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## The influence of expandable polystyrene fillers on properties of water-blown rigid polyurethane foams

### RAPID COMMUNICATION

**Summary** — This work presents the results of study of fillers addition influence on selected properties of polyurethane/polystyrene porous composite. The porous materials based on water blown rigid polyurethane foam (RPURF) matrix containing thermoplastic expandable polystyrene (EPS) beads as the fillers were obtained. In RPURF-EPS composites, the EPS beads were expanded after heating above the glass transition temperature of EPS and vaporizing EPS gas incorporated inside, by using the heat of exothermic reaction of polyol with isocyanate. The thermal conductivity, compressive strength, core apparent density and dimensional stability of new materials were investigated.

**Keywords:** composite, rigid polyurethane foam, expandable polystyrene, thermal conductivity.

WPLYW ZDOLNYCH DO EKSPANDOWANIA NAPEŁNIACZY POLISTYRENOWYCH NA WŁAŚCIWOŚCI SZTYWNYCH PIANEK POLIURETANOWYCH SPIENIANYCH WODĄ

**Streszczenie** — W artykule przedstawiono wyniki badań dotyczących wpływu dodatku napełniacza na wybrane właściwości porowatego kompozytu poliuretanowo-polistyrenowego (RPURF-EPS). W otrzymanym kompozycie spieniany wodą poliuretan (RPURF) stanowił matrycę, natomiast napełniaczem były zdolne do ekspandowania perełki polistyrenu (EPS). Zawarte w mieszaninie perełki EPS pod wpływem ciepła egzotermicznej reakcji polioliu i izocyjanianu zostały ogrzane do temperatury ich zeszczenia i ekspandowały na skutek odparowania poroforu zawartego w ich wnętrzu. Wykonano oznaczenia właściwości użytkowych otrzymanych materiałów tj. współczynnika przewodzenia ciepła, wytrzymałości na ściskanie, gęstości pozornej oraz stabilności wymiarowej.

**Słowa kluczowe:** kompozyt, sztywna pianka poliuretanowa, zdolny do ekspandowania polistyren, współczynnik przewodzenia ciepła.

Fillers are added to polymers in order to reduce composite cost and/or to improve processing behavior and/or to modify product properties [1]. Fillers are inexpensive, thus using them allows making the material cheaper but cost reduction is no longer the only reason, nor even the most important. Fillers influence density and other material characteristic such as: optical, surface, electrical, magnetic, mechanical and rheological properties, and chemical reactivity, thermal stability, flame retardancy *etc.* [2–4].

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Fillers are also added to the formulation of foams although their production is very sensitive to changes in composition and the parameters of processing. The inclusion of fillers complicates the process and careful investigation must be performed to learn the effects that filler incorporation has on material properties. Fillers could reduce shrinkage and combustibility of polymer foams. Typical powder fillers used for preparation of rigid polyurethane foams (RPURF) are aluminum hydroxide, melamine, starch, talk, chalk, borax [5], calcium carbonate, barium sulfate [6], crystallized silica [7] and nanofillers like bentonite [8] and expanded graphite [9].

Cracow University of Technology in cooperation with HIT Consulting Sp. z o.o. carried out investigations on the method to obtain new composites based on RPURF and expandable polystyrene (EPS). The method to prepare RPURF-EPS composite was described in patents [10, 11]. The filler of RPURF matrix should be a thermoplastic

granulate with a blowing agent inside. The filler should have the softening point below 90 °C. The beads' size is suggested to have diameters in the range between 0.2 and 4.0 mm. The amount of EPS added to the polyurethane reaction mixture should be in the range of 20–120 wt. % in relation to polyurethane reagents. The idea of this process is co-expansion of EPS and RPURF and full energy balance. EPS is expanded by using the heat of exothermic polyol and isocyanate reaction. This technology allows creating a new composite material based on well known and widely applied polymers.

The aim of this work was to study the influence of the addition of EPS in amount 40 wt. % on selected properties of RPURF-EPS composites such as thermal conductivity, compressive strength, core density and dimensional stability.

## EXPERIMENTAL

### Materials

The materials used for composite preparation are listed below:

– Owipian<sup>®</sup> 1325 (Synthos S.A., Oświęcim, Poland) in the form of polystyrene beads with main particle size 1.60–2.40 mm, containing 5–6 wt. % of pentane isomers as an expanding agent and hexabromocyclododecane (up to 0.5 wt. %) which reduces flammability of the foam.

– PUREX WG 2034 NF (product of Polychem Systems Sp. z o.o., Poznań, Poland) which was a two component system for production of rigid polyurethane foam for insulation. Component A was a mixture of polyols, a catalyst, a silicone surfactant and water as a blowing agent. Component B was a monomer and polymer mixture of methylene-4,4'-diphenyl diisocyanate (30–32 % of NCO group). Both components were mixed with mass ratio A:B = 100:150 (start time 18–24 s).

### Foams sample preparation

The RPURF-EPS composites and reference RPURF were synthesized using the one-shot method at the room temperature. The reference foam did not contain EPS beads. The RPURF-EPS composites contained 40 wt. % of EPS beads (66.7 wt. % according to the mass of polyurethane reagents) were prepared with the method described in [12, 13]. Component B was mixed with the EPS beads. Then, an appropriate amount of component A was added to the mixture and vigorously stirred at 1 200 rpm for 10 seconds. After mixing, the mixture was poured into the 300 × 300 × 100 mm metal mold which was heated to 50 °C. The foams were cured in the mold for 30 min in order to get dimensionally stable products. After that foams were removed from the mold and conditioned for 24 hours at the room temperature before being cut into appropriate specimens for testing.

### Methods of testing

– The core apparent density of the RPURF-EPS composites was determined according to the ISO 845 standard using cubical samples of 50 mm side by measurement their volume and mass.

– Closed cell content was measured according to the PN-EN ISO 4590 standard using samples of size 100 × 25 × 25 mm. The test is based on measurement of gas volume displaced by samples inserted into a closed cell apparatus chamber.

– Water absorption was measured according to the PN-69/C-89084 standard. Samples (100 × 100 × 25 mm) were precisely weighed on an analytical balance and put into water for 24 hours. After that time samples were weighed again and the amount of absorbed water was calculated.

– Dimensional stability of the RPURF-EPS composites was studied according to the PN-EN 1603 standard using original samples. Linear dimensions (width) of samples were measured using caliper after 24 hours, 48 hours and 28 days.

– Mechanical properties were determined on a Zwick 1445 universal testing machine at the room temperature. The measurement of compressive strength of the RPURF-EPS composites was performed according to the ISO 844 standard. The force required to deform the composites by 10 % of their original thickness has been taken as the compression strength of the foams. The speed of the crosshead movement was 5 mm/min. Tests were performed in the parallel and perpendicular directions to the foam rise.

– Thermal conductivity of the RPURF-EPS composites was measured using the FOX 314 apparatus according to the PN-EN 12667 standard using samples 50 mm thick.

## RESULTS AND DISCUSSION

As it was mentioned above, the RPURF and the RPURF-EPS composite were prepared in order to estimate the influence of EPS on the material properties. The addition of EPS as a filler to RPURF made it possible to obtain products that differ in apparent density, dimensional stability, mechanical properties and thermal conductivity. The cross-sections appearance of the prepared foams and characteristics of the samples are presented in Figure 1 and Table 1, respectively.

The result shows that the apparent density of RPURF-EPS is considerably higher than that of RPURF. The EPS was expanded by using heat of exothermic polyol and isocyanate reaction, therefore the density of the composite filled with 40 wt. % of EPS rose only to 50 kg/m<sup>3</sup>. The value of water absorption is similar to the reference foam and composites, but the addition of EPS influences divergence of results. It was observed that the compressive strength of the RPURF-EPS composites in

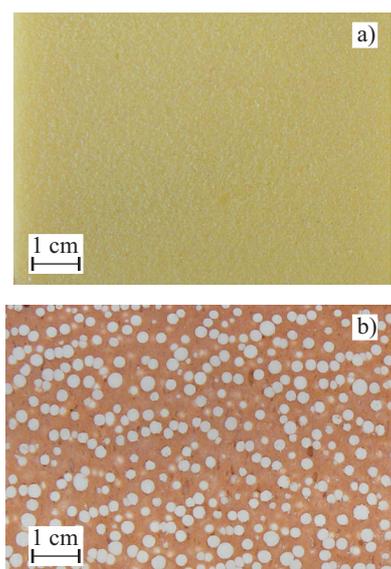


Fig. 1. The cross-section appearance of foam samples: a) RPURF, b) RPURF-EPS

the parallel and perpendicular direction to the foam rise direction was worse in the presence of EPS.

**Table 1. Physical properties of reference RPURF and RPURF-EPS (with 40 wt. % of EPS beads) composite foam**

Property of sample, unit	RPURF	RPURF-EPS
Apparent density, kg/m <sup>3</sup>	39.8 ± 0.2	50.3 ± 0.6
Closed cell content, %	75 ± 2	37 ± 4
Water absorption, wt. %	2.1 ± 0.2	2.0 ± 0.8
Dimensional stability after 2 days, %	-0.90 ± 0.02	-0.70 ± 0.02
Dimensional stability after 28 days, %	-2.10 ± 0.02	-0.73 ± 0.02
Compressive strength – parallel to the rise direction, kPa	131 ± 4	104 ± 3
Compressive strength – perpendicular to the rise direction, kPa	105 ± 8	72 ± 5
Thermal conductivity after 24 hours, mW/(m · K)	24.5	26.1

Additionally the use of EPS in composite improves the sample dimensional stability in comparison to the reference foam. The width of RPURF-EPS sample after 2 and 28 days changed in both cases by -0.7 % and was stable while the width of the reference RPURF foam changed by -0.9 and -2.1 %, respectively.

The RPURF-EPS composite had lower percentage of closed cell content than the reference foam. It may have been caused by the addition of EPS which expands in polyurethane matrix and upsets (destroys) their structure. A relatively high thermal conductivity measured after 24 hours, observed for the RPURF-EPS composite may be explained by a worse thermal conductivity of expanded EPS beads and lower percentage of closed cells in the composite.

Thermal conductivity ( $\lambda$ ) of foams changed gradually during long ageing, what is shown in Figure 2. Increasing of  $\lambda$  is strongly influenced by the blowing agent diffusion from the foam. Seven days after materials were made,

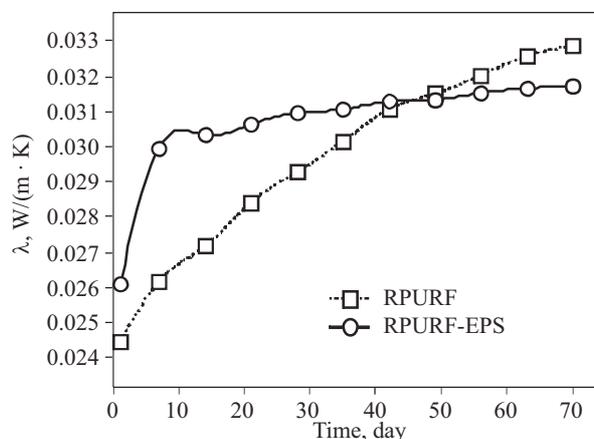


Fig. 2. Effect of the ageing time on thermal conductivity for reference RPURF and RPURF-EPS composite samples

thermal conductivity of the RPURF-EPS composite increased by about 15 %, while the reference RPURF by only about 7 %. This phenomenon can be explained by the higher content of open cells in the composite material, which affects a higher diffusion rate. The diffusion rate of the blowing agent in the reference foam is stable for all time periods, while in the composite it decreases after 7 days. Therefore both foamed materials show the same heat insulating properties after the 40 days period. After the 50 days period of ageing, thermal conductivity of the reference foam raised above the RPURF-EPS composite value. As the blowing agent the reference foam had only carbon dioxide, while RPURF-EPS composite had carbon dioxide and pentane isomers. The pentane contained in the EPS beads has a lower diffusion rate than carbon dioxide therefore the increase of  $\lambda$  is faster in the case of the reference foam.

## CONCLUSION

The obtained results prove that modification of the RPURF, foamed by the chemical blowing agent with the addition of EPS beads, is possible and the prepared composites have desirable properties. Rigid polyurethane foams filled with EPS show very good mechanical properties and dimensional stability. With regard to this, the foams are equal to and sometime better than those without the EPS. Such composites are cheaper than polyurethane foam and can have industrial application.

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