

# The effect of ground coffee on the mechanical and application properties of rigid polyurethane-polyisocyanurate foams (*Rapid communication*)

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**Abstract:** Rigid polyurethane-polyisocyanurate (PUR-PIR) foams were obtained based on the commercial polyol, the newly synthesized polyol (Pn), and with addition of stabilizer (2.5–15 wt %) in the form of ground coffee. The produced foam materials with the filler were characterized by lowered brittleness and compressive strength, and increased absorptivity and impregnability in comparison to the reference foam. The foam density increased slightly as well (it ranges between 46 kg/m<sup>3</sup> and 54 kg/m<sup>3</sup>). The addition of the coffee significantly affected the foam processing parameters. The structure of the foam material was also slightly modified. The presence of the modifier caused an increase in the softening temperature by approx. 20 °C (from 156.7 °C for F0 reference foam to 173.0 °C for F6 foam, containing 15 wt % of coffee). The aging parameters (changes in linear dimensions, volume and mass) of foams with coffee were not affected in a significant way and their values were very close to the values of F0 foam.

**Keywords:** PUR-PIR foam, fragility, ground coffee.

## Wpływ dodatku kawy mielonej na właściwości mechaniczne i użytkowe sztywnych pianek poliuretanowo-poliizocyjanurowych

**Streszczenie:** Sztywne pianki poliuretanowo-poliizocyjanurowe (PUR-PIR) zsyntetyzowano z poliolu handlowego, nowo opracowanego poliolu (Pn) i stabilizatora (2,5–15,0 % mas.) w postaci kawy mielonej. Otrzymane tworzywa piankowe charakteryzowały się mniejszą kruchością i wytrzymałością na ściskanie oraz większą chłonnością wody i nasiąkliwością niż pianka referencyjna. W niedużym stopniu zmieniła się też struktura pianek. Dodatek kawy wpłynął na niewielkie zwiększenie ich gęstości (46–54 kg/m<sup>3</sup>), znaczne skrócenie czasów przetwórczych, a także wzrost o 20 °C temperatury mięknięcia gotowego tworzywa piankowego (z 156,7 °C pianki referencyjnej F0 do 173,0 °C pianki F6, zawierającej 15 % mas. kawy). Poddane starzeniu pianki PUR-PIR napełnione kawą mieloną tylko nieznacznie zmieniły swoje wymiary liniowe, objętość i masę.

**Słowa kluczowe:** pianki PUR-PIR, kruchość, kawa mielona.

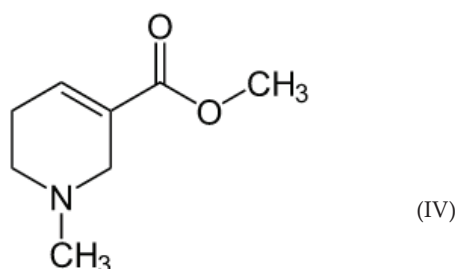
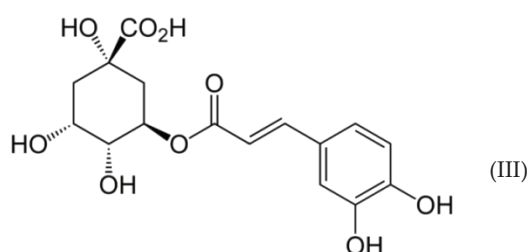
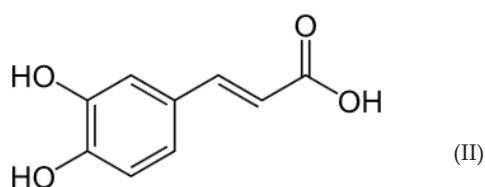
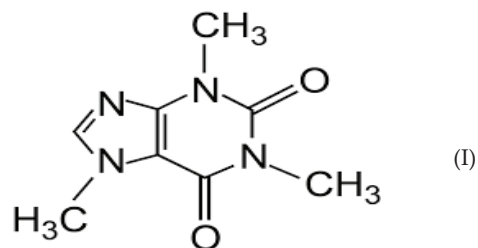
Stabilizers are the chemical compounds which are introduced to materials to stop or slow down the aging processes during material processing and utilization, and at the same time to increase its durability. The most important stabilizer groups used in plastic material technologies are antioxidants, antiozonants, photostabilizers, heat stabilizers, and metal deactivators. Antioxidants are used to stop the aging of polymer due to temperature, light and moisture influence [1]. The additives most commonly used in those technologies are phenol antioxidants, which contain sulfur that provides resistance to thermal oxidation degradation. Researchers often use peroxides

whose decomposition products are the initiators of the polymer crosslinking process. As a result of this process the polymer loses its thermoplastic characteristics and can be used in a higher temperature than the original material [2].

Coffee, which contains caffeine, is a well-known antioxidant. Caffeine (Lat. *caffeinum*), also known as theanine (from tea), guaranine (from guarana), mateine (from yerba mate), is an organic chemical compound, a purine alkaloid, present in floral raw materials [Formula (I)]. It can also be obtained synthetically. It was discovered (isolated in its pure form) by German chemist Friedrich Ferdinand Runge in 1819 [3–6].

Currently the main source of caffeine is the decaffeination of coffee – the extraction of caffeine from coffee seeds [7, 8]. Roasted coffee contains caffeic acid [Formu-

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la (II)] and non-roasted coffee contains chlorogenic acid [Formula (III)].

There are various coffee extracts, for example *Coffea Arabica* Seed Extract. Beside caffeine, it contains also theobromine, theophylline, caffeic acid, chlorogenic acid, and trigonelline – Formula (IV) [9–11]. The influence of coffee smoking on its antioxidant properties was studied by Perrone *et al.* [12] and Small *et al.* [13]. Additionally, it was determined that coffee contains large amount of carbohydrates (*e.g.*, polysaccharides, sucrose), nitrogen compounds (*e.g.*, proteins, caffeine), lipids (coffee oil, diterpenes), and mineral substances (*e.g.*, potassium compounds) [14]. The waste obtained during ground coffee production comprises 50 % of the input [15]. The known methods of its utilization are composting, gardening, bioenergy production or as subsoil for mushroom cultivation. Newer methods include production of biodiesel, sugar sources, precursors of active charcoal or sorbents for the removal of metal ions [16].

The aim of our researches was to examine the effect of ground coffee on the properties of rigid polyurethane-polyisocyanurate (PUR-PIR) foams obtained with the help of new polyol (Pn). The purpose of the research was to use ground coffee for rigid PUR-PIR foams as a stabilizer for aging parameters (change in mass, dimensions and volume) and heat properties (softening temperature). Ground coffee was first used as a stabilizer for acrylonitrile-butadiene-styrene (ABS) by Rytlewski [17], but coffee wasn't used as an filler for polyurethane until now. This modifier was introduced based on its availability. This kind of research can help obtain new type of polyurethane foam with new and broader applications, *e.g.*, in construction (as insulation) and packaging industries. This paper presents the results of investigations on the effects of amount of ground coffee on mechanical, heat, aging and application properties of PUR-PIR foams. Dimensional, volume and mass stability of the foams and the abovementioned heat parameter are very important. Further researches on using coffee in foams will describe its effect on thermal and combustible properties of PUR foams.

## EXPERIMENTAL PART

### Materials

Polyester with trade name Rokopol RF-551 [polyoxypropylenediol with hydroxyl number 420 mg KOH/g, molecular weight 660, product of Chemical Plants PCC Rokita S.A. in Brzeg Dolny] was used to prepare rigid PUR-PIR foams [18, 19]. As a crossing compound the technical polymeric diisocyanate Ongromat 30-20 (BorsodChem, Hungary) was used, whose main component is 4,4'-diphenyl-methane-diisocyanate (MDI). Density of Ongromat 30-20 at temperature of 25 °C was 1.23 g/cm<sup>3</sup>, viscosity was 200 mPa · s, and content of NCO groups 31.0 %. Polyether and diisocyanate were characterized according to standards: ASTM D 2849-69 and ASTM D 1638–70. Catalyst in the process of foam preparation was anhydrous potassium acetate (POCh Gliwice) applied in the form of 33 % solution in diethylene glycol (Catalyst 12) and DABCO 33LV (diazabicyclooctane in triethylendiamine, Hüls, Germany) used as 33 % solution in diethylene glycol. Stabilizer of foam structure was poly(oxyalkylene siloxane) surface-active agent Tegostab 8460 (Witco, Sweden). Carbon dioxide formed in reaction of water with isocyanate groups acted as blowing agent. Moreover, liquid flame retardant tris-(2-chloropropyl) phosphate – Roflam P (Albright and Wilson, Great Britain) was introduced into foam composition. Lewis acid – tetraisopropyl titanate was applied as catalyst in synthesis of new polyol Pn. Its trade name is Tyzor® TPT (Du Pont). It is colorless, yellowish liquid with freezing temperature of 19 °C, highly sensitive to moisture. Beside the industry polyol Rokopol, the new compound (polyol Pn, straw-yellow liquid) was used as a polyol. It was obta-

**Table 1. Characteristics of Pn polyol**

Hydroxyl number mg KOH/g	Acid number mg KOH/g	Content of water %	Density kg/m <sup>3</sup>	Viscosity mPa · s	pH	Color	Molecular weight	Content of C %	Content of H %
572	71	3.35	904	3972	4.2	Straw	204	54.26 ± 0.28	10.72 ± 0.31

ined at the Department of Chemistry and Polyurethanes' Technology at UKW in Bydgoszcz. For synthesis of polyol Pn 2-hydroxypropane-1,2,3-tricarboxylic (citric) acid (Brenntag Poland LLC company in Kędzierzyn Koźle) and pentane-1,5-diol (POCh Gliwice) were used. Pn properties are shown in Table 1. Ground coffee (Cafe Sati Franc Et Intense) was added as a filler in amount of 2.5 wt % (F1 foam), 5.0 wt % (F2 foam), 7.5 wt % (F3 foam), 10.0 wt % (F4 foam), 12.5 wt % (F5 foam), and 15.0 wt % (F6 foam).

### Preparation of rigid PUR-PIR foams

The foam was prepared in a laboratory scale by one-stage method from the two-component system at the equivalent relation of –NCO to –OH groups equal 3 : 1. The component A was obtained by the precise mixing (1800 rpm, 10 s) of the suitable amounts of Rokopol RF-551 [0.7 R (R – chemical equivalent), 46.8 g, in case of F0 reference foam it was 1 R, 66.8 g], polyol Pn (0.3 R, for F0 foam 0 R), Catalyst 12 (2.5 wt %, 8.0 g), catalyst DABCO (3.2 R, 1 g), flame retardant Antiblaze TMCP (15 wt %, 47.6 g), surface-active agent Tegostab 8460 (1.7 R, 5.4 g) and porophor – distilled water (0.7 R, 3.15 g). Component B, polyisocyanate Ongromat 30-20, was added in amount of 3.7 R (250.7 g). The amount of isocyanate was chosen based on the ratio of isocyanate groups to hydroxyl groups, which for rigid polyurethane-polyisocyanate foams was 3 : 1. The calculated amount of isocyanate was increased by the mass of isocyanate necessary for reaction with water. During the reaction, gas (CO<sub>2</sub>) was produced and it foamed the reactive mixture. The isocyanate group chemical equivalent was calculated according to Equation (1):

$$R_{\text{NCO}} = \frac{4200}{\%_{\text{NCO}}} \quad (1)$$

where: %<sub>NCO</sub> – content of NCO group in polyisocyanate.

The hydroxyl group chemical equivalent was calculated according to Equation (2):

$$R_{\text{OH}} = \frac{56 \cdot 100}{L_{\text{OH}}} \quad (2)$$

where: L<sub>OH</sub> – hydroxyl number of polyol Pn.

Polyol Pn was added in amount of 0.3 R, (14.7 g, for F0 reference foam – 0 R). Components A and B were mixed (1800 rpm, 10 s) at respective volume ratio and poured onto an open rectangular tray of internal dimensions 190 × 190 × 230 mm. The F0 foam contained only petrochemical polyol Rokopol RF-551 (without Pn nor coffee) whereas the series of foams F1–F6 contained ground coffee [from 2.5 to 15 wt %, relatively to the sum of the

masses of polyols (Rokopol RF-551 and polyol Pn) and polyisocyanate].

### Methods of testing

After synthesis the obtained product polyol Pn was cooled and analyzed. The following parameters were determined: hydroxyl number (PN-93/C-89052.03), viscosity (PN-86/C-98082.04, Hoeppler viscometer), density (PN-92/C-04504), water content (PN-81/C-04959, by Carl Fisher's method), pH (microcomputer pH-meter CP-551).

During foam preparation, the processing times were measured: start, rise and gel times. The obtained foams, after removing them from the mold, were thermostated for 4 h at 120 °C. Then, they were seasoned for 48 h at 20 ± 4 °C, cut into pieces and basic properties were determined according to valid standards (Table 2): aging parameters (change in linear dimensions, volume and mass of foams), apparent density, structure, water absorbing (absorptivity) and impregnability, compressive strength, brittleness, softening temperature, FT-IR spectra were analyzed as well [18, 20].

**Table 2. PUR-PIR foam test methodology**

Parameter	Standard, sample size, formula
Aging tests	Thermostating 48 h in dryer with 120 °C, Δl, ΔV, Δm
Apparent density	ISO 845-1988, a cube of size 50 mm
Structure	Optical microscope, Axiotech Carl Zeiss, Hal 100 (magnification 10×)
Determination of water absorbing capacity (absorptivity and impregnability)	DIN 53433
Compressive strength	PN-93/C-89071 (ISO 844), cubes: 50 ± 1 mm, strength machine Instron 5544
Brittleness	ASTM C-421-61, 12 cubes of 25 mm
Softening temperature	DIN 53424, cube of sides 20 mm, with Vicat apparatus
FT-IR analysis	Nicolet iS10 FTIR spectrophotometer by Thermo Scientific, spectral range of 7800–350 cm <sup>-1</sup> and maximum resolution capability < 0.4 cm <sup>-1</sup> , with a deuterated triglycine sulfate (DTGS) detector

## RESULTS AND DISCUSSION

The newly synthesized polyol Pn (the result of condensation reaction of citric acid with pentane-1,5-diol) was

**Table 3.** Processing, aging and other selected parameters of foams ( $\Delta l$  – change of linear dimensions in the direction of foam growth,  $\Delta V$  – change of foam volume,  $\Delta m$  – change of foam mass)

Foam	Start time, s	Rise time, s	Gel time, s	Density, kg/m <sup>3</sup>	$\Delta l$ , %	$\Delta V$ , %	$\Delta m$ , %	Compressive strength, kPa	Impregnability, %	Absorptivity, %
F0	20	148	230	46.73	-2.40	-3.28	6.01	377.0	24.3	0.36
F1	20	104	156	47.22	-2.33	-3.02	7.06	239.4	55.2	2.40
F2	21	91	109	48.51	-3.03	-4.30	9.90	198.1	85.1	7.53
F3	19	77	88	48.92	-2.14	-4.32	8.07	179.2	112.4	21.22
F4	18	56	71	49.31	-2.09	-4.47	7.96	162.1	187.5	38.70
F5	17	54	67	51.11	-1.45	-4.00	6.30	146.4	190.1	40.12
F6	16	52	62	54.57	0.98	-3.86	4.45	133.7	194.0	41.00

used for foam synthesis. In article of Liszkowska *et al.* [21], a process of obtaining similar polyol was described (with  $L_{OH} = 436$  mg KOH/g), using the same components, but the content of catalyst was 0.04 wt % and a polyol with 0.1 wt % Tyzor® TPT was used. Due to those conditions preparation time decreased from 8.5 to 4.5 h and the reaction temperature was lowered from 161 °C to 140–160 °C. Also, the researchers obtained 41 g more of the final product, which characteristics is presented in Table 1.

As the amount of ground coffee in the foam increased, the processing times decreased (Table 3). Roasted coffee, which contains caffeic acid and other compounds [9–11] was used. Due to the increase of temperature in foam core, the caffeic acid and chlorogenic acid (which are carboxyl acids) were probably released during the foam preparation. It acted as a catalyst, shortened the processing parameters by 4 s (start time), threefold (rise time) and almost fourfold (gel time).

The addition of ground coffee caused a slight linear increase in the apparent density of the foams (Table 3), which was in the range of 46–54 kg/m<sup>3</sup>. This means that the addition of ground coffee does not affect the apparent density in a significant manner.

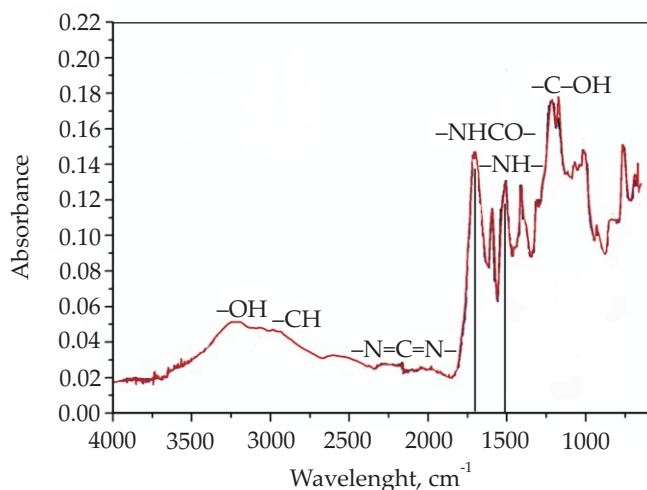
The aging processes of the foam (changes in linear dimensions, volume and mass under temperature, Table 3) were examined as well. The coffee slowed down the aging processes of the polymers [1–6, 17] and ensured

dispersibility with composites with polyolefins [22]. For the PUR foams, the aging is characterized by the stability of the dimensions, volume and mass. In rigid polyurethane foams, aging is very important. It affects the mechanical properties and durability of cells in the foam material. The selection of appropriate components, *e.g.*, polyols and polyisocyanates, can slightly affect the thermal stability of foam products. First 24 to 72 h of foam aging are the most crucial, the most characteristic volume changes occur during that time [22]. The examined aging parameters of foams (changes in volume  $\Delta V$ , linear dimension  $\Delta l$ , and mass  $\Delta m$ ) were barely affected in comparison to F0 foam.

IR analysis of foams (Fig. 1) shows the presence of urethane (1736 cm<sup>-1</sup>) and isocyanurate bonds (2969–2915 cm<sup>-1</sup>). Moreover, hydroxyl group –OH is observed within the range from 3136 cm<sup>-1</sup> to 3360 cm<sup>-1</sup>.

It was observed, that the foam, which did not contain coffee, was characterized by regular pore dimensions, without any structural defects (Fig. 2). The microscopic images of F0 foam show that the cell shape was spherical when the foams were cut perpendicularly to the growth direction. Cells of the same foam cut parallel to the growth direction had an almost spherical shape, only slightly elongated. The microscopic images of foams with coffee cut perpendicularly to the growth direction exhibit a slightly elliptic shape of the pores (Figs. 3 and 4). The coffee was deposited on the cell walls in the foam structure and did not increase the foam strength (Table 3). The structure of the foams modified with ground coffee (F1–F6) contained few irregular pores with slight defects in the structure. They were caused by the presence of coffee particles in the foam.

The values of the softening temperature are presented in Fig. 5. An increase in the softening temperature could be observed in foams with ground coffee (*e.g.*, 173.0 °C in F6 foam) in comparison to F0 foam (157.6 °C). The amount of coffee positively affected the softening temperature. A linear dependency was observed between the coffee amount and the softening temperature. On the contrary, the brittleness decreased with the ground coffee content (Fig. 5): the highest value (over 41.1 %) was obtained for F0 foam and the lowest (3.7 %) – for F2 foam (containing 5 wt % of coffee).



**Fig. 1.** FT-IR spectra of F0 PUR-PIR foam

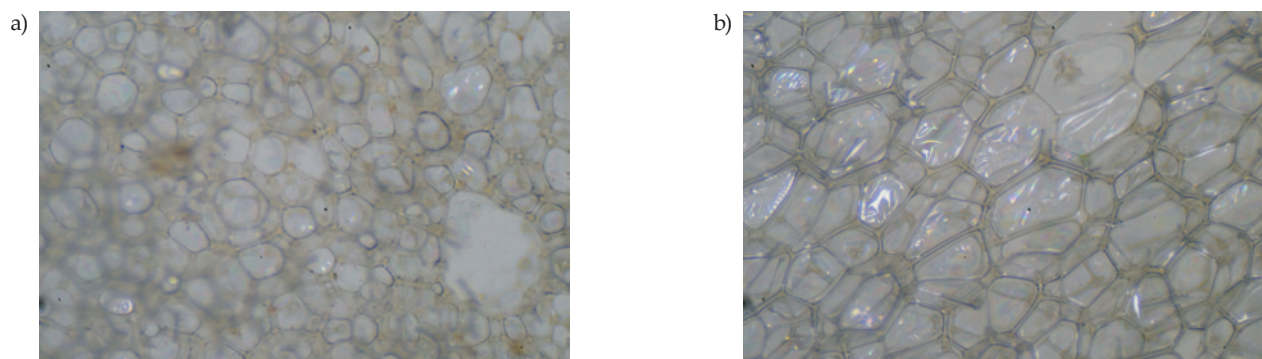


Fig. 2. Microscopic images of F0 foam cross-sections: a) perpendicular, b) parallel to the growth direction

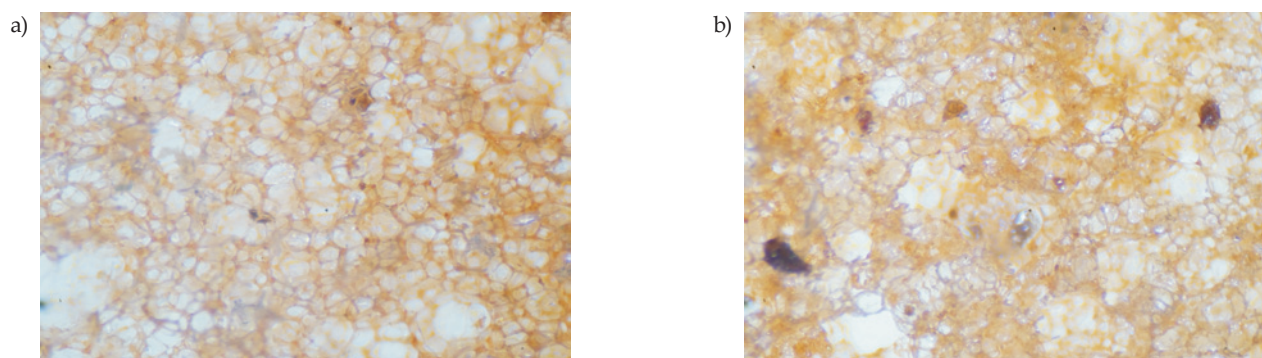


Fig. 3. Microscopic images of cross-sections of foams containing ground coffee, perpendicular to the growth direction: a) foam with 5 wt % coffee content, b) foam with 10 wt % coffee content

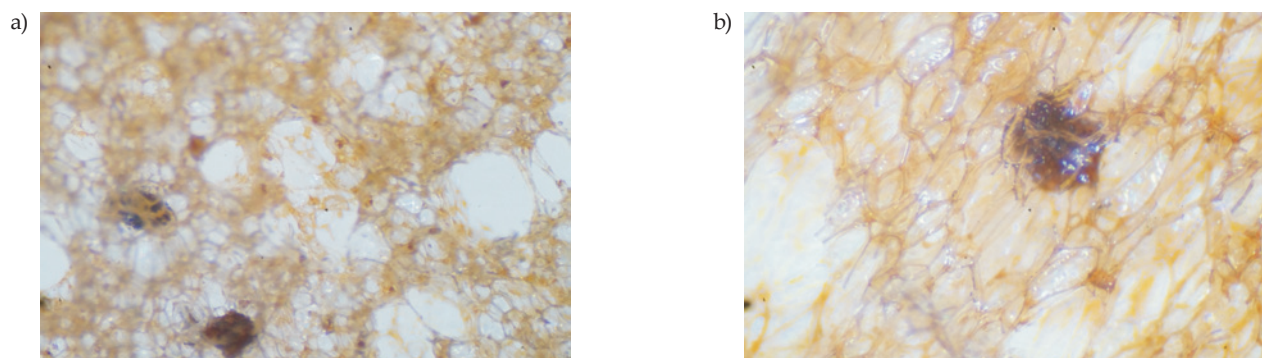


Fig. 4. Microscopic images of cross-sections of foams containing ground coffee, parallel to the growth direction: a) foam with 5 wt % coffee content, b) foam with 10 wt % coffee content

The compressive strength of PUR-PIR foams (Table 3) changed from 377.0 to 133.7 kPa. Based on past studies [23, 24], it can be determined that the decrease of compressive strength decrease was due to the presence of new polyol, not the addition of ground coffee. The results of absorptivity and impregnability for all examined foams are presented in Table 3. Both, the impregnability and absorptivity were the worst for F0 foam (24.3 % and 0.36 %, respectively). For F6 foam the absorption values are 194 % (impregnability) and 41 % (absorptivity). The absorptivity and impregnability of foams were affected by the coffee filler, which had high values of those properties by itself. It is also confirmed by the fact that the coffee filler was embedded in the foam structure. The ab-

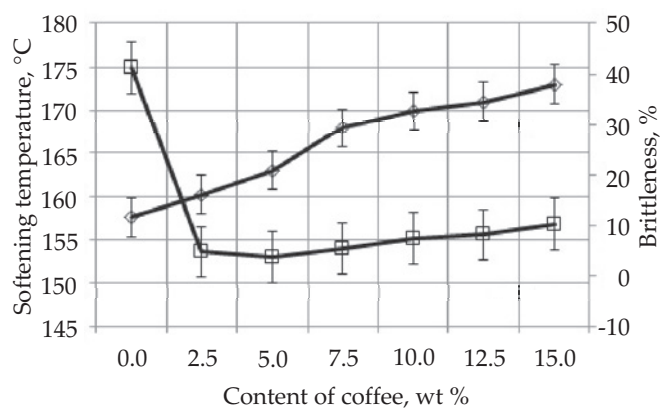


Fig. 5. Dependency of softening temperature and brittleness on the coffee content in PUR-PIR foams

sorption of water indicates that it is easy to penetrate into the degradable part of the structure (coffee), which leads to faster disintegration of this structure. Foam F0 did not have coffee in its composition, hence the migration of water molecules was difficult. Therefore, it had the lowest absorption capacity.

### CONCLUSIONS

The production and researches on the foams containing coffee helped obtain a product with improved heat properties. The addition of this filler increased the softening temperature of the foam materials by 20 °C, from 157.6 °C (F0 foam) to 173.0 °C (foam with 15 wt % of coffee). The brittleness of the foams containing coffee decreased from 41.4 % (F0 reference foam) to 10.2 % (F6 foam with 15 % coffee concentration), and the F2 foam had the lowest brittleness (3.7 %). The absorptivity and impregnability increased in a linear manner depending on the coffee concentration in the foam. In the previously published articles [1–6, 17], whenever coffee was used as a filler, it slowed down the aging processes of the polymers. The examined aging parameters of the obtained foams (changes in linear dimensions, volume and mass) were almost unaffected in comparison to the values of F0 foam. Keeping those parameters at a steady level is important for the purpose of applying those foams in various industries, for example in the construction or as a shock absorbers in packaging.

Further studies on foams containing ground coffee will be focused on the flammability of the foams and their thermal properties. The results will be described in details in the next publication.

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