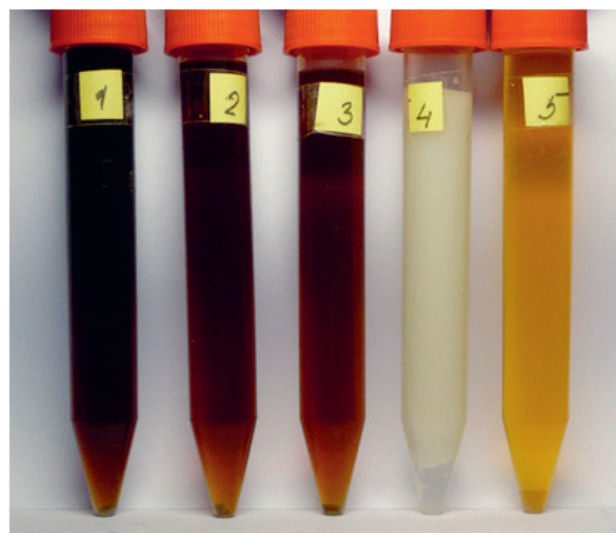


Fig. 2. Compatibility of polyols with blowing agent – Solkane 365/227; samples: 1) RO/PET-2; 2) RO/PET-4; 3) RO/PET-6; 4) NEO; 5) RO/TEA

Polyols compatibility with physical blowing agents

Experiments were carried out to test the compatibility of the polyols with physical blowing agents. Solkane 365/227 was chosen as a blowing agent. As expected, the aromatic polyester polyol NEO, based on PET scraps, showed poor compatibility with the blowing agent (Fig. 2, sample 4). All RO/PET polyols and also RO/TEA polyol showed complete compatibility with Solkane 365/227. This is due to the presence of long, hydrophobic and non-polar dangling chains in the polyol structure, which are introduced with RO.

The darker color residue at the bottom of sample 5 tube (RO/TEA polyol) is glycerol, which forms as a by-product during RO/TEA polyol synthesis. We have also reported this previously [11].

Formulation and foaming parameters of rigid PUR foams

Rigid PUR foam samples were prepared using synthesized polyols RO/PET-2, RO/PET-4, RO/PET-6, RO/TEA and commercially available NEO polyol. Foam samples were named accordingly to the polyol used and adding “PUR” before it. For example, PUR RO/PET-2 is a foam sample from polyol RO/PET-2. The detailed formulations of PUR samples is given in Table 4.

The amount of necessary pMDI was calculated according to isocyanate index 130. It is well known that the apparent density of PUR foams affects several foam properties, such as compression strength and thermal conductivity. Therefore, the amount of blowing agent Solkane 365/227 was adjusted to obtain PUR foams with a unified apparent density $40\text{--}45\text{ kg/m}^3$. This range of apparent density is suitable for rigid PUR foams used for thermal insulation in the construction industry.

Table 4. Formulation of rigid PUR foams

Component	Part by weight				
	PUR NEO	PUR RO/PET-2	PUR RO/PET-4	PUR RO/PET-6	PUR RO/TEA
Corresponding polyol			75		
Lupranol 3422			25		
Trichlorpropylphosphate			16		
NIAX Silicone L6915			1.5		
Water			2.2		
PC CAT NP-10	1.6	1.6	1.6	1.6	0.3
Solkane 365/227	14.0	16.0	16.0	16.0	15.0
pMDI	165.1	137.5	161.7	169.5	166.5

The foaming process was monitored by measuring the duration of start time (when foam begins to rise), gel time (indicating a transition of the mix from liquid to solid state) and tack-free time (when the foam solidifies completely). The results of the foaming parameters are presented in Table 5.

Table 5. Foaming parameters

Sample	Cream time, s	Gel time, s	Tack-free time, s
PUR NEO	70	100	135
PUR RO/PET-2	18	60	95
PUR RO/PET-4	23	53	80
PUR RO/PET-6	26	53	100
PUR RO/TEA	18	50	90

The results of the foaming parameters show that a lower amount of foaming catalyst is necessary for the PUR foam formulation when RO/TEA polyol is used (Table 4). The amount of catalyst PC CAT NP-10 was reduced in the PUR RO/TEA formulation to prolong the start time of foaming. It is necessary to decrease the start time otherwise the buildup of viscosity is too fast and it is not possible to pour the mixture into the mold. This is due to the fact that RO/TEA polyol also contains a tertiary amine group in its structure, which acts as a catalyst. So RO/TEA polyols can be characterized by a catalytic activity in urethane-forming reactions as reported in previous papers by our group [12]. Also, the synthesized bio/recycled polyols RO/PET show increased catalytic activity in comparison to NEO polyol. The start and gel

time of PUR foams was shorter for formulations where RO/PET polyols were used in comparison to the start and gel times of PUR foams from NEO polyol. All four PUR RO/PET formulations contained the same amount of foaming catalyst (Table 4).

Thermal conductivity of rigid PUR foams

Thermal conductivity is one of the most important physical properties of PUR foams and it determines their potential application. The thermal conductivity of PUR foams is from about 18 up to 28 mW/(m·K) [13]. It is influenced by several factors such as conduction through the polymer phase, conductivity through the gas trapped within the closed cell structure and the radiation between cells [14]. In the present study, the closed cell content of rigid PUR foam samples is ~95 %. The thermal conductivity of the obtained PUR foams is in a range from 19.5 to 21.5 mW/m·K (Table 6).

Over time, the thermal conductivity of PUR foams increases because the foaming agent with low thermal conductivity becomes diluted with air as it diffuses into the foam. Air has a higher thermal conductivity than foaming agent. The foaming agent also permeates the cell walls and escapes from the foam [15]. A small change (1.5–2.7 %) in the thermal conductivity of PUR NEO and for all PUR RO/PET samples is observed after 1 week. The best and the worst retentions of thermal conductivity were shown by foam samples PUR RO/PET-4 and PUR RO/TEA, respectively. As shown in Table 6, the thermal conductivity of PUR foams from RO/TEA polyol increased by 9.4 %.

Table 6. Thermal conductivity of rigid PUR foams

Sample	Closed cell content vol. %	Initial thermal conductivity mW/m·K	Thermal conductivity after 1 week, mW/m·K	Change in thermal conductivity after 1 week, %
PUR NEO	96 ± 1	19.49 ± 0.50	19.82 ± 0.50	1.7
PUR RO/PET-2	95 ± 1	20.54 ± 0.06	21.10 ± 0.06	2.7
PUR RO/PET-4	95 ± 1	20.65 ± 0.01	20.96 ± 0.01	1.5
PUR RO/PET-6	96 ± 1	20.86 ± 0.50	21.27 ± 0.50	2.0
PUR RO/TEA	95 ± 1	21.50 ± 0.25	23.52 ± 0.25	9.4

Water absorption of rigid PUR foams

Water absorption was tested by immersing PUR foam samples in water for 7 days. The results of water absorption are shown in Table 7.

Table 7. Water absorption of rigid PUR foams

Sample	Water absorption after 1 day, vol. %	Water absorption after 7 days, vol. %
PUR NEO	1.02 ± 0.05	2.11 ± 0.05
PUR RO/PET-2	1.58 ± 0.08	2.34 ± 0.13
PUR RO/PET-4	1.46 ± 0.10	2.56 ± 0.03
PUR RO/PET-6	1.56 ± 0.05	2.75 ± 0.36
PUR RO/TEA	1.97 ± 0.03	3.42 ± 0.08

The worst performance in water absorption experiments was observed for the PUR RO/TEA sample. It showed the highest water absorption after 1 day (1.97 vol. %) and also after 7 days (3.42 vol. %). PUR foams from RO/PET polyols and NEO polyol were more moisture-resistant.

Thermal stability of rigid PUR foams

The thermal stability of a material reflects the ability of its chemical structure to resist chain termination caused by heat. This greatly depends on the polyol structure.

Rigid PUR foams from aromatic polyols are thermally more stable than foams from aliphatic polyols [16]. Therefore, it can be expected that PUR foams from NEO and RO/PET polyols will show higher thermal stability than PUR foams from RO/TEA polyol. RO/TEA is an aliphatic polyol and does not contain any aromatic rings in its structure. TGA and DTG results are presented in Fig. 3 and Fig. 4, respectively. All PUR foams from RO/PET polyols show similar TGA and DTG curves, therefore only PUR RO/PET-4 was chosen for presentation.

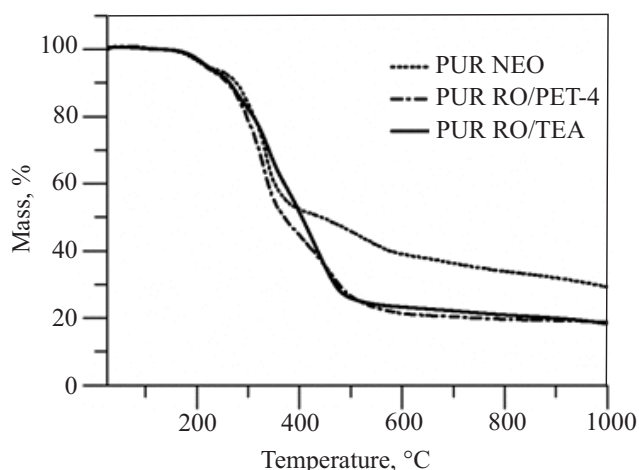


Fig. 3. TGA results of rigid PUR foams

The DTG curves show three peaks corresponding to three steps of degradation (Fig. 4) of PUR foams from NEO polyol and RO/PET-4 polyol. The three peaks are observed at about 200–210 °C, at ~330–340 °C and the

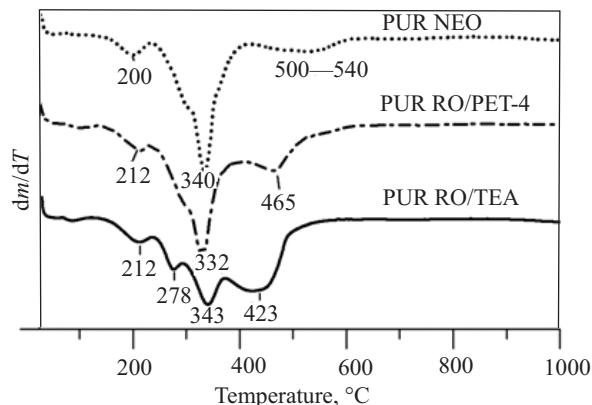


Fig. 4. DTG results of rigid PUR foams

final peak at ~450–500 °C. PUR RO/TEA also showed an additional peak at 278 °C. In the first step, low molecular weight compounds evaporate [16]. Also, allophanate and biuret crosslinks decompose [17]. In the second step, degradation starts at the urethane bond and other segments of the remaining structure [18]. Weight loss in the third stage is due to the degradation of the strongest bonds in the PUR backbone [19].

The thermal stability of PUR foams based on NEO polyol containing aromatic groups was higher than the thermal stability of PUR foams based on RO/TEA and RO/PET polyols. The char residues at 1000 °C for PUR foams from NEO, RO/PET-4 and RO/TEA polyols were 29.3 %, 18.8 % and 18.3 %, respectively (Fig. 3). The higher char residue of PUR NEO is due to the presence of aromatic groups in the polyol structure.

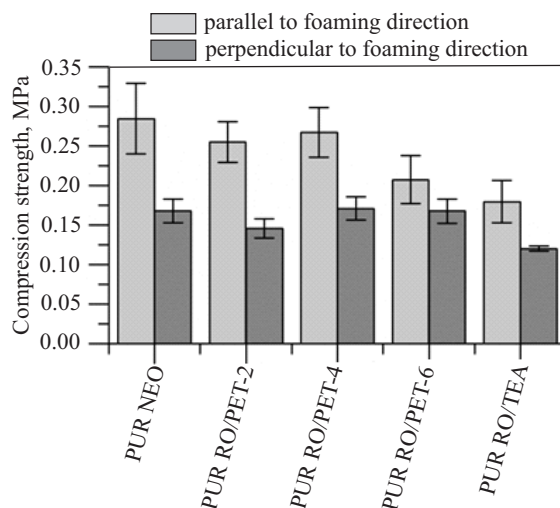


Fig. 5. Compression strength of rigid PUR foams

Compression strength of rigid PUR foams

It is well known that for rigid PUR foams obtained by the free rise method that anisotropy is observed. Therefore, compression tests were carried out in the foam rise direction and perpendicular to it. The mechanical properties of rigid PUR foams are significantly related to their apparent density. Due to this, all compression strength results were normalized with respect to a density of 45 g/m^3 using the equations of Hawkins *et al.* [20]. The coefficients of anisotropy calculated from the compression strength results (Fig. 5) are in the range from 1.2 to 1.7.

PUR foams obtained from RO/PET polyols showed higher compression strengths than foams from RO/TEA. The highest compression strength ($0.27 \pm 0.03 \text{ MPa}$) of RO/PET foams was for the sample PUR RO/PET-4. Within the margin of experimental error, it reaches the same compression strength as for PUR NEO ($0.28 \pm 0.03 \text{ MPa}$).

CONCLUSIONS

The synthesis of polyols from recycled poly(ethylene terephthalate) and rapeseed oil was successfully achieved using a two-step continuous synthesis method. With higher molar ratios of diethylene glycol, the rapeseed/poly(ethylene terephthalate) polyols showed increased hydroxyl numbers and decreased viscosity. High reactivity and high bio/recycled content combined with excellent compatibility with blowing agents make these polyols suitable for the preparation of rigid polyurethane foams. Further studies on detailed chemical mechanisms are crucial to understand the chemical reactions and their effect on the properties of polyols and rigid polyurethane foams.

Rigid polyurethane foams from bio/recycled polyols showed decreased thermal conductivities and water absorptions in comparison to polyurethane foams obtained from rapeseed oil triethanolamine polyol. In addition, polyurethane foams from rapeseed/poly(ethylene terephthalate) polyols showed a better retention of thermal conductivity. The results show that the most favorable molar ratio of poly(ethylene terephthalate) to diethylene glycol is 1:4. Rigid polyurethane foams obtained using this polyol showed the best compression strength, water absorption, retention of thermal conductivity and foam samples were dimensionally stable without shrinkage. Overall, the results indicated that the rapeseed/poly(ethylene terephthalate) polyols can be successfully used for preparing polyurethane foams as energy efficient, thermal insulation material. Also, it is a promising and perspective material for the use in construction industry and domestic appliances *etc.*

ACKNOWLEDGMENTS

The financial support of the European Regional Development Fund Contract No 2014/0043/2DP/2.1.1.1.0./14/APIA/VIAA/063 is gratefully acknowledged.

REFERENCES

- [1] <http://www.petcore.org/sites/default/files/generated/files/news/Press%20release%20-%20Petcore%20Europe%20-%20Europe%20recycled%20over%2065%20billion%20PET%20bottles%20in%202013%21.pdf> (access date 12.12.2014)
- [2] <http://ec.europa.eu/environment/waste/studies/pdf/plastics.pdf> (access date 12.12.2014)
- [3] Sinha V., Patel M.R., Patel J.V.: *Journal of Polymers and the Environment* **2010**, 18, 8.
<http://dx.doi.org/10.1007/s10924-008-0106-7>
- [4] Sabnis S., Bhavne V.G., Kathalewar M.S. *et al.*: *Archives of Applied Science Research* **2012**, 4, 85.
- [5] Vitkauskienė I., Makuška R.: *Chemija* **2008**, 19, 29.
- [6] Ionescu M.: "Chemistry and Technology of Polyols for Polyurethane", Smithers Rapra Press., Shawbury, Shrewsbury, Shropshire, UK, 2008, p. 602.
- [7] Beneš H., Slabáb J., Walterová Z., Raisa D.: *Polymer Degradation and Stability* **2013**, 98, 2232.
<http://dx.doi.org/10.1016/j.polyimdegstab.2013.08.019>
- [8] Yakushin V., Stirna U., Sevastyanova I. *et al.*: *Materials Science (Medžiagotyra)* **2008**, 14, 333.
- [9] Tana S., Abrahamb T., Ferenceb D., Macosko C. W.: *Polymer* **2011**, 52, 2840.
<http://dx.doi.org/10.1016/j.polymer.2011.04.040>
- [10] Cakić S.M., Ristić I.S., Cincović M.M. *et al.*: *Progres in Organic Coatings* **2015**, 78, 357.
<http://dx.doi.org/10.1016/j.porgcoat.2014.07.012>
- [11] Stirna U., Fridrihsone A., Lazdiņa B. *et al.*: *Journal of Polymers and the Environment* **2013**, 21, 952.
<http://dx.doi.org/10.1007/s10924-012-0560-0>
- [12] Stirna U., Fridrihsone-Girone A., Yakushin V., Vilsone D.: *Journal of Coatings Technology and Research* **2014**, 11, 409.
<http://dx.doi.org/10.1007/s11998-013-9545-8>
- [13] Zatorski W., Brzozowski Z.K., Kolbrecki A.: *Polymer Degradation and Stability* **2008**, 93, 2071.
<http://dx.doi.org/10.1016/j.polyimdegstab.2008.05.032>
- [14] Septevani A.A., Evans D.A.C., Chaleat C. *et al.*: *Industrial Crops and Products* **2015**, 66, 16.
<http://dx.doi.org/10.1016/j.indcrop.2014.11.053>
- [15] Shankland I.R.: "Insulation Materials: Testing and Applications, ASTM STP 1030", (Eds. McElroy D.L., Kimpflen J. F.), American Society for Testing and Materials, Philadelphia 1990, p. 759.
- [16] Vitkauskienė I., Makuška R., Stirna U., Cabulis U.: *Materials Science (Medžiagotyra)* **2011**, 17, 249.
<http://dx.doi.org/10.5755/j01.ms.17.3.588>
- [17] Chattopadhyay D.K., Webster D.C.: *Progress in Polymer Science* **2009**, 34, 1068.
<http://dx.doi.org/10.1016/j.progpolymsci.2009.06.002>
- [18] Oprea S.: *Polimery* **2009**, 54, 120.
- [19] Narine S.S., Kong X., Bouzidi L., Sporns P.: *Journal of the American Oil Chemists' Society* **2007**, 84, 65.
<http://dx.doi.org/10.1007/s11746-006-1008-2>
- [20] Hawkins M.C., O'Toole B., Jackovich. D.: *Journal of Cellular Plastics* **2005**, 41, 267.
<http://dx.doi.org/10.1177/0021955X05053525>