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Recycling Through Comminution: Characterization, Separation and Recycling Barriers of Metal Coated Polymers and Metallized Polymer Foams

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Abstract: The increasing global demand for raw materials underscores the importance of lightweight construction and sustainable material use, drawing attention to composite techniques like galvanic coating of plastics. To support recycling efforts, the development of efficient separation and material recovery processes is critical, particularly for end-of-life products containing metal-plated polymers. This study investigates the recyclability of metallized polymer foams and coated polymers through comminution, focusing on the potential for effective separation of metal and polymer components. Cu-ABS samples showed 27% of the products in the 8–10 mm fraction and 48% in the 10–16 mm fraction during primary comminution, while Cu-PUR achieved a more even distribution. Microscopic analyses revealed decoating rates of up to 95% for Cu-ABS compared to 19% for Cu-PUR. The comminution energy required for Cu-PUR was three times higher, with a fivefold lower decoating rate than solid materials. Particles larger than 200 µm exhibited interlocking, complicating the separation process. These findings highlight the need for optimized recycling processes to enable efficient raw material recovery and support a circular economy.

Keywords: metal foam; recycling; comminution; metalized polymerfoam; shredding

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1. Introduction

The increasing generation and accumulation of plastic waste have created significant global ecological challenges. Innovative approaches in polymer recycling and upcycling offer promising pathways to support the transition to a circular economy, focusing on reducing, reusing, and recycling plastics. Mechanical recycling methods play a key role, particularly in returning polymers to the production cycle, although technological and economic barriers, such as efficient plastic sorting, persist [1]. Chemical recycling methods, such as depolymerization, further enable the recovery of monomers for the resynthesis of polymers, thus enhancing the value and lifespan of materials [1].

In the field of additive manufacturing (AM), polymer recycling is increasingly recognized as a pivotal lever for promoting circular economy practices. The ability to produce filaments for 3D printing from recycled plastics opens new perspectives for sustainable production and the reduction of plastic waste [2]. As such, AM can serve as a centralized tool for supporting a transition to a circular economy by enabling the recycling and reuse of polymers and composites into new functional products [2].

Simultaneously, the number of electric vehicles and electric drives in general is expected to rise significantly in the future. This increase will lead to a sharp growth in the demand for copper to support this type of drive [3]. Addressing this surge in demand requires rethinking approaches in various technological domains. To satisfy the rising demand for raw materials, current research investigates the use of metallic lightweight materials, such as metal foams and metallized polymer foams, in cooling systems within the mobility sector. Previous studies have highlighted the exceptionally efficient heat transfer properties of metal foams [4].

The metal foam industry is undergoing a notable transformation, with Europe projected to experience a steady annual growth rate of 4.5% until 2032 [5,6]. Despite the dominance of Asian and North American markets, this growth signifies an important shift. To further enhance material, cost, and energy savings, metallized polymer foams produced through electroplating are being explored alongside metal foams. This approach aims to reduce overall production costs and improve process efficiency.

Given the growing market trends and increasing demand for copper, recycling entire structures will become increasingly important. The metallized nature of these structures requires specific processing methods for recycling at their end of life. Studies indicate that analyzed modules contain a metal content exceeding 75%. However, the absence of established recycling methods for such materials places emphasis on developing appropriate recycling strategies for electroplated foams. Current techniques include incinerating the polyurethane (PUR) core, but this step may be eliminated for multiple reasons. Consequently, this research explores and contrasts conventional methods for removing coatings from galvanized components.

In order to gain a better understanding of the layer adhesion of electroplated components, this is described again in the following section.

Adhesion between metals and polymers is based on various phenomena. On the one hand, adhesion can be caused by interfacial energy interactions such as van der Waals forces, hydrogen bonds or electrostatic forces, as well as by primary and secondary valence bonds. However, mechanical anchoring, also known as the push-button effect, occurs most frequently. This is caused by the introduction of undercuts, for example through processes such as grinding, pickling or etching. The coating material penetrates into these undercuts and anchors itself there. It should be noted that all bond types presumably contribute to the overall adhesion of the coating [7].

Necessity of Recycling and Use of Metallized Polymer Foams

On the one hand, structures are being investigated that are produced by investment casting and therefore consist entirely of the metal used. This means that recycling is largely unproblematic, as they can simply be recycled along the metallurgical process route or even be directly reused when dismantled. In addition, a very high burn-off is to be expected with the metallurgical route, as the ignition temperature is greatly reduced by increasing the specific surface area of powders and granulates, despite prior compaction. When using metallized polymer foams, recycling is not so trivial. It is possible to pyrolyze the metallized polymer foams after electroplating and thus remove the plastic core from the metal. This is not possible in all applications. For example, the research will investigate the possibility of attaching the foam structure directly to the baseplate and the power module during the electroplating process. In this case, the component cannot be pyrolyzed, as the power modules cannot be exposed to such high temperatures.

Furthermore, leaving the polyurethane core inside the structure has both thermodynamic and structural-mechanical advantages. As can be seen in Figure 1, irregularities occur in the layer thickness.

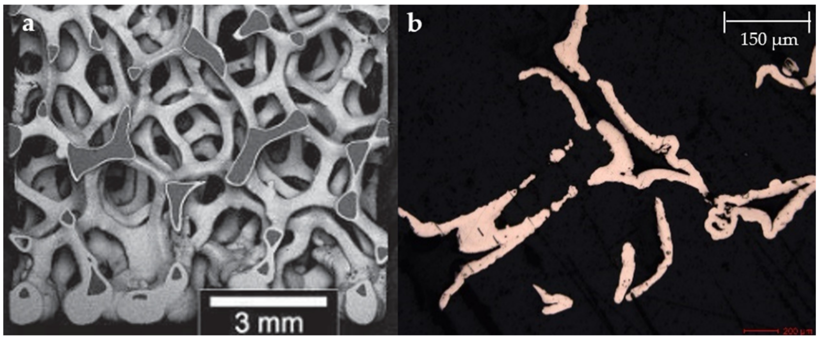

Figure 1. Aluminum foam coated with Ni (**a**) [8] and polyurethane (PUR) foam coated with Cu in section (**b**).

A few years ago, it was still an enormous challenge to coat foams of any kind (see Figure 1a. As can be seen in the figure, no regular layers were formed, but due to the structure of the foams, the field lines were positioned along the foam in such a way that the electric field was stronger on the outside than on the inside of the foam. In the meantime, the problem of the inhomogeneous layer thickness has been reduced somewhat. Figure 1b shows a cross-section of a coated PUR foam. The copper is shown in red. The PUR are the black inner parts. However, it can still be seen in (Figure 1b) that it is still irregular and sometimes even has holes. Simulations have shown that, depending on the viscosity of the fluid flowing through, these imperfections can fail and, due to the change in flow resistance, this imperfection becomes a kind of channel and the cooling capacity decreases enormously. In addition, the different thermal conductivity coefficients of air (0.025 W/mK [9]) and polyurethane (0.035 W/mK and as solid material 0.245 W/mK [10]) have a thermal effect. Although this is not significantly different, it noticeably improves the thermal behavior of the structure.

For these reasons, it can make sense to leave the polyurethane core inside the cooling structure. This also saves a very energy-intensive process step, which also produces a lot of emissions, which can be replaced in recycling by a purely mechanical process step that is significantly more energy and emission efficient.

2. Results

2.1. Shredding of Metallized Polymer Foams and Metallized Solid Materials

The comminution of metallized polymer foams has already been investigated in several other studies and compared with other materials. In [10], for example, metallized polymer foam was compared with solid PUR material and it was found that with the same sequence of process control, it was possible to achieve a factor of 5 better evaporation (separation of Cu and PUR).

As seen in Figure 2, solid material was fully separated in the initial comminution stage (approx. 55%). This is because PUR solid material is unsuitable for coating due to its waxy, smooth, and soft surface, ideal for industrial plain bearings and rollers. Consequently, the adhesion of the coating to the polyurethane surface was insufficient, as reflected in the results. A decoating degree of 95% was achieved with the solid material compared to only 19% with the PUR foam [11]. This was computed using the Formula (1): $DGDG =$ Degree of Decoating in %, $m_{pf} =$ Mass of polymer in the light fraction, and $m_{pi} =$ Mass of polymer in the input material:

$$DG = \frac{m_{pf}}{m_{pi}} \times 100 \tag{1}$$

The polymer content in the input material was given by the manufacturer and the mass of the polymer in the light fraction was calculated by gas pycnometry and a ratio equation using the volume and the real density of the mixture. It should be noted that a light fraction was only considered as such if it had a purity of 99.9% by weight. Otherwise, this sample was added to the heavy fraction.

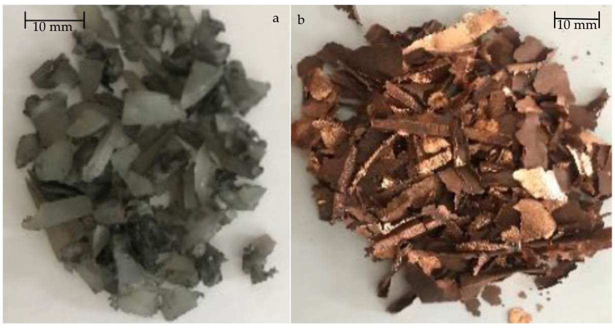

Figure 2. Complete polyurethane material after comminution with separated PUR (**a**) and Cu (**b**).

Similarly, in the pure investigation of decoating degree, Acrylic butadiene styrene (ABS) solid material was compared to PUR foam [12]. This comparison aimed to validate previous results concerning PUR foam decoating. ABS foam exhibited a decoating effect of approximately 19.5% by weight, consistent with findings in [10]. ABS decoating results mirrored those of PUR, reaching 92% by weight [12]. Notably, ABS couldn't be comminuted in the same manner as solid PUR materials.

As can be seen in Figure 3, the ABS melted partially because it became too hot inside the machine. As a result, only the sieve grate became clogged and no comminution took place, but the tools were blocked, which is why this test was aborted in order to soften the material on another machine. The machine used was the universal mill from Jehmlich, with the test parameters described in the next chapter. As the material could not be comminuted in the universal mill, it was subsequently comminuted in the granulator. The procedure is described in the following chapter.

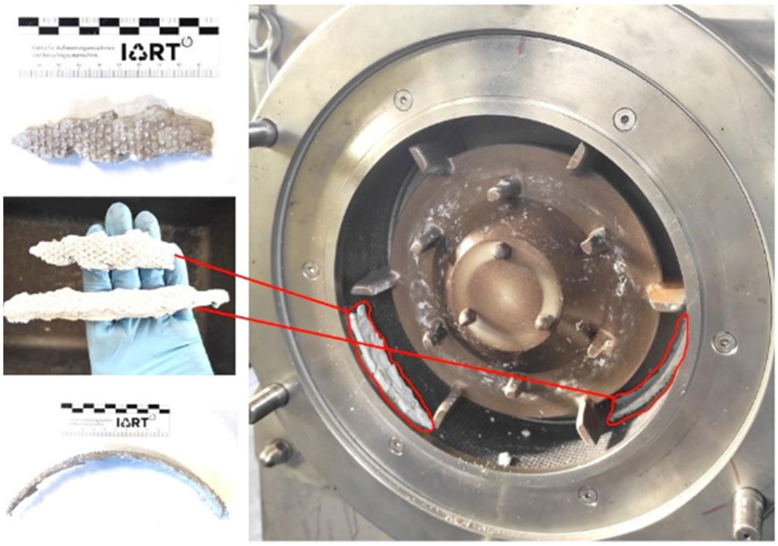

Figure 3. Melted ABS inside the machine.

2.2. Particel Form of Pre Grinding

In the case of Cu-ABS, easily distinguishable "liberated" copper particles with the morphology (Figure 4c) platy-flat; (Figure 4d) rod-shaped straight; (Figure 4e) rod-shaped slightly deformed. In addition, several ABS particles adhere to their copper coating.

It can be seen that the ABS fracture (white) is typical of a ductile material and produces soft, non-sharp-edged grains. Metallic lamellar particles of over 6 mm have detached "cleanly" and relatively easily from the polymer substrate, confirming the slight delamination of a solid and rigid substrate. Bends and radii can be seen in the metal particles, but their compactness is minimal. The back of the copper particles shows a dark gray activation layer formed in the early stages of electrodeposition.

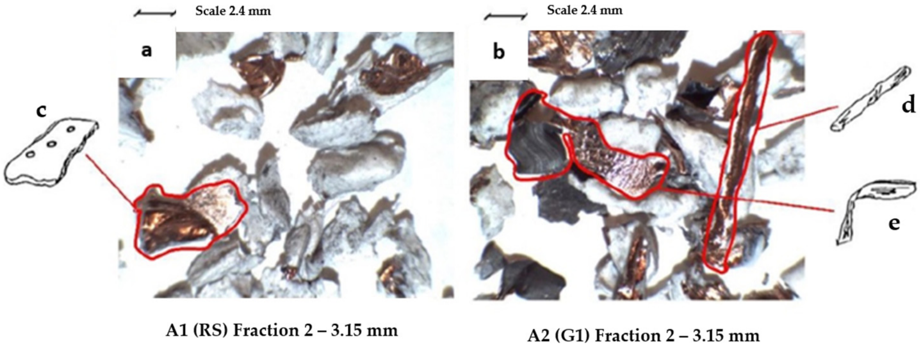

Figure 4. Particle form of Cu-ABS in pre shredding after Rotary Shear (**a**) and Granulator (**b**).

Cu-PURS produces a particle collective with a fluffy appearance and spongy morphology, in which the dark polyurethane substrate is visible, but which does not show any significant decoating. As can be seen in Figure 5, the particle morphology and the structural features appear in a significantly different form than can be seen in the Cu-ABS samples in Figure 4. As already mentioned, the copper particles in Figure 4 are already detached in a platy and lamellar manner, while as can be seen in Figure 5, the coated polymer foam only deforms and balls up due to the ductility of copper, which makes it impossible to decoat the polyurethane. The respective types of deformation are shown for better illustration.

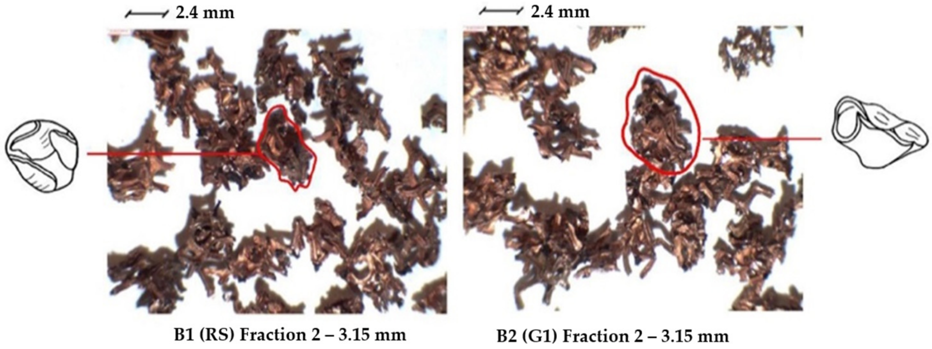

Figure 5. Particle form of Cu-PUR in pre shredding.

In this material morphology, deformations and compressions can be observed in which the "holes" of the foam have collapsed on themselves and the material tends to form "lumpy" particles. The samples crushed in the rotary shear show this collapsed appearance more pronounced. This is directly related to the cutting load applied to the material and the grinding mechanism used by the machine. In the secondary grinding (fine comminution), the particle shape is similar to that after pre-grinding, which can be explained by the similar stress. After the second grinding, the Cu and ABS particles are almost completely separated from each other.

2.3. Particle Size Distribution (PSD)

Pre-grinding. After sieving the particle collective, the mass data for each product fraction was listed in a table. These values are presented graphically for each sample according to the type of material and the size reduction machine used. The PSD data is best presented in the form of logarithmic scale graphs.

As you can see in Figure 6, the comminution of the Cu-ABS samples yielded approx. 27% of the products in the fraction of 8 to 10 mm and approx. 48% between the fractions of 10 to 16 mm. In contrast, the Cu-PUR samples showed a more balanced fraction distribution with significant proportions in many fraction sizes. On the basis of the increase in the graphs, conclusions can be drawn about the density distribution of the particle sizes. This difference is related to the tensile strength and the brittleness of the material, with that of

Cu-PUR being significantly lower and therefore it can be crushed with less energy and produces finer particles.

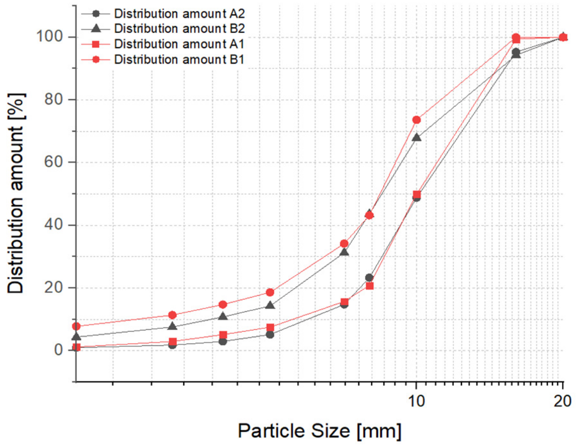

Figure 6. PSD of pre shredding.

2.4. Layer Seperation

Even if the decoating effect was not determined quantitatively, the images in Figure 7a,b clearly show how the copper layer in one of the larger particle classes detaches from each other after the first comminution step. This is due to the fact that in the foam structures (Figure 7b) the particles start to ball up and thus the bars are locked together. In addition, the copper layer encloses the substrate in its entirety and thus, in addition to the mechanical interlocking, which is the predominant bonding mechanism in Cu-ABS, the cladding is also created. In order to break through this, the Cu-PUR particles must be reduced to less than 200 µm. This is also the reason why a significantly lower comminution energy has no significance for foam and was not used as a comparison here. The comminution energy of the solid material is higher by a factor of 3, while the decoating is lower by a factor of 5 according to [12].

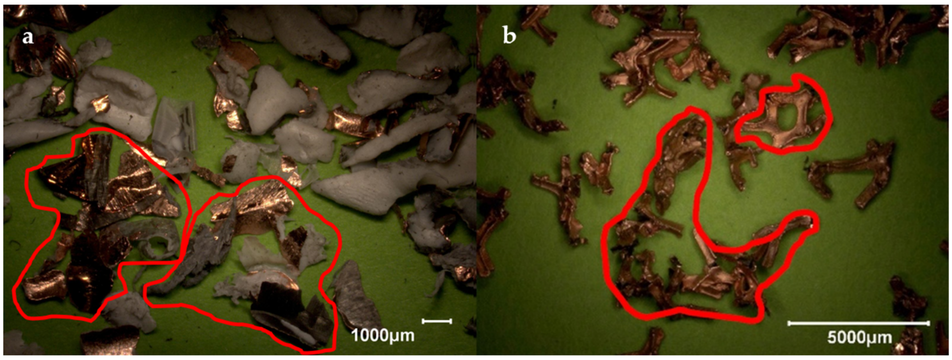

Figure 7. Cu-ABS (from A1) (**a**) and Cu-PUR (from B1) (**b**) in particle size of 2–3.15 mm.

3. Test Material and Test Procedure

3.1. Test Material

The tests here relate only to the characterization of the different samples in terms of particle size distribution, shape and energy stress and not to the decoating effect.

The test material had the same dimensions as that used in the decoating tests. The exact dimensions, characteristics and components can be found in the table below. It should be noted that palladium was only used in very small quantities to activate the surface and is therefore negligible. Figure 8 shows the test material and Table 1 shows the data of the test material.

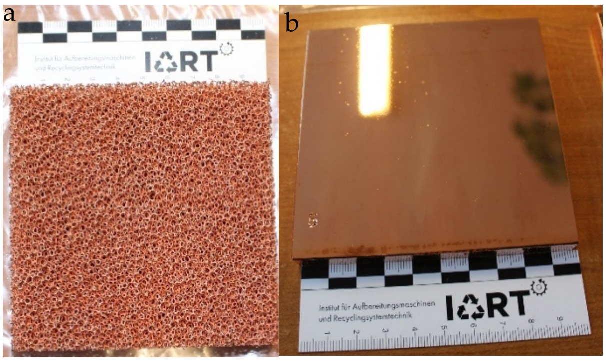

Figure 8. Coated foam (**a**) and coated ABS in sheet form (**b**).

Table 1. Sample Parameter and Data.

Parameter	Coated Metalized Foam (Figure 4a)	Coated Solid Material (Figure 4b)
Dimensions	100 × 100 × 20 mm	100 × 100 × 20 mm
Activation Layer	Nickel germination	Nickel germination
Layer Thickness	Ca. 30 µm	30–50 µm
Materials	PUR, Cu, (Ni)	ABS, Cu, (Ni)
Total Mass	399.3 g (divided into 2 test runs)	898.9 g (divided into 2 test runs)
Naming	Cu-ABS	Cu-PUR

3.2. Test Procedure

As you can see in Figure 9 both materials underwent a similar grinding process. Initially, pre-grinding occurred using a rotary shear (TUBAF type—Twin-shaft axial-splitting rotor shear—Built at the Technical University of Freiberg in Germany) equipped with two independently driven, counter-rotating shafts. Each shaft is powered by a 4 kW motor, with an axial gap width between the grinding tools of approximately 0.1 mm and a tool geometry of 20 × 30 mm^2 for the first process route. For the second route used toll was a granulator (ZM-300) from the company THM in Eppingen, Germany with a grate of a width of 10 mm. This granulator utilizes square tool geometry, with a 0.2 mm gap between the tool and the blow bar, and approximately 5 mm gap between the tool and the grate for the solid material. For metalized foam there where used subsequently a secondary grinning process employing the impact mill (REKORD 224-M-universal mill of the Company JEHMLICH, Nossen, Germany), which features an impact nose rotor and a 2 mm wide grate. This mill is driven by a 15 kW electric motor, and grinding occurred at a velocity of 82.1 $\text{m}\cdot\text{s}^{-1}$. In the second grinding of the solid materials, the impact mill could not be used for the above-mentioned reasons and the granulator with a grate width of 6 mm was used again under the same parameters as in the first comminution stage.

Subsequently, after each comminution stage, a classification was carried out using sieves with the following parameters for the primary comminution stage, the following sieves were used for Cu-PUR and Cu-ABS: (16; 10; 8; 7.1; 5 4; 3.15; 2 mm). For the secondary stage of comminution, the following sieves were used for Cu-ABS: (5; 4; 3.5; 2; 1 mm and 710; 500 µm) and for Cu-PUR: (2; 1 mm and 710; 500; 355; 200 µm). The sieves selected for the test are arranged in a stack. The sieve stack is then placed in a Retsch (Germany) AS 200 sieve tower and sieved with an amplitude of 1.60 mm, and intervals of 10 sec for 3 min. Table 2 shows the sample masses of the individual process routes and the losses recorded. The samples were weighed using a scale of the manufacturer Mettler Toledo (Columbus, Ohio, USA, model: TLE3002E).

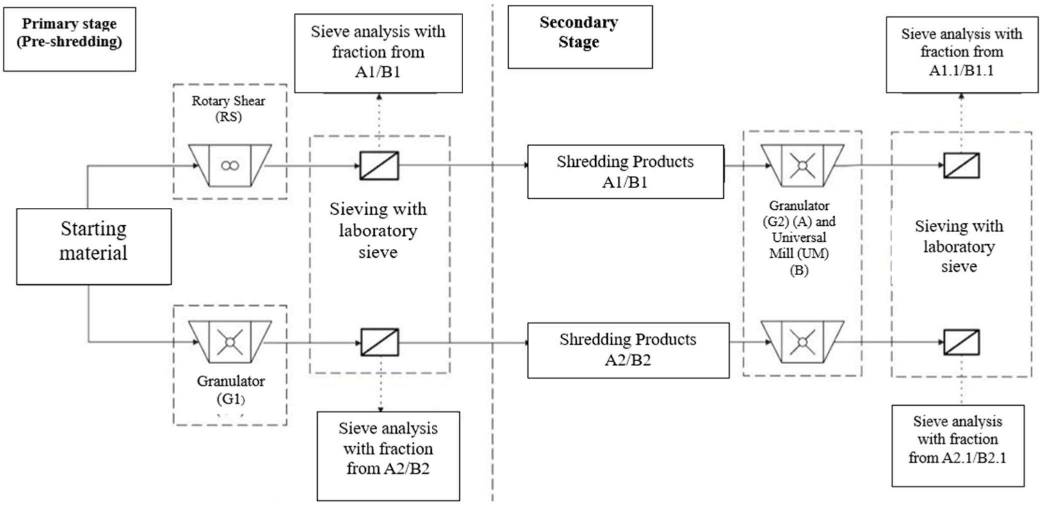

Figure 9. Test sequence of the two process routes with solid material in Granulator (G1/2) and metallized foam in universal mill (UM).

Table 2. Saple masses and losses.

Code	Naming	Input Mass [g]	Output Mass [g]	Loss [%]
A1	Cu-ABS in RS	450.9	447.4	0.77
A1.1	Cu-ABS in RS + G2	447.4	431	3.67
A2	Cu-ABS in G1	448	447.9	0.02
A2.1	Cu-ABS in G1 + G2	447.9	431.8	3.6
B1	Cu-PUR in RS	194.5	192.1	1.23
B1.1	Cu-PUR in RS + UM	192.1	188.3	1.99
B2	Cu-PUR in G1	204.8	202.2	1.25
B2.1	Cu-PUR in G1 + UM	202.2	198.8	1.68

RS: Rotary Shear, G1: Granulator pre grinding, G2: Granualtor second grinding, UM: impact mill.

It can be seen that higher losses are recorded in the granulator, especially with solid material, because the particles are significantly larger and get stuck in the tools or are accelerated outside the collection container. The foam particles are much lighter and therefore the braking effect due to air resistance is much greater.

4. Conclusions

This study investigated the comminution behavior of coated solid materials and foams. The tests carried out provided valuable findings, particularly with regard to particle shape and size distribution. The analysis of the particle shape in equal sizes already allowed a clear distinction between the materials and indicated potential improvements in terms of decoating. These results were further supported by the particle size distribution. Microscopic images confirmed these assumptions and showed effective decoating of the substrate for the solid materials. The particle shapes reflected the properties of the different materials very well and also show the problems that arise during comminution. While the ABS behaves in a brittle-breaking manner and forms flat particles, the PUR foam behaves in a ductile manner and does not break, but is shielded or deformed. This behavior is a problem for the decoating process, as it forms balls that make it extremely difficult to reach the PUR core in order to decoat it.

Important conclusions are:

- ABS exhibited brittle-breaking behavior, forming flat particles, whereas PUR behaved in a ductile manner, leading to the formation of spherical particles. These spheres hinder access to the PUR core and complicate the decoating process.
- Primary comminution of Cu-ABS resulted in 27% of particles in the 8–10 mm range and 48% in the 10–16 mm range, while Cu-PUR displayed a more uniform distribution.

- Particles larger than 200 µm exhibited interlocking, further complicating the separation process.

It was found that the exclusive consideration of the mass-specific comminution energy is not a sufficient basis for solving the problem. The complexity of the decoating process therefore requires a more differentiated approach.

For future studies, it is crucial to investigate the decoating behavior in more detail and to narrow down the causes of the comminution phenomena in foams. Particular attention should be paid to decoating in the upper particle size classes. This requires a multidisciplinary approach that takes into account both the physical and chemical properties of the materials. By gaining a deeper understanding of these processes, more effective decoating methods can be developed that make an important contribution to the circular economy and resource conservation.

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Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

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