

Article

Research into Efficient Technology for Material Recovery of Waste Polyurethane Foams

Lubomír Šooš ^{1,*}, Miloš Matuš ^{1,*}, Stanisław Legutko ² and Jozef Bábics ¹

¹ Faculty of Mechanical Engineering, Institute of Production Engineering and Production Quality, Slovak University of Technology in Bratislava, Námetie Slobody 17, 812 31 Bratislava, Slovakia; jozef.babics@stuba.sk

² Faculty of Mechanical Engineering, Institute of Mechanical Technology, Poznań University of Technology, Piotrowo 3 Street, 60-138 Poznań, Poland; stanislaw.legutko@put.poznan.pl

* Correspondence: lubomir.soos@stuba.sk (L.Š.); milos.matus@stuba.sk (M.M.)

Abstract: The recovery of waste from old vehicles generates different types of waste. Most waste can be recovered with more or less success. Among the wastes that are problematic is foam. It is large in volume and light in weight, and there is currently no highly efficient technology to recover this waste and produce new products. The aim of this paper is to analyse the current situation in the processing, research, development, design and testing of test samples as a basis for the production of a machine to produce new 3D products made from waste foam. The paper begins with an analysis of the amount of plastic waste generated in the automotive industry. It describes the current state of waste management and the possibilities of its use in the production of new products. The core of the paper is the selection of suitable technology and the design and verification of experimental measurement and evaluation of test samples at different temperatures and pressures and with different endurance times.

Keywords: polyurethane foam; material recovery; PUR recycling



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1. Introduction

Polyurethanes' versatility is founded in a broad spectrum of properties, which can be achieved by choosing suitable combinations of starting materials, the key raw materials being polyol and isocyanate. These properties can range from flexible to rigid and from compact to foamed [1].

The worldwide production of polymeric foam materials is growing due to their advantageous properties of light weight, high thermal insulation, good strength, resistance and rigidity. In the year 2022, the market attained a volume of 15.5 million tonnes, experiencing a growth of 6.9% (2019–2022) [2]. Only a small part is recycled, while the majority becomes waste and ends up in landfills or in incinerators [3]. The environmental impact is substantial and derives primarily from the nature and quantity of the materials involved and, therefore, from the resulting consumption of resources [4]. Other aspects, such as high volumes due to the low density of foams, make the management of waste not easy as regards both logistics and delivery to incineration plants [5]. The recycling and recovery of PU foam cover a range of mechanical, physical, chemical and thermo-chemical processes [6,7].

Flexible polyurethane (PUR) foams account for about 33% of total polyurethane production. They are used for the production of seats in the automotive industry, mattresses, furniture, laminating textiles, packaging purposes (impact protection), and the production of insulation and sealing strips. They are also used in the construction industry. The fact

that they are commonly used both in industry and as consumables raises the issue of waste disposal and recycling. There is also the possibility of waste in the production, where it can reach up to 10% of the total foam production. PUR can be used as foams, elastomers, varnishes, adhesives, elastic fibres or artificial leather. In the automotive industry, there is a demand for moulded products made from flexible PUR foams. Their density ranges from $15 \text{ kg}\cdot\text{m}^{-3}$ to $70 \text{ kg}\cdot\text{m}^{-3}$ [8].

Rigid foams are prepared in closed or open moulds. They are mostly used as insulation material in construction and engineering (pipes, refrigerators, cars), but also in aircraft as radar covers. This takes advantage not only of their good insulating capacity but also of the hardness of rigid PUR foams. Their hardness ranges from $10 \text{ kg}\cdot\text{m}^{-3}$ to $600 \text{ kg}\cdot\text{m}^{-3}$ [9,10].

Polyurethane foams are used in automobiles as seat fillers, armrests, roof upholstery, door trim, carpet pads, noise and vibration insulation for engine compartments, etc.

The implemented project is focused on the efficient design of the treatment of PUR waste from old vehicles. As the output is to be an optimised design of the technology of material (or energy) recovery of waste—polyurethane foams—it is necessary to quantify the quantities of this problematic waste from old vehicles in the Slovak Republic. In 2022, 46,354 old vehicles were processed in Slovakia. Based on an average vehicle weight of 1400 kg and a percentage by weight of PUR foam (foam) in the car of 1.75%, it is possible to conclude that 1135 tonnes of this problematic waste were generated from old vehicles in 2022 alone [11].

However, in terms of designing and optimising a technology suitable for the treatment and recycling of such waste from old vehicles, the following two objectives need to be pursued:

- Recycling of pure PUR foam from seats;
- Recycling of polyurethane foam from other parts of the vehicle, which has an integral surface layer, i.e., PUR foam injected directly into moulded parts, upholstery with an adhesive surface layer—textile, leather, and artificial leather (armrests, door upholstery, roof, etc.).

1.1. Analysis of Technological Options for the Recovery of Polyurethane Foams

The practice of recycling pure PUR foam from seats is widespread and technologically manageable. A significant problem is the recycling of PUR foam from other parts of the vehicle, as it has an integral surface layer. This problem is not currently being addressed. At the same time, for the conditions and annual quantities of waste PUR foam from old vehicles in Slovakia, it is necessary to design and optimise a technology that would allow material recycling of both types of waste.

Research, studies and testing have led to a range of areas and methods of recycling and using polyurethane that may be economically and environmentally viable [12]. The four main categories [13] are mechanical recycling, advanced chemical and thermo-chemical recycling, energy recovery, and recycling of the product itself (Figure 1). Each method provides unique benefits that are particularly suited to specific applications or requirements [14,15]. Mechanical recycling (i.e., material recycling) involves physical treatment, while in chemical and thermo-chemical recycling (i.e., raw material recycling), the waste is transformed into feedstock products or chemicals for the chemical industry. The energy recovery of this waste includes the full or partial oxidation of the material [16], the production of heat and electricity and/or gaseous fuels, oils and coal, except for by-products such as ash, which must be disposed of [17]. Due to the typically long service life of products containing polyurethane, a fourth option—product recycling or so-called closed-loop recycling—is limited [18,19], as markets are changing rapidly and the concept of downcycling or open-loop recycling applies strongly to chemical-based products such as

polyurethanes. Therefore, mechanical, chemical and thermo-chemical recycling and energy recovery are the only four ways to effectively recycle polyurethane [20].

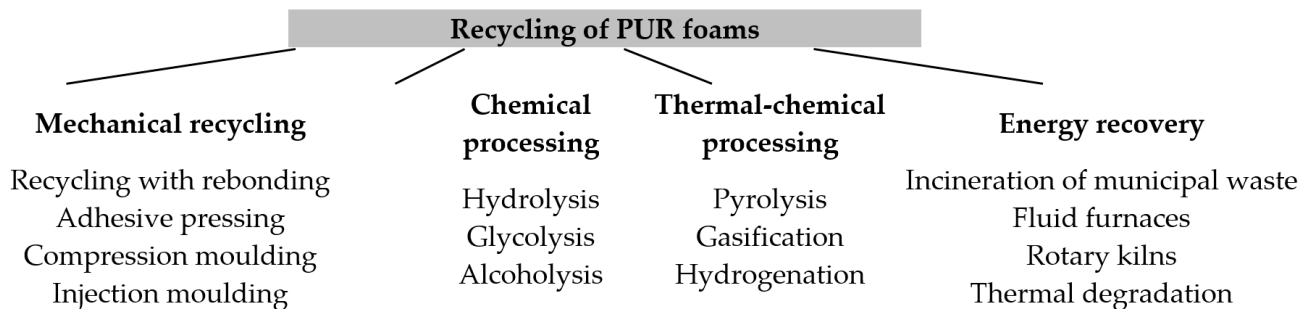


Figure 1. Overview of PUR recycling options.

Repair and reuse are generally not applicable for PU foam waste [6]. The mechanical and chemical primary treatment options are described in detail in review books [1,21,22] and papers [23–25], among others.

Regardless of the recycling technology used, the following two factors play a key role in determining the technical and economic feasibility of recycling polyurethane materials:

- Increasing the density of the bulk polyurethane foams, enabling cost-effective transport from the collection point to the recycling plant;
- Size reduction in polyurethane products (mattresses, car seats, insulation boards, etc.) suitable for further processing in the selected recycling process.

1.2. Mechanical Recycling

An important and first step is to process the waste materials into smaller particles, which are then easier to process. This can be flakes, pellets or dust, depending on the type of PUR that is recycled. For polyurethane foams, recycling by grinding is used. The resulting dust can be reused as a filler in the production of new PUR foams. In other cases, waste material is shredded [23]. The required fraction size for downstream processing of polyurethane ranges from particles smaller than 200 μm for reuse as fillers in polyurethane to larger pieces for chemical processing or energy recovery. There are four basic methods of mechanical recycling: recycling with rebonding, adhesive pressing, compression moulding, and injection moulding.

Recycling by rebonding with the addition of a binder is one of the most widely used recycling processes that are described in detail in studies [23,26–28]. It has been in use for 50 years. In the rebonding of polyurethane waste, 90.0% polyurethane scrap and 10.0% binder are usually added [23]. Currently, the most widespread mechanical recycling technology of flexible polyurethane foams that has found application on an industrial scale is the rebonding technique, which is almost only used for the recycling of pre-consumer flexible polyurethane waste [29]. In the rebonding process, the shredded polyurethane foam waste with dimensions of about 1 cm is mixed with the monomers or prepolymers used for the production of soft polyurethane foams, which are generally based on diphenylmethane diisocyanate (MDI) and polyols (ester- or ether-based). The main limit of this process is the minimum obtainable density, which generally exceeds 60 $\text{kg}\cdot\text{m}^{-3}$ [30]. The final stabilisation of the product is carried out by steam. The addition of steam completes the binding to make padding-type products of varying mechanical properties [6]. This process has been only used to produce foams for other applications (e.g., thermal insulation and construction) [23].

Adhesive pressing consists of layering polyurethane crumb and adhesive and then curing under the influence of temperature and pressure [19]. Granular polyurethane

particles are pressure-coated with a binder and heat- and pressure-cured. A semi-finished product is reobtained [6]. Moulded parts for the automotive industry, such as mats or spare tyre covers, are produced using this method. Adhesive pressing is applicable to many types of plastic waste and their mixtures. The shredded PUR foam with a particle size of approximately 1 cm can be reassembled into a compact unit by the addition of MDI and subsequent pressure moulding under temperature in the range of 100–200 °C and pressure of 3–20 MPa. Material recovery of the enormous amount of PUR foam from old vehicles can satisfy a large part (nearly 50% in the US) of the carpet underlay market [14]. This recycling method is also very interesting for PUR foam from construction waste [13].

This compression moulding method, which primarily uses reaction-injected polyurethanes as recycled raw material in the moulds, is capable of producing high-quality recycled products. The moulded parts contain 100% recycled material. The processed waste is ground to fine particles and subjected to high pressures and temperatures to create a solid material that is ideal for many automotive applications. Compression moulding [31,32] involves forming polyurethane particles at temperatures and pressures high enough (180 °C, 35 MPa) to generate shear forces needed to melt and bond the individual particles together, without the need for additional binders. The technology focuses on the production of upholstery from the processing of polyurethane and polyurethanes obtained from old vehicles. It is suitable for the production of rigid and complex three-dimensional parts such as shaped pump and motor casings. Products produced in this way are particularly suitable for the automotive industry because they achieve high stiffness [14,24,32].

Injection moulding technology enables partial recycling of polyurethane. One method (Bayer high-temperature pressing) processes granular polyurethane with a grain size of 250 to 1000 µm at a temperature of around 180 °C and pressures greater than 35 MPa, making it possible to produce thermoformed products such as various automotive parts [33]. It consists of melting granulate from waste plastics, including the fine fraction of PU, in the extruder chamber by external heating and subsequent injection of liquid plastic into the mould. Injection can be performed through a single extruder. Dual injection moulding with two (or more) extruders enables the recycling and reuse of waste thermoplastics and thermosets. The advantages of this technology lie in the increased mechanical properties of the product, in the higher surface quality, and in the possibility of any colour of the product.

A study [34] shows that by recycling PU foams, it is possible to produce products with higher density (almost 1 g·cm⁻³, which is approximately 14 times the original density of the foam) with good mechanical properties, using direct moulding technology, which is the compression moulding of particles without the addition of virgin material. The idea behind this technology is very simple: grinding the materials in order to give the resulting particles new reactivity. In fact, the broken bonds on the outer surface of the particles can act as polymerisation sites in subsequent processing steps. If residual reactivity of the bulk material is also present, it is useful to increase the reactivity of these particles.

As part of the research work [3], a novel method for the recycling of end-of-life mattress foam using an AIR-LAY process which employs a bi-component fibre as a binder for the polyurethane foam has been developed and optimised. This method permits obtaining materials with the same density as the starting foam (25 kg·m⁻³) and with a significantly lower density with respect to that obtainable with the rebonding process (above 70 kg·m⁻³) that uses isocyanates to bond the foam particles. The obtained recycled foams have been tested by mechanical compression and recovery tests showing that compression values of 3.7 KPa, similar to that of the mattress foam (3.0 KPa), can be achieved using 20% of bi-component fibres as binders and a density of 35 kg·m⁻³. On the contrary, only three times stiffer and denser materials can be obtained using the rebonding technology.

One of the necessary objectives for securing the European Union's Waste Management Strategy is the modification of industrial policy for the sustainable prosperity of the national economy. Waste prevention or material recovery should be a fundamental principle in the design of new products. From the point of view of waste management, emphasis must be placed on the possibility of efficient recycling of waste, with the result of obtaining full-value secondary raw materials that significantly replace the original primary raw materials.

The scientific hypothesis is based on the assumption of whether we can design an environmentally friendly and affordable technology for material recovery of PUR foam waste for 3D-shaped products that meet the thermal and acoustic insulation properties. The environmental criterion is based on the assumption that the recycled waste will not be affected by chemicals, such as adhesives, which would allow the new product to be recycled again using secondary raw materials. Affordability assumes that the investment and operating costs will not be high and that the proposed technology will be affordable for small- and medium-sized operations.

2. Proposal of the Appropriate Technology for the Stated Project Objectives

On the basis of an extensive analysis of technologies for the recovery of waste PUR foam, the technology for the moulding of PUR recycle under pressure will be further developed in detail for the project objectives mentioned above as the only suitable and efficient technology meeting the requirements and satisfying the constraints. This technology enables the recycling of pure PUR foams, as well as PUR foams with an integral surface layer. It enables the production of inherently flat insulating products, as well as shaped elements without the addition of binders or other chemical additives. The proposed technology of the material recovery of PUR foam from old vehicles does not pose a risk to the environment, as there are no chemical reactions or pollution of the environment by solid dusts, liquids or gases. Recycled polyurethane foam is also used in the construction industry as an insulating element for walls or floors thanks to its excellent thermal and acoustic insulation properties. This insulation is also suitable for soundproofing production halls. It can be produced either as plates, strips or blocks cut to the required thickness. Due to its higher strength and density, which can be adjusted during recycling, it is also successfully used as a floor layer for sports halls.

Utilisation of Products from Secondary Raw Materials

In the automotive industry, recycled polyurethane foam has great applications in soundproofing the bodywork and preventing vibration transmission (Figure 2a). In addition to the excellent sound-insulating and damping properties, the thermal insulation properties of such products are also exploited. Compared to the new PUR foam, they have a higher density and hardness. In automobiles, they are used in the form of plates or belts as insulation to damp vibrations and transmit them to the cabin (e.g., engine compartment insulation, door insulation, etc.).

Recycled PUR foam products can also be installed in white goods, doors and windows, machinery, and various equipment and structures as an acoustic element with a noise-absorbing function. Thanks to its excellent thermal insulation properties, it can contribute to reducing the energy consumption of buildings (Figure 2b). Such products can be recycled and turned into the same or a completely new product again.

With the proposed technology of moulding PUR recycle under pressure, it is possible to produce other rigid and shape-complex moulded parts for the automotive industry (Figure 2c). Parts moulded from recycle achieve guaranteed mechanical properties that may in fact be better than new polyurethane material.

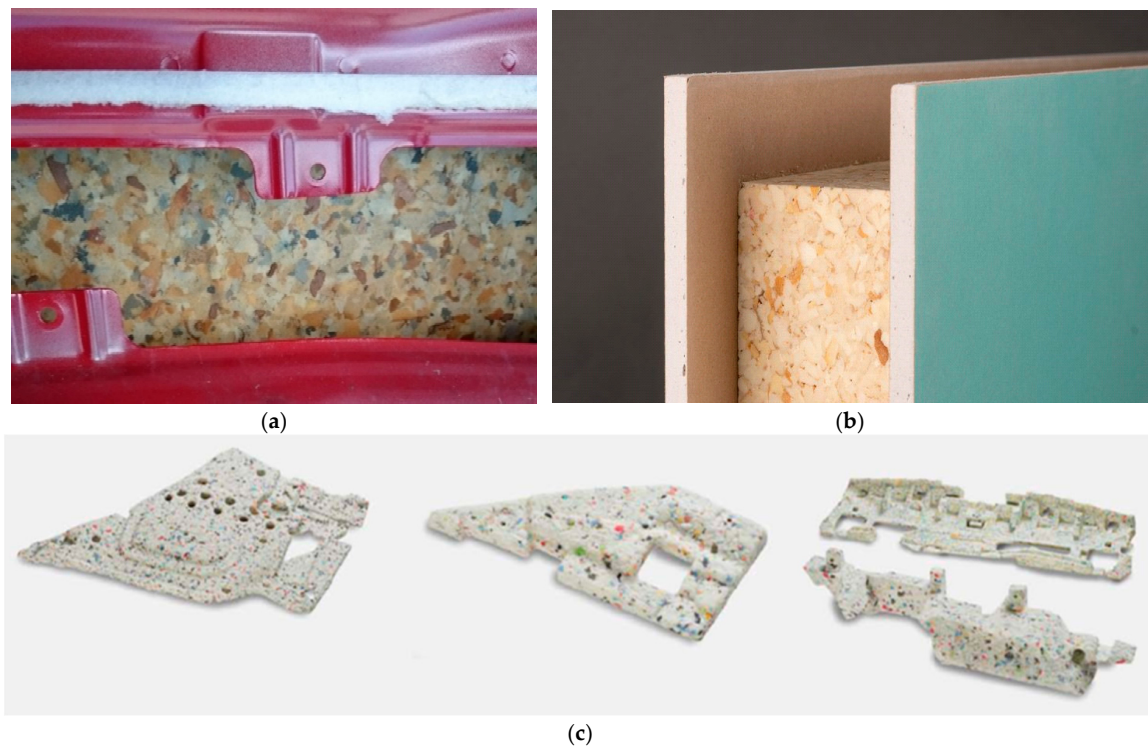


Figure 2. (a) Insulation for damping noise and vibration in automobiles, (b) sound and thermal insulation in construction, (c) shaped elements made of recycled PUR foam.

3. Experimental Development and Optimisation of the Proposed Technology

Within the experimental development of technology for the application of material recycling of waste PUR foams, especially from old vehicles, the aim is to define suitable (optimal) technological conditions for the production of shape-complex and precise elements. Figure 3 shows the potential applicability of the technology for the production of a number of elements in an automobile.



Figure 3. Acoustic filler in vehicle interiors and filler for car seats.

In the proposed method of hot compression (moulding) of PUR recyclate without the addition of adhesive, it is important to heat the shredded material to a temperature of 180 to 220 °C; when this flexible PUR foam softens, the edges of the individual flakes are melted, and the pressure can make the particles bond. The decisive technological parameters investigated in the experiments were the effect of pressing temperature, pressing force and endurance time. The other parameters were kept constant during the experiments: the shredded fraction with the fractional composition shown in Figure 4, the shape of the mould or the shape element.



Figure 4. Shredded waste from PUR foam.

For the above experiments, technological waste from the production of PUR foam, shredded into flakes, was used (Figure 5). To determine the fractional composition of the material, fractional analysis was performed on a Retsch Vibratory Sieve Shaker AS 200 (Retsch GmbH, Haan, Germany) digit and the material was divided into 5 fraction sizes.

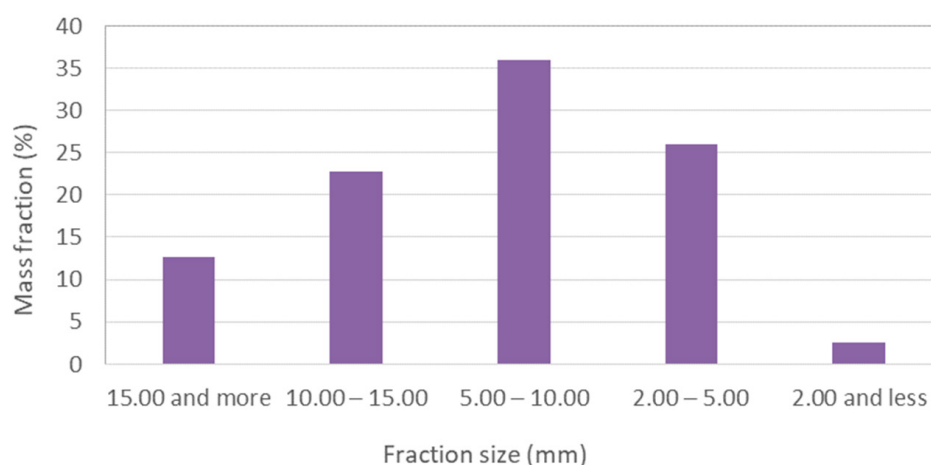


Figure 5. Fractional composition of the tested material.

For the experiments, a mould (Figure 6) was made to allow the simultaneous production of 4 samples. Each sample is a cuboid with the following dimensions: width of 50 mm and length of 100 mm; the thickness depends on the individual variable parameters examined in the experiment. A muffle furnace is used for heating. The empty mould and weights are put into the furnace and heated to the desired temperature. The mould is then

filled with shredded PUR foam; 25 g of material is placed in each part of the mould. After the mould is filled, the material of each sample is loaded with a precisely determined pressure applied by the weight of the weights. Filling is very fast and takes place in conditions that ensure that there is very little cooling of the mould and weights. The filled and loaded mould is placed back in the furnace for a precisely defined period. After the mould is emptied (Figure 7), the whole procedure is repeated under precisely defined experimental conditions. Figures 8 and 9 show the effect of loading on the resulting thickness of the samples.



Figure 6. Mould and weights for the production of experimental samples.



Figure 7. Filling of the mould with material.



Figure 8. Loading of samples with different pressing pressures.



Figure 9. Removal of finished samples from the mould. Different loads represent different heights.

The aim of the experiments is to define the optimal ranges of technological parameters of dry moulding under pressure and to determine the marginal values of the parameters' intervals in their individual combinations. The pressing temperature interval has critical limits based on PUR's state transition temperatures. Therefore, the following are the variable parameters of the process:

- Pressing pressure (2.0 kPa, 4.0 kPa, 6.0 kPa, 8.0 kPa);
- Heating temperature (200 °C, 225 °C, 250 °C);
- Heating period (10 min, 15 min, 20 min, 25 min, 30 min).

The examined parameters are evaluated with respect to the quality parameters of the samples: sample density, dimensional precision, stability and compressive strength. The experiments include 60 different combinations of technological parameters. For the sake of a sound statistical evaluation of the experiment, 10 samples of moulded-shaped elements were produced for each combination of technological parameters (a total of 600 samples).

4. Results of Experiments Using Dry Forming of PUR Foam Under Pressure

The developing technology of dry forming flexible polyurethane foam under pressure and heat without a binder has not been investigated in detail so far. All available studies used different binders, and most of them used steam for higher efficiency. The goal of our research is primarily to eliminate binders for cost-effective material recovery. And secondarily to eliminate the use of water steam—to use dry heating, because the semi-finished product made from recycled material is intended primarily for thermal insulation elements. Therefore, low humidity is essential.

The density of recycled PUR foam products produced by the compression moulding technology depends on the pressing pressure, heating temperature and heating period. The given intervals of the variable technological parameters were chosen on the basis of an analysis of the properties of the moulded material and an analysis of the results of published scientific papers. The results of the experiments show that with the defined intervals of the variable technological parameters, the magnitude of the loading pressure has the greatest effect on the increase in the density of the samples. The absolute values resulting from the experiments with the same intervals of the variables (Tables 1–3) show that it is the heating temperature that has the least effect on the density of the samples. Test samples achieving different quality were produced for all combinations of the variable technological parameters. Representative samples from each experimental setup are shown in Figure 10.

The results in Tables 1–3 show that the change in density in relation to the heating period is nearly linear at each temperature. The experiments have shown that in the examined temperature range, the heating period has a significant effect on the cohesiveness and dimensional stability of the sample. As the moulded material is a thermal insulator, the required heating period allows its volumetric overheating and the melting of the contact points between the individual flakes. The necessary heating period depends on the heating

temperature. As the temperature increases, the minimum period required for a cohesive and stable sample to form decreases.

The experiments also showed the time limits for the length of heating of the material. At temperatures of 200 °C and 225 °C, it is not possible to produce a cohesive and dimensionally stable sample with an endurance time of less than 15 min. The material does not overheat sufficiently and does not melt. The samples were not sufficiently cohesive and showed significant dimensional instability after removal from the mould. On the other hand, moulding at 250 °C has an upper endurance time limit of 25 min. With prolonged heating, degradation of the moulded material and caking occurred. Samples produced within the above limits showed good dimensional precision and stability, differing in their density.

An interesting result of the research is the effect of the individual technological parameters on the percentage increase in density. Figure 11 shows the results of PUR foam moulding at the lowest examined pressing pressure, i.e., 2 kPa. The results show a significant effect of the heating period as well as heating temperature. On the other hand, the results of moulding at 8 kPa pressing pressure shown in Figure 12 show that a change in heating temperature has a minimal effect on the change in density of the samples. At this level of pressing pressure, only the heating period has a significant effect on the density change. It is clear from these results that a detailed energy analysis will be necessary for practical applications, as both temperature and heating periods significantly affect the cost-effectiveness and competitiveness of the overall technology.

The resulting density of the samples represents a fundamental physical parameter influenced by the technological parameters in a non-linear way. Considerable attention should be paid to the study of the results of density increase caused by the variation in the individual parameters. A more comprehensive picture of the increase in density due to an increase in pressing pressure as well as extended heating periods at 250 °C is shown in Figure 13.



Figure 10. Test samples in the dry compression moulding experiment. Sample marking: Heating temperature (°C)/Heating period (min)/Pressing pressure (kPa).

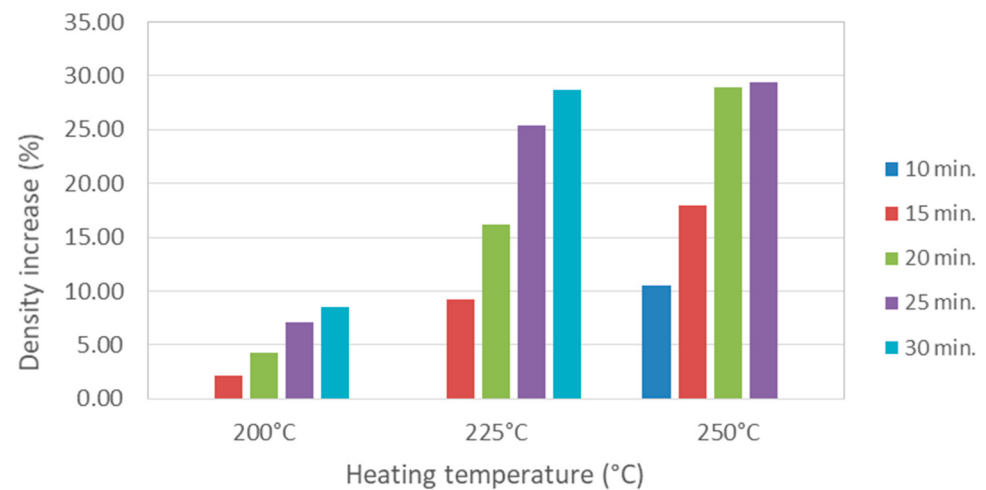


Figure 11. Effect on density change depending on temperature and heating period at 2 kPa pressing pressure.

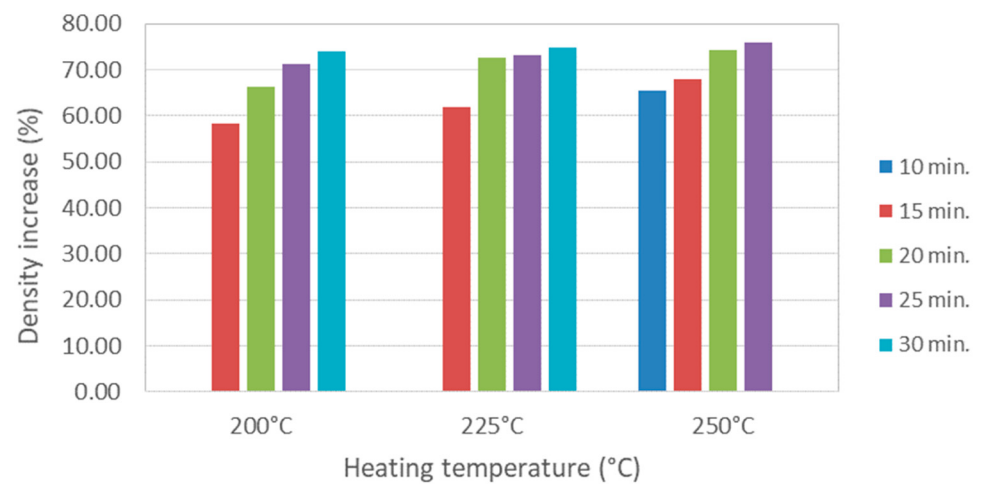


Figure 12. Effect on density change depending on temperature and heating period at 8 kPa pressing pressure.

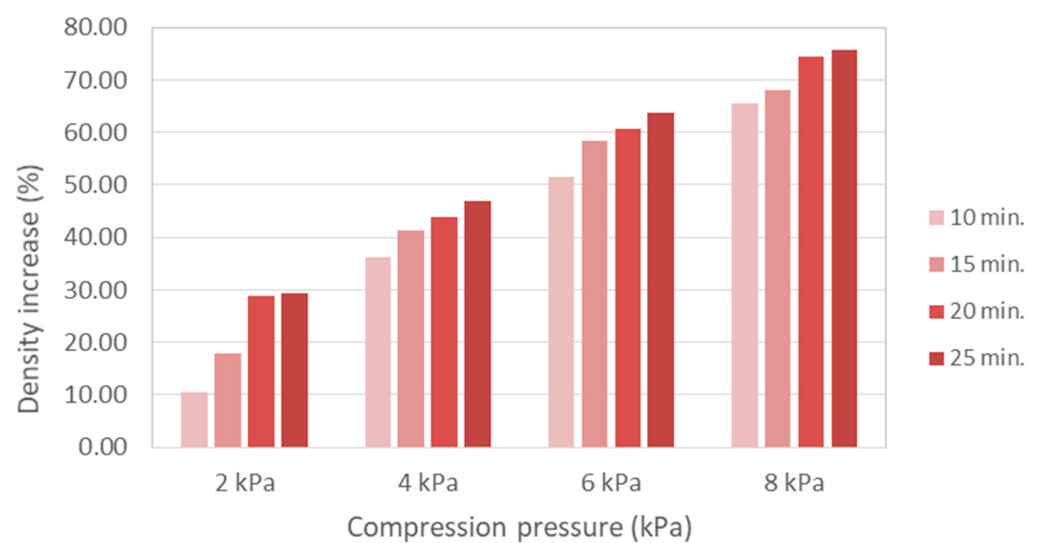


Figure 13. Effect on density change depending on pressing pressure and heating periods at a heating temperature of 250 °C.

Table 1. Average density values of samples produced at a heating temperature of 200 °C using various combinations of technological parameters.

Heating Temperature 200 °C					
Heating period (min)	10	15	20	25	30
Pressing pressure (kPa)	Density (kg·m ⁻³)				
2.00	Incohesive, dimensionally unstable	102.10	104.30	107.11	108.49
4.00		131.00	138.93	142.85	147.82
6.00		148.43	157.85	160.65	163.09
8.00		158.31	166.40	171.24	173.98

Table 2. Average density values of samples produced at a heating temperature of 225 °C using various combinations of technological parameters.

Heating Temperature 225 °C					
Heating period (min)	10	15	20	25	30
Pressing pressure (kPa)	Density (kg·m ⁻³)				
2.00	Incohesive, dimensionally unstable	109.17	116.20	125.33	128.67
4.00		133.55	140.14	147.05	148.15
6.00		148.60	159.78	163.25	166.88
8.00		161.84	172.60	173.18	174.87

Table 3. Average density values of samples produced at a heating temperature of 250 °C using various combinations of technological parameters.

Heating Temperature 250 °C					
Heating period (min)	10	15	20	25	30
Pressing pressure (kPa)	Density (kg·m ⁻³)				
2.00	110.4865	118.00	128.90	129.38	Material degradation, caking
4.00	136.2667	141.33	143.77	146.86	
6.00	151.4286	158.46	160.67	163.68	
8.00	165.60	168.00	174.36	175.83	

The results show that by adjusting the technological parameters, it is possible to control the density of the products while maintaining appropriate mechanical parameters. Experimental research has shown that with the right combination of parameters, it is possible to recycle waste polyurethane foam into new products without adding a binder and steaming.

Further Research Plan

It should be noted that these were only the first validation tests of the experimental samples. We will thoroughly analyse, verify and repeat these tests as needed. Testing of samples for compressive strength, tensile strength and flexural strength and determination of the required density for the mouldings is currently underway. Other parameters, such as steam, are likely to be added to the monitored parameters (temperature, pressure, endurance). The final results of these tests will be used in the definition of requirements and in the design of experimental equipment for the production of shape test samples.

A series of extensive compression strength measurements of PUR foam samples are currently underway in accordance with the Standard [35]. The measurement is carried out on standard laboratory equipment for tensile and compression tests (Figure 14). The method consists of placing the sample between two flat surfaces—a static and a moving plate (Figure 15). Due to the action of the moving plate, the sample is pressed to the appropriate thickness value. During the test, the load on the sample is measured in four cycles. The sample is pressed by 70% of its original thickness for the first three cycles and by 40% for the fourth cycle. At this point, the value of the pressing force is recorded, and the value of the compressive stress can be defined from the corresponding relationships.

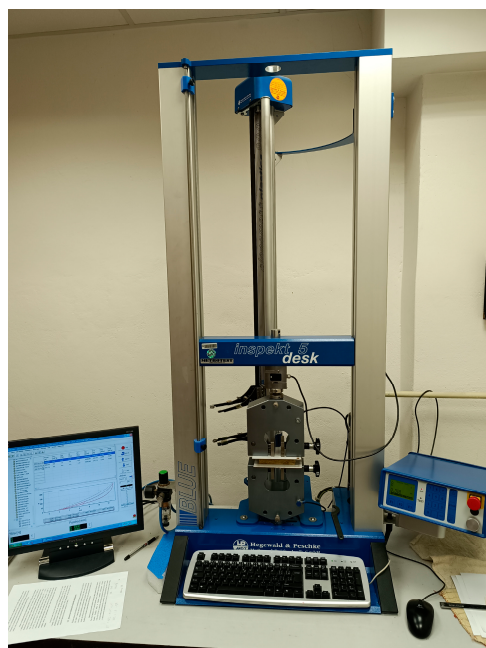


Figure 14. Measuring equipment for measuring the compression resistance of a sample – the universal testing device Inspekt Desk 5 kN (Hegewald & Peschke, Nossen, Germany).

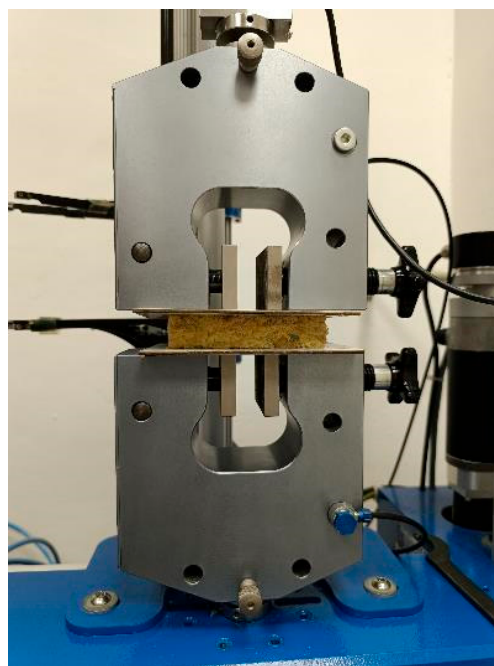


Figure 15. Location of the measured sample during the compression test.

5. Conclusions

The aim of the present paper is to investigate the material recovery of waste PUR foam. In doing so, the research was orientated towards a “clean technology” of material recovery of waste. This means that the recycling process and the subsequent production of new products take place without any chemical process and the addition of additives. The required shape, strength and damping properties are achieved only by the appropriate temperature and pressure ratio. By suitable combinatorics and optimisation of the named parameters, we can easily change the desired properties of future products. These include new lightweight porous materials with high thermal and acoustic insulation as well as materials with higher density, strength and required properties for mechanically stressed components.

The research has resulted in an environmentally friendly and affordable material recovery technology for PUR foam waste for 3D moulded products. In conclusion, the proposed and validated material recycling technology of waste pressure-formed PUR foam is applicable, under suitable technological conditions, for the production of precision moulded components with wide application not only in the automotive industry. The desired mechanical, acoustic, damping and thermal insulation properties of this material in the form of moulded elements make it suitable for a wide range of practical applications.

On the basis of the above, we conclude that we have succeeded in creating a technology for recycling PUR foam that is both cost-effective and affordable for small- and medium-sized operations. This method of recycling waste polyurethane foam materials, not only from old vehicles, has a very positive economic and environmental effect.

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