## Influence of the Use of Recycled Polyols Obtained by Glycolysis on the Preparation and Physical Properties of Flexible Polyurethane

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**ABSTRACT:** Polyurethanes (PUs) represent one of the most important groups of plastics, and so the increasing quantity of wastes makes their recycling an urgent task. The general purpose of PU chemical recycling is to recover constituent polyol, a valuable raw material. Among the suitable processes, glycolysis in two phases allows better quality products. The objective of this work is the evaluation of the option to apply the recovered polyols to obtain PU with identical characteristics to the starting raw material, and so several foaming tests were carried out according to the evaluation method employed in free expansion foaming of conventional flexible slabstock foams. To achieve this objective, a formulation recipe for flexible

foams was selected, in which the raw polyol was totally or partially replaced for recovered polyol. The foaming formulations were modified because of the different amount of active hydrogens in the recovered polyols and the virgin polyol. Amounts up to 50% could be applied without relevant changes in rising profiles and the physical properties of the foams. The foams were characterized, and according to its appropriate characteristics they can be employed in the same applications where a commercial one made with raw polyol is used. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 617–626, 2008

Key words: polyol; polyurethane; foam; recovery

### **INTRODUCTION**

One of the best alternatives for the recycling of polyurethane (PU) foams includes chemical treatment to convert the PU back to its starting raw materials, especially polyols. Glycolysis has been described as a suitable procedure to break down the PU chain.<sup>1-5</sup> Most of the glycolysis processes proposed in literature produce a liquid mixture of products containing hydroxyl active groups, which can be used only in blending with raw materials. Nevertheless, better quality products can be achieved from flexible PU foams using a two-phase glycolysis, enabled by the higher molecular weight of polyols used in this kind of PU. By means of an excess of glycolysis agent, much larger than the stoichiometric quantity, the reaction product splits into two phases, where the upper layer is mainly formed by the recovered polyol from the PU and the bottom layer by the excess of glycolysis agent.<sup>6</sup>

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As described in previous works,<sup>7–9</sup> the authors have been working in the design of an integrated process for the recovery of polyols from flexible PU foams by means of two-phase glycolysis to obtain quality recovered products. The choice of diethylene glycol (DEG) as glycolysis agent and potassium octoate as catalyst allows to obtain high-quality polyol. Further washing and drying of the polyol phase allows to obtain recovered polyol with high purity and with chemical properties quite similar to that of fresh commercial polyol.<sup>10</sup> But it is well known that the main test that one polyol has to pass through to confirm its applicability is the foaming test.

It is usual that only traces of contaminants in a given polyol change dramatically the properties of the PU foam prepared with it. For this reason the foaming capacity of the recovered polyols has to be confirmed to ensure their reuse.

PU foams are made by the reaction of polyols and isocyanates in the presence of other agents such as amines, tin catalyst, blowing agents, surfactants, and other additives. During this process, there is multitude of competing reactions that form different species, which in turn gives the foam different physical properties. Key reactions are the blowing reactions that generate the gas responsible for some cell features, and the gelling reaction that contributes to the formation of the three-dimensional network and structure. The balance of these two reactions

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determines most of the physical properties of PU foams. At first stages, the blowing agent produces a dissolved gas in the liquid phase up to the saturation limit. Then, the gas escapes from the polymerization mix forming bubbles. When the bubble begins to be formed the gas sphere is surrounded by a liquid layer in polymerization. As the gas is produced the number of bubbles and their size increase and the gas diffuses from some bubbles to others in such a way that the volume of the foam grows and the liquid layer thickness decreases. The bubbles lose their spherical form as the liquid layer becomes thinner, adopting a definitive three-dimensional structure surrounded by plane membranes of polymer.<sup>11</sup> Any insignificant change in this complicated sequence of interdependent stages can fail the foaming process.

These cells may be closed, retaining the gas that has formed in them, like in rigid foams, or open, with several or all faces broken, like in flexible foams. The gas that leads to the formation of the cells is obtained by the addition of foaming agents that are classified in function of its nature.

Water can be used as a blowing agent. The formation of gas is the result of a chemical reaction of isocyanate with water, leading to an unstable carbamate that breaks up to carbon dioxide. In a similar way the gas can be generated by means of thermal decomposition, as in the case of the azo compounds, which produce nitrogen.

The most employed surfactant agents in flexible foams are silicone-based compounds such as polydimethylsiloxanes with no pendant side chains, or complex polyoxyalkylene polysiloxane copolymers with varying levels of pendant side chains. Organometallic compounds such as tin octoate or dibutyltin dilaurate are usually used as polymerization catalysts. The catalysts that lead to production of gas are constituted by a blend of two amines. One of these amines has low molecular weight and high activity and catalyzes the reaction between water and isocyanate; this amine is eliminated quickly from the foam because of its volatility. The other amine used has high molecular weight and less activity and remains in the system ensuring a suitable curing. Examples of this catalyst are N,N-dimethylcyclohexylamine, diaminobicyclooctane, and N,N-dimethylaminoethanol.<sup>11–13</sup>

Despite the great industrial significance of PU foams, there is lack of information and experimental data in relationship to quantitative aspects of the foam formulation and their influence on the characteristics of the polymeric three-dimensional network during foaming. Several experimental difficulties exist, such as numerous chemical reactions, the volume and viscosity changes, or the heterogeneity of the mixture that implies uncertainties related to the analysis methods. Another problem lies on the

experiment scale. There is great variability among the processes investigated at laboratory scale and the manufacturing of industrial slabstocks, where the foam dimensions are around 2 m wide by 1 m high.<sup>12</sup> To develop new products for its later application, foaming free expansion rehearsals at laboratory scale are usually carried out, characterizing the foaming process by means of temperature profiles inside the foam with a infrared camera or the foam growth with a ultrasounds distance meter.<sup>14–16</sup> These profiles and the physical properties of the final foam determine their subsequent applicability.

The objective of this work is the evaluation of the option to apply the recovered polyols to obtain PU foams with identical characteristics to the starting raw material. Several foaming tests have been carried out according to the evaluation method employed in free expansion foaming of conventional flexible slabstock foams to verify their reusability. The potential level of raw polyol replacement with recovered polyol and its influence on the physical properties of the foam prepared with the recycled polyol has been studied.

### **EXPERIMENTAL**

## Materials

The recovered polyether polyol for foaming assays was obtained as upper phase after glycolysis of industrial samples of flexible PU foam. The PU was based on polyether polyol [poly(propylene oxide)block (ethylene oxide),  $M_w \sim 3500$ , functionality with respect to OH groups of three] and toluene diisocyanate. Silicones, amines, and catalyst composition and providers are not given to preserve certain confidentialness on the recipes of the supporting company. The scrap foam was reacted within DEG (PS, from Panreac, Spain) using potassium octoate (NUSA, Spain) as catalyst. The glycolysis reaction was carried out in a 2-dm<sup>3</sup> flask equipped with stirrer, heating control system, and refluxing condenser under nitrogen atmosphere to avoid oxidation. After reaction, polyol phase was purified by liquid extraction with an aqueous hydrochloric solution and dried.<sup>10</sup> The operational conditions of each step of polyol recovery are summarized in Table I. The method has been previously described in detail in previous articles.<sup>7,8,10</sup> Polyol properties are shown in Table II.

### Foaming assays

The foaming experiments were carried out according to the evaluation method in free expansion foaming of conventional slabstock foams. A formulation recipe for flexible foams was employed based on a

	Operation	in contantions for Obtaining	the factovered i oryon	
Glycolysis reaction				
PU Foam (g)	Glycol DEG (g)	Catalyst K octoate (g)	Temperature (°C)	Reaction time (min)
300	440	10	189	150
		Extraction		
Solvent pH	Mass ratio solve	ent : Polyol phase	Temperature (°C)	Phase separation
4	1	:1	60	Decantation
		Dehydration		
Pressure (mmHg)		Temperature (°C)		Time (min)
10		110		240

 TABLE I

 Operational Conditions for Obtaining the Recovered Polyol

polyol of  $M_w \sim 3500$  in which the virgin polyol was totally or partially replaced for recovered polyol. The foaming formulations were modified because of the different amounts of active hydrogen between the recovered polyols and the virgin polyol.

The growth tests were carried out using a wood box with dimensions of 20 cm long by 20 cm wide by 13 cm high where the growth of the foams was monitored by means of SQS-01, a system for the qualification of PU foaming by means of an ultrasound sensor supplied by Sensing S.L. (Madrid, Spain), and the times corresponding to 50% of the maximum growth were determined (T50%), as well as to 75% (T75%) and the time for the maximum growth (TMG). This characteristic time, TMG, corresponds to the time lapses between the mixture of the products and the moment in which the maximum growth of the foam is reached. This is determined as 99.5% of the maximum height measured.

The measurements of the mechanical properties of the foams have been carried out according to the standard methods and using the appropriate testing device in each case. These methods are explained in the corresponding section of this work for a better understanding of the measure objective and its meaning.

## Characterization

The characterization was carried out at a temperature of 23–24°C and a relative humidity of 20–25%. The density was measured according to the ISO 845 standard. The tear resistance was determined according to ISO 8067 standard. The resilience was measured according to the ISO 8307 standard. The remaining deformation 50% was measured according to the ISO 1856/A standard. The tensile strength and the elongation were measured according to ISO 1798 standard. The thickness variation and the compression strength after the dynamic fatigue are determined according to the standard NF-T56-114/85. The compression strength is measured according to the standard ISO 3386.

## **RESULTS AND DISCUSSION**

# Experiments with upper phase without further polyol purification

The first series of foaming experiments were carried out with nonpurified recovered polyol (SPR100-I) to verify if an exhaustive polyol cleaning is necessary after reaction before foaming. In Table III, the different recipes with varying amounts of catalyst and isocyanate, together with the characteristic foaming times are shown. A nonappropriate growth profile was obtained with upper phase as obtained from the glycolysis process and can be observed in Figure 1. This foaming growing profile shows a high growth speed at the beginning so the value observed for T50% (time to reach the 50% of the final height) is very low. After the curing process the foam suffered an important contraction in its structure. The contraction and dimensional deformation of the foam is due to the cells are not completely opened or present a low permeability, so that the cells retain the hot

 TABLE II

 Properties of the Purified Polyol After Water Treatment

Viscosity	Hydroxyl number	Acidity	Water	Amine content	Alkaline
(cp)	(mg KOH/g)	(mg KOH/g)	content (%)	(mg KOH/g)	content (ppm)
591	63	0.011	0.103	6.23	<1

	TABLE III		
Foaming Experiments With Upper Phase Without Further			
	SPR100 (I)	SPR100 (II)	
Raw polyol 3500 $M_w$	_	_	
Recovered polyol	100	100	
Hydroxyl number polyol mixture	120 <sup>a</sup>	120 <sup>a</sup>	
Water	4.60	4.60	
Amine A	0.10	0.10	
Amine B	0.05	0.05	
Silicone	1.40	1.40	

**Polyol Purification** 

0.20

68.48

105

68

83

153

contraction

Noticeable

0.10

68.48

105

90

100

124

cavernous crack

Collapse and

Recipes are given on the basis of 100 arbitrary mass units of polyol. Theoretical value.

gas that formed them. When the gas is cooled down after the curing process, the cells volume reduces with the consequent contraction of the cell.

Sn Octoate

T50 % (s)

T75 % (s)

TDI (80 : 20)

Isocyanate Index

TMG 99.5% (s)

Observation

The formation of closed cells and their subsequent contraction can be due to the presence of a high quantity of gelation catalyst. Stannous octoate makes increase the polymerization rate, therefore increasing the viscosity of the foaming media, leading to thick and closed cellular walls. It can be due to either an excess of isocyanate (the isocyanate index applied is too higher) or the incorrect proportion of the amine catalyst and gelation catalyst. The amount of gelation catalyst, stannous octoate (SPR100-II), and the isocyanate index (SPR100-III) were modified in the formulation, keeping constant the amount of isocyanate isomers and the rest of conditions.

As a result of this modification in the recipes, the reaction mixture was strongly catalyzed to the reaction of gas formation, decreasing the cream time. The foam SPR100-II suffered a collapse because of a disproportionate ratio of gelation catalyst and amine catalyst (Fig. 2). On the other hand, the foaming experiment carried out with a decrease of the isocyanate index and keeping the gelation catalyst content constant (SPR100-III) did not improve the foaming process either. In both cases the reaction mix began to grow before the polymeric network acquired enough consistency. As a consequence of this strong growth the bubbles of gas formed escape from the reaction mix causing the collapse of the foam. This collapse produces a cavernous crack inside the block and an irregular cellular structure. The rise profiles (Fig. 1) also show the collapse and sinking of the foam with a strong decrease after reaching a maximum height.

The modification of the quantity of stannous octoate and isocyanate index did not solve the problem of the foam collapse. The proposed recipe

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adjustments have not get the objective of a proper foaming probably because not only DEG that modifies the hydroxyl number but also the byproducts containing amine groups that can acts as catalyst modifies the unstable equilibrium of the foaming process. The washing of the upper phase with a slight acid pH and further dehydration provides a polyol with an improved quality<sup>10</sup> and so experiments with clean recycled polyol were carried out.

SPR100 (III)

100  $120^{a}$ 4.600.100.05

1.40

0.20

65.21

100

84

94

114

cavernous crack

Collapse and

## Foaming assays using polyol purified by extraction liquid-liquid

The polyol was replaced completely by purified recovered polyol in the former formulation and two different experiments were carried out. In the first experiment the foaming recipe was maintained assuming that the hydroxyl number of the recovered polyol is the same that the hydroxyl number of the



Figure 1 Rising profiles of nonpurified upper phase in free expansion foaming test. [Color figure can be viewed in the online issue, which is available at www.interscience. wiley.com.]



Stage 1

Stage 2



Stage 3

**Figure 2** Process of growth and collapse of the foam in exp SPR100-II. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

standard polyol (A0-R100 (I)). In the other the formulation was recalculated (Table IV). The hydroxyl number used for the adjustment of the recipe was the hydroxyl number measured for the recovered polyol, according to the actual value obtained by analysis (A0-R100 (II)).

Figure 3 shows the rising profiles of foams formulated only with recovered polyol and blends recovered with commercial polyol, and the curve obtained with the standard polyol. The direct replacement of the polyol (A0-R100 (I)) in the formulation without considering the real hydroxyl number of the recovered polyol gave place to a quick foam formation (Fig. 3), in which the cream time was reached during the mixing, and after the curing process the foam contraction took place. Since a lower polyol hydroxyl number

TABLE IV				
Foaming Experiments	With Recovered Polyol Purified I	y Extraction Liquid–Liquid		

	A0-R100 (I)	A0-R100 (II)	A50-R50	A75-R25
Raw polyol 3500 $M_w$	_	_	50	75
Recovered polyol	100	100	50	25
Hydroxyl number polyol mixture	48 <sup>a</sup>	63	58	50.5
Water	2.92	4.60	4.60	4.60
Amine A	0.10	0.10	0.10	0.10
Amine B	0.05	0.05	0.05	0.05
Silicone	1.40	1.40	1.40	1.40
Sn Octoate	0.20	0.20	0.20	0.20
TDI (80 : 20)	54.58	57.02	56.21	54.99
Isocyanate index	105	105	105	105
T50 % (s)	-	83	98	103
T75 % (s)	_	98	110	120
TMG 99.5% (s)	-	208	174	179
Observation	Noticeable contraction; $T_{\text{cream}}$ reached during the reactive mixing	Contraction	OK	OK

Recipes are given on the basis of 100 arbitrary mass units of polyol.

<sup>a</sup> Theoretical value.



**Figure 3** Rising profiles of the mixtures of purified recovered polyol and standard polyol in free expansion foaming test. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

was considered, the isocyanate added to the polyol was not enough to provide a stable structure to the PU foam. However, when the real hydroxyl number was considered to adjust the formulation (A0-R100 (II)) a similar growth profile was obtained in comparison to the growth profiles obtained for standard foams, showing better dimensional stability than the foam A0-R100 (I). The main difference with the standard is that a smaller and quicker growth was observed.

After that, there blends were prepared with both polyols (recovered and commercial) in different proportions and the formulation was modified taking into account the real hydroxyl number of the blend. The foams synthesized with blends showed quite similar rising profiles to those of the standard polyol.

The physical appearances of the foams prepared are shown in Figure 4. It can be observed that all the

foams exhibits regular structure with not internal defects, only pinholes or bubbles of air occluded in the foam, probably because of the incorporation of this bubbles in the mixture reaction during the mixing process. Depending on the quantity of recovered polyol employed, a slight increment of the color of the foam is observed (darkness in b/w) changing from white, when only virgin polyol is used, (A100-R0) to cream color, when 100% of the same is prepared with recovered polyol (A0-R100). This slight coloration is due to the intense color of the recovered polyol. The polyols are compounds sensitive to the light and the environment, and therefore, easily oxidized. Even so, the influence in the final color of the foam is very small and it does not suppose a requirement of strict fulfillment because most of the commercial foams are tinted with different colors depending on its density.

The foams A75-R25 and A50-R50 gave place to homogeneous distributions of cells and good skin formation (external surface of the superior layer of the foam cake) similar to the standard foam. The foam synthesized with 100% of recovered polyol (A0-R100) led to a more compact structure and formation of smaller cells, because of the contraction suffered after the curing.

#### Physical characterization of the foams

To characterize the foams and ensure that can be employed in the same applications than a commercial one made with raw polyol, their mechanical properties as well as the durability have been measured by means of diverse standardized tests.



**Figure 4** The patterns of the foam synthesized with the standard raw polyol (a) and the foam synthesized with different percentages of recovered polyol with a constant isocyanate index: (b) A75-R25, (c) A50-R50, (d) A0-R100. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]



**Figure 5** Dependence of foam density as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).

The density of the foams is defined as the weight per unit of volume of the foam and it is determined in some specific conditions of temperature and relative humidity as is described in the ISO 845 standard. The density of foam prepared with a standard polyol is 23 kg/m<sup>3</sup>. The foams obtained were synthesized with an initial formulation in which the water content was adjusted to maintain the same density of the initial foams. The density values obtained for the aforementioned foams were quite similar to the standard one. For A75-R25 foam a slight decrease of density was observed (Fig. 5). This slight difference is so small that a correction of the density is not required.

The adjustment of the density would suppose a change in the water content and therefore a proportional change in urea domains in the foam and a decrease in the foam flexibility could be attained. Shendijarevic<sup>17</sup> also observed that small percentages of recovered polyol entailed a density decrease, although high percentages of recovered polyol increase the density.

The tear resistance is defined as the force required to initiate or to continue a tear or a laceration of a foam sample subjected to a tensile stress under specified conditions. It was determined according to ISO 8067 standard. Figure 6 shows the values of the tear strength varying the recovered polyol content. The foam prepared with recovered polyol only exhibits lower strength than that from blends or the standard while the foam from the blend 50% shows tear strength similar to the standard.

The resilience is defined as the absorbed energy by a sample of foam when it is subjected to a cyclic compression (absorbed energy) and stress-relaxation (released energy) and it is related to the capacity of the material to recover its original state after these pressures cycles. Resilience of conventional flexible foams takes values among 33-51%. This property was measured according to the ISO 8307 standard. Figure 7 shows the influence of the content in recovered polyol on the resilience. The foam from the 50% blend exhibits an appreciable resilience decrease, probably because of the presence of low molecular weight chains, while the foam obtained with a replacement of 25% of the commercial polyol showed a resilience value even slightly higher than that measured for the foam pattern. Van der Waal<sup>18</sup> observed a decrease of the resilience in conventional flexible slabstock foams when polyol is recovered by two phases of aminolysis, while Hemel et al.<sup>6</sup> and Hicks et al.<sup>19</sup> described an improvement in this property when recovered polyols from high resilience foams are used. The glycolysis process and later purification implies a reduction of unsaturation content for the recovered high molecular weight polyol, and therefore could suppose even an improvement in certain properties like resilience.

The remaining deformation of 50%, according to ISO 1856/A standard, measures the permanent



**Figure 6** Dependence of foam tear resistance as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).



**Figure 7** Dependence of foam resilience as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).

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**Figure 8** Dependence of the remaining foam deformation 50% as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).

changes of the thickness that take place in a test probe under compression of 50% at a specified humidity and temperature conditions, during a certain period of time (22 h), and it is expressed in percent of loss of the initial thickness. The values of residual deformation of the samples can be observed in Figure 8. Increases with the percentage in recovered



**Figure 9** Dependence of foam tensile strength and elongation as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).

polyol used as a consequence of the addition of segments with low molecular weight chains and low functionality dissolved in the recovered polyol, decreasing the flexibility of the structure. This deformation represents the permanent loss of the initial height of the foam because of the partial collapse of the cellular structure. The remaining high deformation percentages would cause premature shape losses in the PU products.

The tensile strength and the elongation were measured according to ISO 1798 standard. The variation of these properties with recovered polyol content is represented in Figure 9. The test determines the tensile strength at which the test probe failure occurs. The elongation is the percentage of the initial length that the test tube reaches in that moment. The tensile strength and the elongation show a similar comportment when the quantity of recovered polyol is modified. The PU foams based on the commercial polyol takes values of 66-150 kPa for tensile strength and 110-260% for elongation. The slight elongation increase observed in blends is due to short ramifications and an increase of the quantity of the covalent bonds that is a direct consequence of a decrease of the functionality of recovered polyol. The effects of introducing recovered polyol to the formulation are



**Figure 10** Dependence of foam compression strength and thickness after dynamic fatigue as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).



**Figure 11** Dependence of foam compression strength as a function of the recovered polyol percentage introduced into the formulation (replacing the raw standard polyol).

a slight decrease in molecular weight and in polyol functionality because of the presence of residual compounds with molecular weight lower than the polyol and also with low functionality (mainly two).

The flexible foams based on polyether polyol present a low vitreous transition temperature  $(-55^{\circ}C)$  in such a way that the foam shows viscoelastic characteristics. So, when the foam is subjected to deformations during short periods it shows elastic behavior, but after long compression periods it begins to flow. The fluency ratio is quantified as the variation of the strength after a period of time being subjected to a constant stress. The dynamic fatigue is produced subjecting the foams to numerous compression cycles (200,000), and after that, the thickness variation and the compression strength are determined, being compared with the values of the initial test probes, according to standard NF-T56-114/85. As it can be seen in Figure 10, only the remaining deformation is affected significantly by the introduction of recovered polyol in the original formulation.

Figure 11 shows the compression strength of the synthesized foams. The compression strength is measured according to the standard ISO 3386/1 and it is related with the foam hardness defined as the necessary load to produce a deformation of the 25, 40, 50, or 65% of the initial test probe thickness when the stress is carried out uniformly on its entire surface. The compression strength decreases when the recovered polyol percentage introduced in the formulation increases. Surprisingly, this decrease is smaller in the case of using 50% of recovered polyol. The compression strength generally depends on the density, and the foam hardness increases when the foam density decreases. This fact is experimentally proven by comparison with foam densities in Figure 5. Both foam density and compression strength present a minimum value for 25% of recovered polyol. As it was previously commented, foam density is a parameter determined by the water quantity introduced as foaming agent. Additionally, the presence



Figure 12 Microphotographs of the PU foam framework for different percentages of recovered polyol, replacing the raw standard polyol.

of water influences notably in the formation of urea domains, conditioning in this way the foam hardness. The same as in the case of other properties, the presence of byproducts with smaller molecular weight and functionality in the reaction mix modifies the final foam hardness, thus increasing the possibility of covalent bond formation by isocyanate and the reticulation of the foam.

The variation of the polymeric chains composition due to the addition of the recovered polyol, including the reticulation degree and the rigid domains, as well as the internal morphology of the synthesized foam are two decisive aspects that condition the final physical properties of the cellular PU. In this way, the microscopic structure of the foam reflects directly the macroscopic physical properties. To understand better these connections, in Figure 12, the microphotographs of the foams synthesized with several replacement are shown.

The cells formed in the foam synthesized with the standard polyol and the foams synthesized with a percentage replacement of virgin polyol 25 and 50% (A100-R0, A75-R25, and A50-R50) show a homogeneous aspect. A slight decrease in the cell size of the foam A50-R50 is observed. The fact that different foams present a similar distribution of cells sizes and shapes is in good agreement with the growth curves that exhibit similar profiles, since the foaming, as explained earlier, is the consequence of two parallel processes, the formation of the polymeric structure and the formation of gas. If one of these processes accelerates regarding the other one because of the addition of different compounds, this fact will be reflected in the morphology of the cellular structure. On the other hand, obtaining some regular and homogeneous cells implies that the mixture of recovered polyol and commercial is stable, avoiding the formation and segregation of domains.

Additionally, the presence of small polymer membranes is observed in certain cells, and the presence of these membranes increases when the addition of recovered polyol increases. For a complete replacement of virgin polyol with recovered polyol (A0-R100), a lot of these closed cells are observed in the three-dimensional network. As it was proven with the foams growth profiles (Fig. 3), the foam contraction is caused by the formation of a continuous membrane closing the cells that retain the gas inside. When the gas is cooled down after the curing process, the cells volume reduces with the consequent contraction of the cell. In Figure 12, the contraction, as well as cells irregularities, and squashed cells can be observed. Furthermore, the presence of closed cells contributes to reduce the elastic capacity of the foam, which is reflected in its resilience and elongation decrease, in addition to other related properties such as the remaining deformation and the fatigue strength.

## CONCLUSIONS

Since the main application of the flexible polyether polyols is the production of flexible foams of PU, the fundamental parameter that allows defining its quality and utility is the capacity of the polyol to produce quality cellular PUs. The foaming tests and the physical properties of the foams have demonstrated that first-quality PU can be produced using blends of recycled and raw polyol. In this way, the introduction of 25% recovered polyol instead of the virgin one does not cause any detriment in the required physical properties similar to a commercial quality PU foam. The incorporation of high percentages of recovered polyol would require modifications in the formulation to keep constant all the physical properties, especially those related with the elastic properties of the foam.

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