Mechanical Recycling of Autoclave Laminated Plastic

Towards Sustainable Waste Management

BM51035: BME MSc Thesis

Mervin Hansen
4945697

Delft University of Technology

Mechanical Recycling of Autoclave Laminated Plastic: Towards Sustainable Waste Management

MSc Thesis Biomedical Engineering

Mervin Hansen
MSc Student Biomedical Engineering
Delft University of Technology

Supervisors:
B. van Straten
T.J. Horeman
H.F.J Lieshout

***Abstract*—*Introduction:* The environmental impact of single-use medical plastics is a growing concern. One such plastic component, autoclave laminated plastic (ALP), commonly used in medical sterilization pouches, is typically discarded after use, but it holds potential for recycling. This research paper investigates the feasibility and effectiveness of mechanical recycling as a solution for ALP waste.**

***Methods:* The study commences by analyzing the composition of collected ALP materials using X-ray diffraction analysis. Subsequently, recycling techniques, including melting, and shredding, are explored and assessed.**

***Results:* After multiple melting setups, the high-pressure melting setup, augmented by a continuous pressure spring mechanism, successfully yielded recycled ALP blocks that could be further processed into dogbone-shaped specimens. The results of the tensile tests revealed a Young's modulus of 269 ± 85 MPa for the recycled ALP.**

***Discussion:* These findings hold promise in the transformation of ALP waste into valuable recycled products, mitigating the environmental impact of single-use medical plastics. However, the study underscores the need for further optimization of the recycling process to enhance mechanical properties and overall material quality, ultimately advancing sustainability efforts in the medical sector.**

***Index Terms*– Mechanical recycling, Medical sterilization pouches, Medical waste, Packaging waste, Plastic waste, Polypropylene, Sustainable healthcare**

I. INTRODUCTION

In recent years, there has been a growing concern over the environmental impact of plastic waste, particularly in the healthcare sector, where many plastics are disposable [1]. Single-use plastics, in particular, have raised significant environmental concerns [2]. Autoclave laminated plastic (ALP) used in medical sterilization pouches is a noteworthy example among various types of plastic waste generated by the medical industry. Medical sterilization pouches provide a critical barrier to maintain sterility and protect medical supplies such as medical instruments during transportation and storage [3], [4]. These pouches are used in healthcare facilities where individual instruments are packaged, such as dental clinics, general practices, and hospitals. However, their disposal often results in significant waste accumulation. Nevertheless, there is potential for recycling paper and plastic packaging used in the operating room, which could significantly contribute to addressing the plastic waste issue [5].

Although several studies have addressed the recycling of various types of medical plastic waste, such as blue polypropylene wrapping paper [6] and plastic medical syringes [7], to date, no research has been conducted specifically on the recycling of the plastic component found in medical sterilization pouches. This gap in the literature highlights the need for further investigation into the recycling potential and challenges associated with this specific type of medical plastic waste. Understanding the feasibility and effectiveness of mechanical recycling the plastic part of medical sterilization pouches could contribute to more sustainable waste management practices in the healthcare industry.

Prior to this study, a literature review [8] was conducted to explore potential methods for recycling autoclave laminated materials. The recycling of ALP offers a promising avenue for waste management, potentially reducing the environmental burden associated with its disposal and enabling the recovery of valuable resources. The literature review [8] indicated that mechanical recycling of ALP is the most suitable recycling technique. This preference is based on its advantages over chemical recycling, namely its superior performance in terms of CO2 emissions reduction and cost-effectiveness. Additionally, mechanical recycling is favored for its lower energy consumption and ease of handling. In contrast, landfilling and incineration, as waste management methods, are considered less sustainable compared to both mechanical and chemical recycling. Eventually, a mechanical recycling method was suggested, which involved a melting, shredding, and injection molding process.

This research paper aims to investigate the suggested (thermo) mechanical recycling method, illustrated in Figure 1 [8]. The proposed approach combines thermal and mechanical treatment processes for the recycling of the plastic component of medical sterilization pouches.

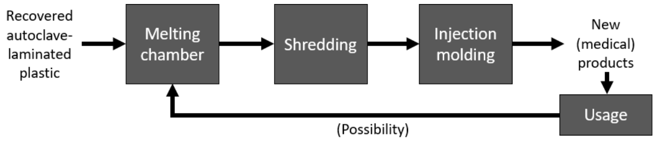

Fig. 1. Transformation of autoclave-laminated material to recycled plastic [8]

The following **research question**, along with its corresponding *sub-questions*, was formulated for this study.

- **How can autoclave-laminated material (ALP) be recycled mechanically?**
- *What is the composition of the collected material?*
- *What is the Young's modulus and ultimate tensile strength of the recycled ALP?*

For the design of the various melting setups, a set of requirements was established. These requirements included:

- Compatibility with a circular induction furnace (34 cm diameter, 35 cm depth)
- Reusability
- Capability to produce flat or circular dogbones using post-processing
- Simple retraction of recycled plastic
- Ease of assembly and disassembly
- Ease of fabrication
- Cost-effective design
- Safety

This research paper is structured as follows: Chapter 2 provides a description of the methodology for the different experimental steps taken. Chapter 3 presents the findings and results obtained from the various research steps. Chapter 4 discusses the findings per research step taken and also provides insights into the limitations of the study, and suggests avenues for future research. In Chapter 5 the conclusion can be found.

II. METHODS

This study intentionally involved eight steps to analyze the feasibility of mechanically recycling ALP waste for utilization in various products. The different steps undertaken to address the research and sub-questions can be found in Figure 2. These steps encompassed collecting ALP, conducting X-ray diffraction analysis on the ALP material, developing a melting process, shredding the remolded ALP, establishing an injection molding process, and analyzing the injected molded dogbones. However, it is important to note that the actual experimental steps deviated from some of the initially proposed steps. Detailed information regarding this can be found in the results section and subsection II.E.

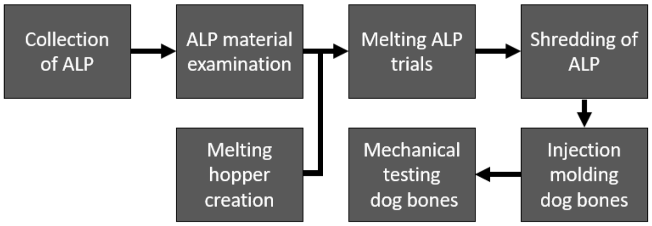

Fig. 2. Overview of Experimental Steps

A. Collecting process of ALP

Initially, an amount of ALP weighing 630 grams was already available and obtained from Greencycl (Utrecht, the Netherlands). However, for experimental testing, a larger quantity of ALP waste was desired. To address this, a small logistical process was established to collect additional ALP waste from the oral surgery department at Ikazia Hospital (Rotterdam, the Netherlands). With the assistance of Dr. B.I. Pluijmers, the staff at the policlinic was requested to collect used medical sterilization pouches in separate plastic waste bags. After a collection period of 2 weeks, the bags were transported from the hospital to Delft University of Technology (Delft, the Netherlands) for further investigation. This study utilized uncontaminated medical sterilization pouches that had not come into contact with patients. Consequently, this type of waste is classified as general waste [9].

B. X-ray diffraction analysis

The collected ALP underwent X-ray diffraction (XRD) analysis. A total of four XRD studies were conducted, including one with green ALP, one with blue ALP, and two with white ALP. During the separation process of the different types of white ALP, it was observed that there were two distinct laminated sheets: one flexible and one stiff. Consequently, two separate XRD studies were performed on the different white ALP samples. The specimens were folded several times and mounted on a SI50 zero-background wafer. The XRD test was conducted using the Bruker D8 Advance diffractometer, and the obtained data was subsequently evaluated with the Bruker software DiffracSuite.EVA vs 6.1. R. Hendrikx at the Department of Materials Science and Engineering of the Delft University of Technology is acknowledged for the X-ray analysis.

C. Melting process

For the granulation of the laminated sheets, the decision was made to create plastic blocks due to the successful recycling methods employed in other research studies on plastic materials [6], [10]. To facilitate the creation of plastic blocks from the laminated sheets, a melting hopper was constructed. The hopper was made using a 100x100 mm steel square tube and folded sheet metal. It was designed for easy assembly and disassembly, allowing for convenient removal of the produced plastic blocks. Inside the tube, a filter was installed to separate the molten material from the laminated sheets. Additionally, a weight was positioned on top of the laminated sheets to exert pressure and facilitate the passage of the melted ALP through the filter. The Melting hopper, which can be seen in Figure 3, was then placed in an electrical melting oven (KOS, Electric crucible, series 219029).

Fig. 3. Melting hopper for the creation of plastic blocks

According to a paper by Patil et al. (2021) [11], the composition utilized ALP (a-polypropylene and carbon nanotubes) has a glass transition temperature of approximately 162-165 degrees Celsius. Figure 4 illustrates the trend of the glass transition temperature, showing an initial increase followed by a decrease. The changes observed in the glass transition temperature were found to be minimal, indicating that the presence of carbon nanotubes has a minimal impact on the thermal behavior of polypropylene. So, in this research, the influence of carbon nanotubes on the glass transition temperature of polypropylene is considered negligible. In the various melting processes, temperatures ranging from 200 to 325 degrees Celsius were employed, as they were also utilized for polypropylene recycling in the study by Van Straten et al. (2021) [6].

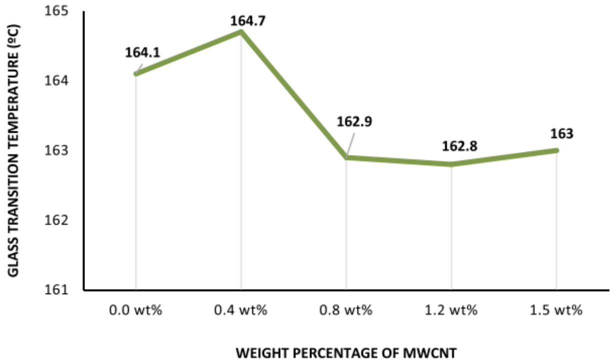

Fig. 4. Glass transition temperature of Polypropylene with carbon nanotubes [11]

D. Shredding and injection molding process

The Moditec Goliath Plus Granulator (Figure 5), located at van Straten Medical (Utrecht, the Netherlands) was utilized for granulating of the ALP. The resulting granules were collected in a plastic bag and subsequently transported to Model Engineering (The Hague, the Netherlands) for injection molding. The injection molding process was carried out using a BabyPlast Moditec injection molding machine (Figure 5), and a specific mold was employed to create a dog bone-shaped sample. The injection molding process is being conducted at a temperature of 210 degrees Celsius.

Fig. 5. Left; The granulator, Right; The Babyplast injection molding machine

E. High-Pressure Melting

Due to the difficulties encountered during the melting and injection molding phase, which will be discussed in the results section, an alternative recycling method was developed for melting plastic under high pressure. For this melting process, a basic setup was used to test the potential of melting under pressure, consisting of a 36 mm inner diameter steel pipe closed at one end, a steel pressure device, and multiple bolts. The setup can be found in Figure 6.

Fig. 6. First high-pressure melting setup: 1. Steel Pipe, 2. Closure Plate Welded to Steel Pipe, 3. Compressing Rod, 4. Bolts Ensuring Compressing Rod Locking.

The steel pipe was utilized to contain ALP flakes, which were subsequently subjected to compression using a glue clamp. Through repeated filling and pressing cycles within the steel pipe, the ALP flakes experienced elevated pressure, which was maintained by the presence of screws inserted into pre-drilled holes. To facilitate the melting process, the entire setup was positioned inside an electrical melting oven (KOS, Electric crucible, series 219029) and exposed to a temperature of 210 degrees Celsius. This temperature choice was determined based on the results obtained from the non-burned plastic in the BabyPlast.

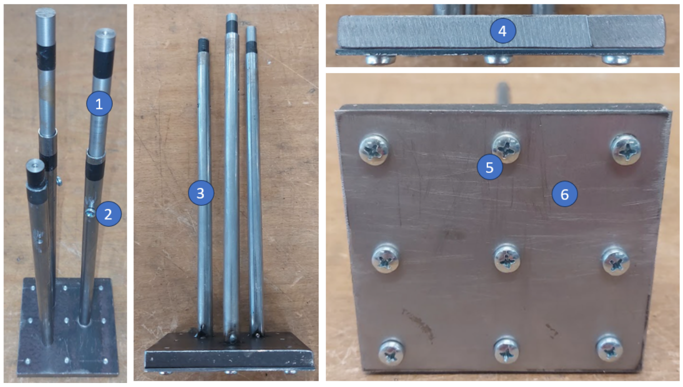

Fig. 7. Second high-pressure melting setup: 1. Compressing Rod (3x), 2. Locking Bolts Ensuring Compressing Rod Locking (3x), 3. Compressing Tubes (3x), 4. Stability Block Welded to Compressing Tubes, 5. Closing Bolts for Closure Plate, 6. Closure Plate for Tube End Sealing.

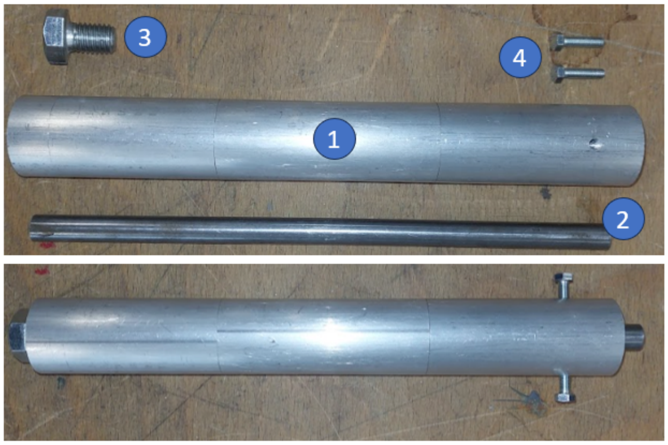

Fig. 8. Third high-pressure melting setup: 1. Compressing Pipe, 2. Compressing Rod, 3. Closure Bolt for Sealing One End of the Pipe, 4. Locking Screws to Secure the Compressing Rod.

Following the testing of the setup depicted in Figure 6, a new setup was developed while adhering to the same logic. The revised setup featured a smaller tube with a 10 mm inner diameter closed on one end, a compressing rod, and a bolt per tube. This updated configuration will be loaded with a workshop press, and due to its reduced dimensions, is expected to achieve higher pressure levels. This setup can produce three ALP rods during a melting process, which can be easily retracted by opening the bottom of the setup. The details of this setup can be found in Figure 7. Additionally, the melting process was conducted at a temperature of 220 degrees Celsius, due to the results of the prior setup.

Following the testing of the second high-pressure setup, it became apparent that certain flaws needed to be addressed, necessitating the development of a new setup. This updated setup incorporates an increased wall thickness to mitigate the problem of deformation or nodding during the pressing process. Furthermore, a locking mechanism and a closing system with a bolt were implemented to facilitate the retrieval of the recycled ALP rod with ease. These modifications are expected to enhance the structural integrity of the setup and improve the overall performance of the recycling process. The new setup comprises a steel pipe with an inner diameter of 12 mm and an outer diameter of 15 mm. Additionally, aluminum shafts with an inner diameter of 15 mm and an outer diameter of 35 mm are used for reinforcement, pressed around the steel pipe. One of the holes in the setup is closed using an M14 screw, while from the other side, a rod that fits inside the steel tube is used to compress the ALP. Finally, two screws are employed on the side of the tube to securely hold the pressure and prevent any movement of the rod. The setup can be found in Figure 8. Additionally, the melting process was conducted at a temperature of 230 degrees Celsius for two hours. The adjustment in temperature was made based on the observation that certain internal flakes were not completely melted, while the external layer exhibited a smooth, non-burned surface.

In Table I, the variations among the different high-pressure melting setups can be found. These details include information on the material casing, the diameter of the ALP cylinder, pressure comparisons between the setups, oven temperature, and the duration each setup spent in the oven.

TABLE I
DIFFERENCE IN THE HIGH-PRESSURE MELTING SETUPS

	High-pressure Setup 1	High-pressure Setup 2	High-pressure Setup 3
Material casing	Galvanized Steel	Steel	Steel and Aluminium
Diameter ALP cylinder	36 millimeter	10 millimeter	12 millimeter
Pressure comparison	Lowest	Intermediate	Highest
Oven temperature	210 °C	220 °C	230 °C
Time in oven	100 minutes	90 minutes	120 minutes

F. Spring High-Pressure Melting

Following the unsuccessful attempts with the previous setups, a new setup was designed to produce ALP blocks as originally intended in the initial melting setup, which can be found in Figure 9. The desired blocks will have dimensions of 94x94x10 mm, necessitating approximately 90 grams of ALP. The newly designed setup includes a spring mechanism that exerts pressure on the ALP throughout the melting process. Compression in millimeters can be assessed by measuring the height of the upper section of the setup. An improvement in this setup is the reduced contact surface between the block and the walls, which facilitates the retrieval process. This addresses a previous flaw encountered in the testing setup in Figure 8, making it easier to extract the ALP block from the setup. Additionally, the melting process was conducted at a consistent temperature of 230 degrees Celsius for 1 hour and 15 minutes, aligning with the temperature used in the last high-pressure molding setup.

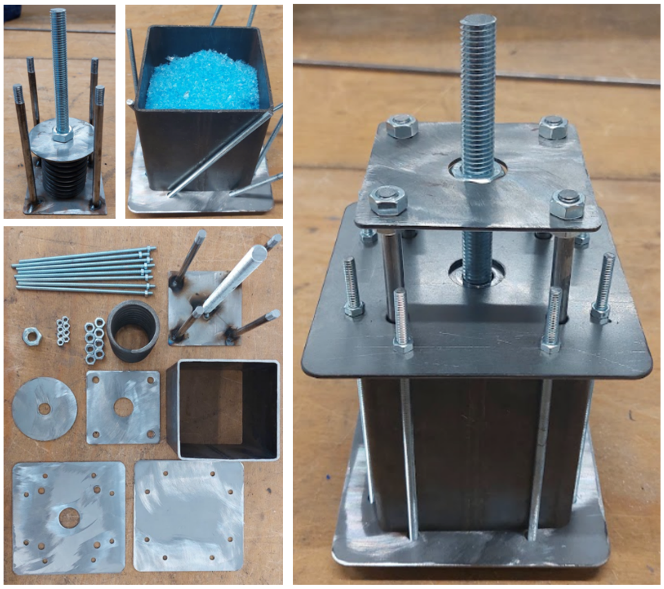

Fig. 9. Final Melting Setup with Spring. Top Left: Spring Preloading and 90g of ALP Flakes in the Melting Chamber. Bottom Left: Individual Components Displayed. Right: Fully Assembled Configuration.

G. *Tensile test*

For the tensile testing, three strips were prepared from the ALP block, which would be utilized to create dogbone-shaped samples adhering to the ASTM D638-V standard for tensile testing [12]. Initially, the block was divided into three equal pieces. These pieces were then processed in a milling machine to ensure their straightness and achieve a uniform height of 4 mm.

Following the milling process, the three pieces underwent further refinement in a laser cutter to precisely shape them into the characteristic dogbone configuration. This was achieved using a cutting speed of 12 and a power setting of 95%. Once the specimens were appropriately shaped, they were subjected to a tensile test using a Zwick Z010 tensile testing machine and subsequently analyzed with the testXpert software. The tests were conducted with a low tensile rate of 2 mm/min, and no preload was applied.

III. RESULTS

This section presents the study's results, organized into subsections, each addressing a specific research aspect. The first subsection shows the results of the collection process. The next subsection shows the X-ray diffraction test results, revealing material composition. The subsequent subsection covers the melting process outcomes. Then, the shredding and injection molding results are detailed. Following that, the findings from the high-pressure melting setups are presented. The next subsection shows the results from the spring high-pressure melting setup. The last section shows the results of the tensile testing on dogbone samples, shedding light on the mechanical properties and behavior of the recycled ALP.

Fig. 10. Left; Different colours ALP from Greencycl, Right; Blue ALP from Ikazia Hospital

TABLE II
WEIGHT PER CATEGORY COLLECTED ALP

Colour ALP	Pure ALP or not	Collected from	Weight
Blue	Pure	Greencycl	375 gram
Blue	Pure	Ikazia	293 gram
Blue	With labeling	Ikazia	874 gram
White	Pure	Greencycl	82 gram
Green	With labeling	Greencycl	173 gram

A. *Collecting process of ALP*

The ALP obtained from Greencycl was available in three different colors: white, green, and blue, as shown left in Figure 10. On the other hand, the ALP collected from Ikazia Hospital was exclusively blue, as depicted right in Figure 10.

A total of approximately 1.8 kg of ALP was collected, with weights distributed among different categories as shown in Table II.

B. *Results X-ray diffraction test*

In Figure 11, 12, 13, and 14, the results of the XRD analysis of the green ALP, blue ALP, white stiff ALP, and white flexible ALP can be found, respectively. Figures 11, 12, 13, and 14 show the measured XRD patterns in black, after background subtraction. The colored sticks give the peak positions and intensities of the possibly present crystalline phases, using the ICDD pdf4 database, see Table III.

According to the XRD graphs, it was observed that the green (Figure 11), blue (Figure 12), and white stiff ALP (Figure 13) samples consist of a-polypropylene (C3H6)n and carbon nanotubes (C). The peaks present in the XRD graph closely

TABLE III
COMPOUND PER TESTED SAMPLE

Sample	Compound	
Green ALP	$\alpha - Polypropylene$	(C3H6)n
	Carbon Nanotubes	C
Blue ALP	$\alpha - Polypropylene$	(C3H6)n
	Carbon Nanotubes	C
White stiff ALP	$\alpha - Polypropylene$	(C3H6)n
	Carbon Nanotubes	C
White flexible ALP	High Density Polyethylene	(C2H4)n

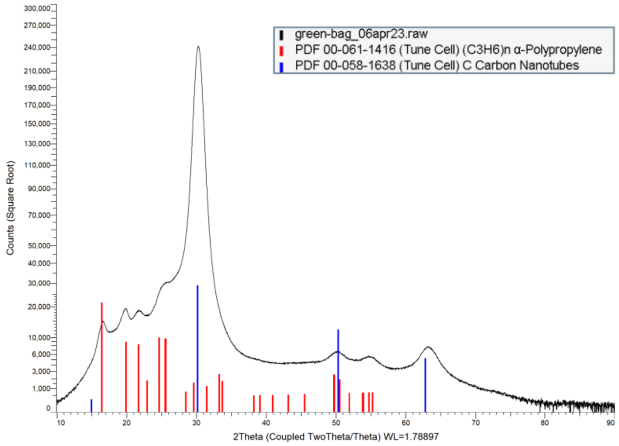

Fig. 11. XRD analysis of green ALP

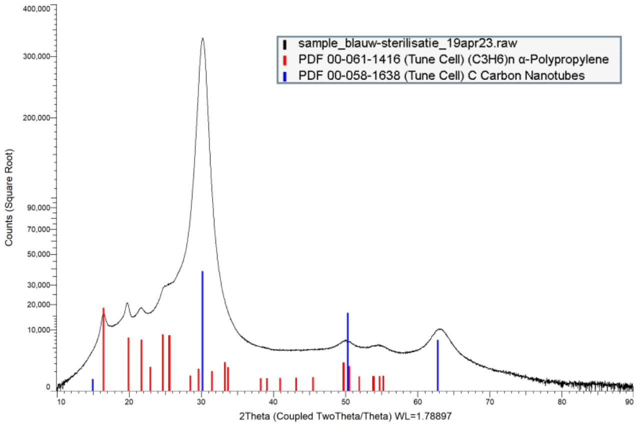

Fig. 12. XRD analysis of blue ALP

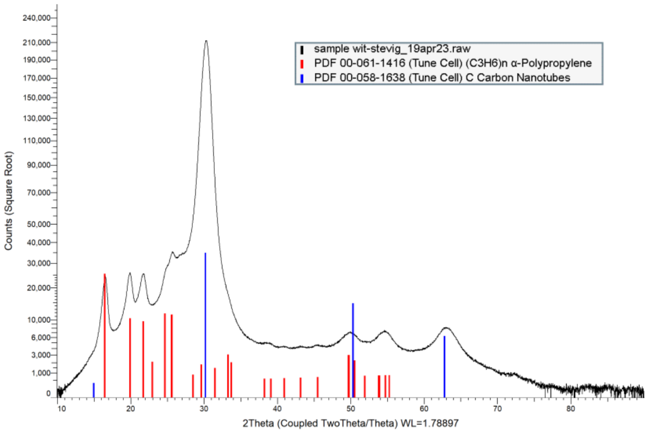

Fig. 13. XRD analysis of white stiff ALP

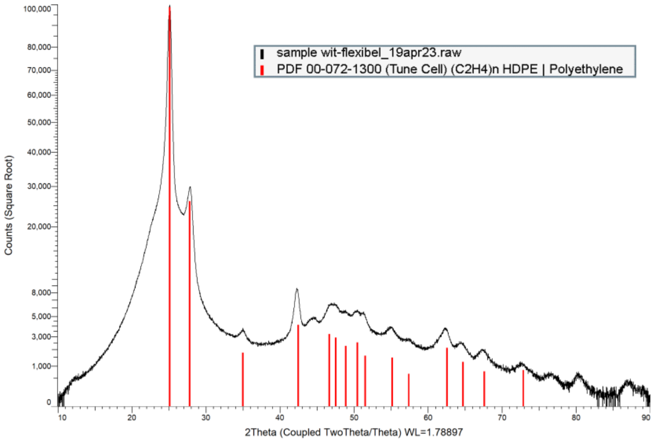

Fig. 14. XRD analysis of white flexible ALP

correspond to the peaks observed in the a-polypropylene $(C_3H_6)_n$ and carbon nanotubes (C) data available in the ICDD pdf4 database. This suggests the presence of these materials in the respective ALP samples. On the other hand, the XRD graph of the white flexible ALP (Figure 14) shows a match with the peaks observed in high-density polyethylene $(C_2H_4)_n$. This indicates that the white flexible ALP consists of high-density polyethylene. This study exclusively utilizes ALP composed of polypropylene with carbon nanotubes.

C. Results melting process

Various melting processes were attempted to identify the optimal recycling method for autoclave laminated plastic (ALP). The different variables of the various trials are presented in Table IV and the corresponding images per melting trial can be found in Appendix; Figure 24.

During the trials, it was observed that the ALP exhibited challenges in the melting process, often resulting in incomplete or partial melting and occasional charring. In the first trial, after a duration of 45 minutes, the ALP showed partial melting, indicating potential for recycling of this material. Trials 2, 3, and 4 were conducted sequentially using the same ALP sample. However, it was observed in trials 2 and 3 that the ALP did not melt as effectively as in the initial trial, even with extended heating times. In trial 4, the temperature was increased to $300°C$, resulting in complete melting; however, the outcome was a charred block of ALP. For trial 5, a new set of materials was used, and a temperature of $300°C$ was employed. After 50 minutes at this temperature, approximately half of the plastic had melted, but the resulting plastic block exhibited some charring. In trial 6, a new melting hopper was utilized without a filter, and pressure was applied using a large block of metal. At a temperature of $250°C$, the ALP did not melt, but after some time at $300°C$, it exhibited charring along the sides of the hexagon shape, while the bottom portion melted, leaving the top slightly deformed but intact. Trial 7 was conducted in a different oven known as the Sterimelt. A piece of compressed ALP was placed in the oven at $260°C$, but after 1 hour, it was completely charred. In the final

TABLE IV
MELTING TRIALS DATA

Trial	Category ALP	Weight	Tbegin	Tend	Time
1	Blue Clean	195 gram	$100°C$	$316°C$	45 min
2	Blue Clean	196 gram	$50°C$	$270°C$	60 min
3	Blue Clean	196 gram	$180°C$	$270°C$	60 min
4	Blue Clean	196 gram	$300°C$	$280°C$	45 min
5	Green Clean	140 gram	$320°C$	$300°C$	50 min
6	Blue Clean	239 gram	$250°C$	$300°C$	100 min
7	Blue Clean	60 gram	$20°C$	$260°C$	60 min
8	ALP flakes	60 gram	$200°C$	$220°C$	120 min

Note.- Tbegin means the temperature the melting hopper went into the oven and Tend means the temperature the melting hopper went out the oven

Fig. 15. Left; Compressing of ALP, Right; Shredded ALP

Fig. 16. ALP melted manually in the Babyplast injection molding machine

Fig. 17. Recycled plastic using high-pressure molding setup 1

trial (Trial 8), shredded ALP was combined with a hexagon melting base. In this particular trial, a lower temperature was employed, approximating the glass transition temperature of ALP. Consequently, the ALP flakes fused together, forming a hexagonal plate. However, it was observed that the individual flakes were still discernible, indicating incomplete melting. These trials highlight the challenges encountered in achieving successful and consistent melting of ALP during the recycling process.

D. Result shredding and injection molding process

During the melting process, it became evident that the ALP posed more challenges compared to materials like blue wrapping paper [6]. Recognizing the difficulties encountered, an alternative approach was explored, bypassing the melting process and directly transforming the ALP into granules. Considering the stiffness of the ALP, it was hypothesized that shredding the material into flakes might be a feasible solution. These flakes could then be utilized in an injection molding machine, offering a potential pathway for further processing and utilization of the ALP waste. Due to the low mass of individual ALP sheets, the blades encountered difficulties in gripping and shredding them effectively. To address this issue, a solution was devised by compressing the ALP in a metal bowl, increasing its overall mass, see Figure 15. This increased mass facilitated easier shredding of the ALP sheets. The shredded material went through the shredder three times to ensure thorough processing. The final result, in the form of flakes, can be observed in Figure 15.

The shredded material was then fed into the BabyPlast injection molding machine. However, due to the small mass of the ALP flakes, the machine was unable to effectively process the flakes provided. An alternative approach involving manual placement of the flakes into the plunger was attempted; however, this resulted in an insufficient volume of plastic to adequately fill the dogbone mold. The outcome of the manual process can be found in Figure 16. This presented a significant challenge, as it prevented the creation of dogbone samples using the injection molding process.

E. High-pressure Melting

The first high-pressure melting setup showed promising results when the compressing rod was removed. In order to recover the plastic cylinder, the test setup was cut at the welding point, allowing for the successful retrieval of the block. Detailed results can be found in Figure 17.

Nevertheless, upon closer observation of the recycled ALP cylindrical block, it becomes evident that the block retained some degree of porosity. The presence of porosity in the recycled ALP cylindrical block poses challenges when conducting a reliable tensile test on the specimens. The block displayed promise, achieving a fully solid block without any porosity necessitated the application of higher pressure. To address this limitation, a new second setup was developed, as described in Method Section E, which allowed for the creation of tensile specimens with enhanced integrity.

During the pressing process of the ALP into the tubes, the pressure became too high, causing the tubes to deform. However, despite this issue, it was still possible to successfully melt the flakes in the oven and produce recycled ALP rods. Unfortunately, retrieving the plastic rods proved challenging due to the deformations in the tubes. However, after opening up the tubes, the rods were successfully retrieved. The results

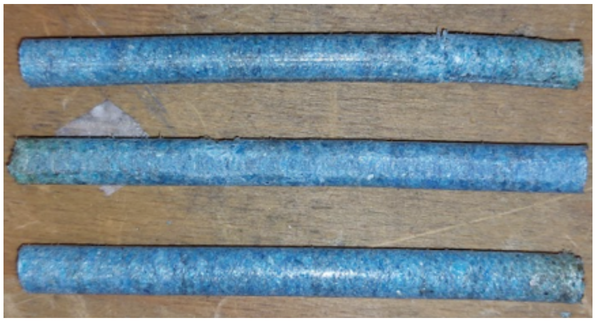

Fig. 18. Recycled plastic using high-pressure molding setup 2

Fig. 19. Recycled plastic using high-pressure molding setup 3

of the second high-pressure setup can be observed in Figure 18.

During visual inspection, the rods appeared to be non-porous and suitable for the creation of tensile test specimens. However, upon closer examination, it became evident that a significant portion of the rods exhibited substantial weakness, as they could be easily broken by applying even a small amount of force or moment. This unexpected fragility of the rods highlights a critical flaw in their structural integrity, indicating potential challenges in conducting accurate and reliable tensile tests using these specimens.

To address the issues encountered with the previous setups, a third high-pressure setup was developed, as outlined in Method Section E. The results of this melting setup can be found in Figure 19.

The final high-pressure melting test initially showed promising results after removing the closure bolt. However, retrieving the ALP rod revealed an unexpectedly high-pressure requirement, causing the steel rod meant for extraction to buckle. Despite numerous attempts, the setup was ultimately cut but only a small part of the ALP rod came free. Regrettably, the recycled ALP did not meet the desired properties, marking a significant setback in achieving the intended outcome.

F. Spring High-Pressure Melting

During the melting process of the final setup, the compression of the spring and the temperature of the oven were monitored at 5-minute intervals. Figure 20 illustrates the changes in spring force and oven temperature over time, providing insights into the behavior of the setup during the melting process. The resulting ALP block from the spring high-pressure setup had a hard outer layer and showed little porosity. This ALP block can be observed in Figure 21.

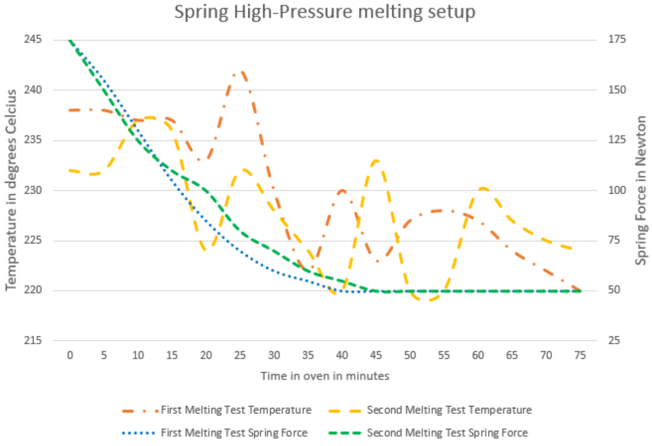

Fig. 20. Change in spring force and oven temperature over time

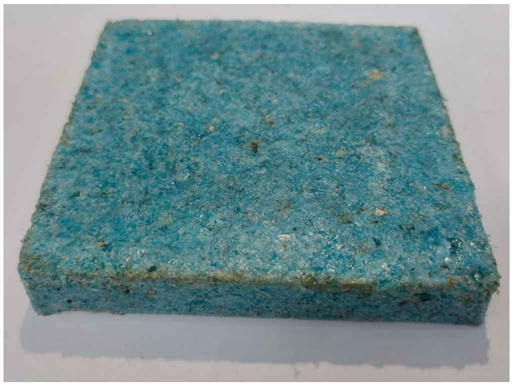

Fig. 21. Recycled ALP using the spring high-pressure molding setup

G. Tensile test

For the tensile testing, six dogbones were initially prepared. Regrettably, specimen 6 inadvertently slipped out of the clamp during the test and had to be excluded from the analysis. Consequently, five successful tensile tests were conducted, providing valuable data. The testing phase is visually depicted in Figure 22. This figure provides an overview of the setup, displays the various broken specimens, and offers a close-up view of the fracture lines on the dogbones of specimen 1.

The results of the tensile tests are presented in Figure 23, which displays five stress-strain curves obtained from five different dogbones, all of the same dimensions, and extracted from the same ALP block. In Table V, the values for Young's modulus and ultimate tensile stress per specimen can be found. The average Young's modulus measures 269 MPa, with a standard deviation of 85 MPa. For the ultimate tensile stress, the average value is 2.9 MPa, with a standard deviation of 0.5 MPa. Regarding the strain at break, the average value is 25%, with a standard deviation of 6%. These values provide insights into the material's mechanical behavior.

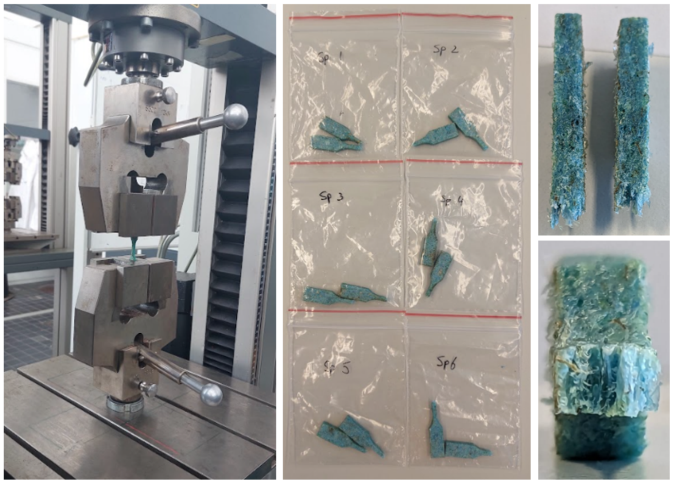

Fig. 22. Left; Tensile testing of ALP dog bone, Middle; Broken specimens, Right; Close-ups of the break line

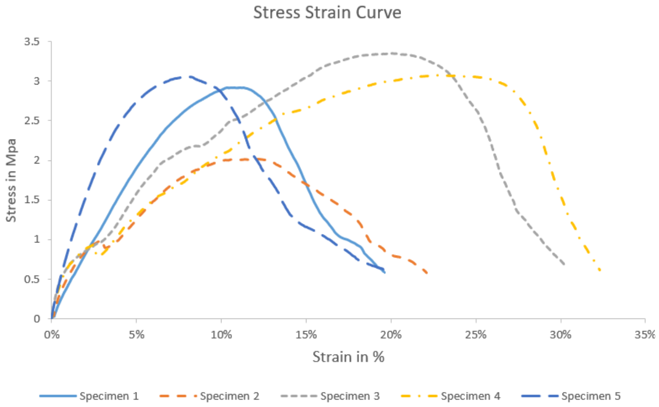

Fig. 23. Stress strain curves of five type D638-V specimens

TABLE V
YOUNG'S MODULUS, ULTIMATE TENSILE STRESS AND STRAIN AT BREAK OF DIFFERENT SPECIMENS

Specimen	**Young's Modulus in Mpa**	**Ultimate Tensile Strength in Mpa**	**Strain at break in %**
1	226	2.9	20
2	244	2.0	22
3	208	3.4	30
4	248	3.1	32
5	419	3.1	20
Average	269 ± 85	2.9 ± 0.5	25 ± 6

IV. DISCUSSION

The results of the research on the recycling of ALP have revealed several significant challenges and limitations. The aim of the study was to explore the feasibility and effectiveness of mechanical recycling as a potential solution for recycling of ALP. After several attempts utilizing different setups and techniques, satisfactory outcomes were eventually realized. The findings from the spring high-pressure melting approach have demonstrated promising potential for the mechanical recycling of ALP.

A. *Collecting process of ALP*

The collected ALP primarily consisted of blue ALP, notably with labeling from Ikazia. Extensive cleaning efforts were undertaken within this category, involving the removal of stickers and paper from the ALP. As a result, the weight of this ALP category is lower than what is indicated in Table II.

For future collections, it is crucial to ensure no ink, stickers, or paper residue remains on the ALP. Leftover inks can affect its appearance, introduce defects, and produce unwanted byproducts [13]. To prevent this, it is advisable to refrain from writing on ALP, or implement a de-inking process in mechanical recycling. Labeling stickers applied to medical sterilization pouches change color upon successful sterilization and are affixed to the packaged instruments before they are sterilized. These stickers are placed on the plastic side to facilitate unobstructed steam penetration into the medical sterilization pouch [14], [15]. While the use of labeling stickers is necessary, it is crucial to establish effective methods for their proper separation from the ALP during the collection process.

B. *X-ray diffraction test*

The x-ray diffraction tests confirmed the presence of polypropylene and carbon nanotubes in the ALP. Figures 8, 9, and 10 displayed consistent patterns, regardless of ALP color variations. These results align with previous literature [8], which highlighted common materials in medical sterilization pouches, including polypropylene and polyester. Due to limited quantities of white flexible ALP and the literature study's findings, it was excluded from the experiments. Instead, the focus remained on stiffer ALP samples, consistent with typical materials found in medical sterilization pouches.

Regarding the XRD graphs of polypropylene carbon nanotubes ALP, it is worth noting that the accuracy of colored sticks representing peak positions and intensities for potential crystalline phases was relatively lower compared to the white polyethylene plastic graph. Peak intensity in XRD graphs is influenced by various factors, including crystal structure, orientation, crystallite size, and material quantity. These variations can result in differences in peak intensities, even for the same crystallographic planes. Thus, while peak positions align, differences in intensity between peaks can be expected in XRD graphs [16].

C. *Melting process*

The constructed melting hopper met most requirements but fell short in terms of reusability and ease of plastic retraction. After multiple melting trials, the hopper's reusability declined due to clamp fitting issues between its components. Additionally, a small amount of plastic residue remained after each trial, contaminating subsequent runs. Retracting the plastic block proved more challenging than initially anticipated, mainly because the block adhered to the closing tub. The raised edges of the tub further complicated block removal, although it remained feasible. Despite these challenges, the hopper performed well in meeting all other requirements.

Multiple trials were conducted for the melting process with the objective of producing a solid block of recycled plastic. However, these trials encountered various challenges. The first issue encountered was the failure of ALP to melt properly, impeding the formation of a cohesive block. Additionally, there were instances where the ALP material charred during the melting process, further hindering the desired outcome.

The behavior of ALP, which consisted of polypropylene and carbon nanotubes, differed from that of polypropylene wrapping paper discussed in Van Straten's (2021) paper [6]. In the aforementioned research, polypropylene was observed to melt at temperatures ranging from 200 to 300 degrees Celcius. However, during the melting of ALP, a temperature of $250°C$ was insufficient to achieve melting, preventing the material from passing through the filter. Moreover, using a temperature of $300°C$ often resulted in the formation of charred blocks of plastic, rendering them unsuitable for recycling with the created melting hopper.

The charring of ALP can be attributed to thermal degradation processes within the polymer matrix. As the ALP was subjected to temperatures around 300 degrees Celsius, vapor emissions indicated the occurrence of thermal degradation. The presence of carbon nanotubes in ALP, known for their high thermal conductivity [17], can influence heat distribution and affect the thermal degradation process. In contrast to the experimental configuration employed by Van Straten (2021) [6], the melting hopper lacked complete airtight sealing, permitting air interaction with the ALP inside the melting hopper, which potentially contributed to the formation of char.

D. Shredding and injection molding

Shredding or mechanically breaking down plastic items can result in the production of smaller plastic fragments, including microplastics and potentially nanoplastics. Improper handling of these plastic fragments can lead to negative environmental consequences, as demonstrated in a study by Suzuki et al. (2022) [18], where mechanical recycling without appropriate wastewater treatment may result in the emission of microplastics into the environment. Nevertheless, when micro and nanoplastics are managed properly during mechanical recycling, the potential environmental consequences can be mitigated.

During the injection molding process, it is crucial to have an adequate amount of material to ensure optimal flow and complete filling of the mold cavities. However, in this case, the bouncing motion in the hopper prevented the granulated ALP material from being effectively fed into the injection molding machine. This issue disrupted the smooth and continuous flow of the material, hindering the injection molding process. Fortunately, the combination of polypropylene and carbon nanotubes did not cause any complications during the melting process when manually inserted into the Babyplast injection molding machine. A temperature of 210 degrees Celsius was found to be adequate for effective melting. This step ensures that the ALP material is in a suitable form for subsequent injection molding, allowing for improved flow and filling of the mold cavities.

E. High-pressure molding

Following the injection molding trial, three consecutive high-pressure melting setups were established. The first setup served as a trial to determine ALP's response to high pressure. However, its design lacked reusability, requiring the cutting of the cylindrical block. Additionally, due to the dimensions of the created block of ALP, it was not possible to produce dogbones from it. The second setup was designed to meet all design requirements but faced challenges in terms of reusability and easy retraction due to issues with the compressing tubes. The dimensions of the ALP rods allowed for the creation of circular dogbones. However, the material's structure posed challenges during post-processing, preventing the successful production of dogbones. The third setup, with an increased wall thickness compared to the second, aimed to prevent nodding of the compressing tube and fulfill all criteria. Unfortunately, it failed in terms of plastic retraction after the melting trial, consequently falling short in reusability and dogbone production as well.

The three high-pressure melting setups showed promising results for ALP cylindrical blocks and rods. They displayed a hard, almost non-porous outer layer. However, when cutting or breaking the recycled ALP, there was found greater porosity inside, hinting at an intriguing structural complexity. This porosity renders the parts unsuitable for post-processing use. The porosity can affect the structural integrity and mechanical properties of the material, potentially leading to inaccurate test results.

F. Spring High-Pressure Melting

After two melting trials, it became evident that the design of the spring high-pressure melting setup met all the specified requirements. The retraction of the ALP block using the Spring High-Pressure melting setup proved more effective compared to previous high-pressure melting configurations. Disassembling the setup revealed that the ALP block could be easily retracted. Furthermore, even after undergoing two melting trials, the setup remained unaltered, demonstrating its reusability and robustness. Much like the outcomes of the high-pressure molding experiments, the recycled ALP block exhibited a hard and non-porous exterior. However, intriguingly, the plate demonstrated a more robust internal structure, as evidenced by its resistance to manual breakage. This resistance was particularly noteworthy given that an 11-millimeter plate could not be broken by bare hands. Cutting the ALP block into three strokes revealed that the individual ALP flakes remained distinguishable, and full melting of all the flakes had not occurred. Nevertheless, the structural integrity was significantly improved compared to the high-pressure molding method, particularly in terms of reduced porosity. This observation was also evident during the smooth milling process. Subsequently, there were no issues encountered during the laser cutting of the 4 mm thick plates.

In Table VI, a comprehensive summary is presented, evaluating the extent to which each setup met the established requirements. Ratings are assigned on a scale that includes the following categories: Excellent, Good, Fair, Poor, and Bad. Notably, the first high-pressure setup is excluded from this table, as it primarily served as a proof-of-concept for assessing the feasibility of high-pressure melting.

TABLE VI
REQUIREMENTS FULFILLMENT PER SETUP

Requirements	**Melting Hopper**	**High-pressure Setup 2**	**High-pressure Setup 3**	**Spring high-pressure setup**
Dimensions	Excellent	Excellent	Excellent	Excellent
Reusability	Fair	Poor	Poor	Excellent
Dogbones	Poor	Poor	Poor	Good
Retraction ALP	Poor	Bad	Bad	Excellent
(Dis)assembly	Excellent	Good	Excellent	Good
Fabrication	Excellent	Good	Good	Excellent
Costs	Excellent	Excellent	Excellent	Excellent
Safety	Good	Good	Good	Good

G. Tensile test

The results of the five tensile tests exhibited notable differences among them. Specifically, when considering Young's modulus, specimens 1 to 4 showed values within a relatively narrow range of 200 MPa to 250 MPa, whereas specimen 5 displayed a significantly higher value, approximately twice that of the other four specimens. In terms of ultimate tensile strength, specimens 1, 3, 4, and 5 demonstrated values falling in the range of 2.9 MPa to 3.4 MPa, while specimen 2 exhibited a slightly lower value, approximately 1 MPa less than the others. Regarding strain at break, two specimens fractured at approximately 30%, while three specimens ruptured at around 20%.

These variations in the mechanical properties among the specimens are likely attributed to the incomplete melting of ALP flakes during the recycling process.This is clearly evident in Figure 22, where distinguishable flakes within the material can be observed. This inherent heterogeneity in the material's structure can lead to divergent mechanical properties from one specimen to another. Another potential explanation is that the dogbone broke due to a small air bubble present in the breaking line. Another important aspect to highlight is that the laser cutting of the ALP resulted in the remelting of the dogbone's edges, subsequently affecting the material's properties during the tensile testing.

When comparing these findings with those from a study conducted by Bikiaris et al. in 2010 [19], where polypropylene was blended with carbon nanotubes and subsequently subjected to tensile tests, it becomes apparent that the mechanical properties of the recycled ALP are notably lower. For polypropylene with 0.5% carbon nanotubes, Bikiaris reported a Young's modulus of approximately 1250 MPa, an Ultimate Tensile Strength of approximately 31 MPa, and a strain at break of approximately 150%. These disparities highlight the need for further refinement and optimization in the recycling process to approach the mechanical properties achieved in Bikiaris' research. However, with the spring high-pressure method of recycling ALP a possible application for this material can be to make acoustic panels to hang up in working rooms. The low weight of the material, the smooth outer surface, and the slight porosity where air is captured can be great for such an application.

H. Future work

This study marks a promising starting point for the recycling of ALP. However, optimal recycling of ALP requires further extensive research. Moving forward, there are numerous potential paths for further research.

Firstly, a key research avenue involves finding a way to recycle ALP into a non-porous and homogeneous material. This objective could be achieved by granulating the ALP blocks into dense granules suitable for injection molding. The promising visual results obtained from the Babyplast injection molding test indicate that this approach may yield a material with non-porous and homogeneous characteristics and so improved mechanical properties. Another approach to achieving a more homogeneous structure is by processing ALP sheets using alternative recycling technologies, such as the S:GRAN system by Next Generation Recycling (NGR) [20]. This system combines shredding, feeding, and extrusion, which may offer improved material homogenization during the recycling process.

Secondly, it is worthwhile to explore the optimization of spring high-pressure molding parameters. The effects of varying parameters such as pressure, temperature, and melting time can be investigated in terms of their impact on the mechanical properties and quality of the recycled ALP material. This research would aim to determine the optimal process conditions for achieving desired properties and product outcomes.

Next, considering comprehensive characterization tests for the recycled ALP is important. These tests should cover various physical, mechanical, and thermal properties using suitable testing methods. This comprehensive data will enable a thorough comparison between the recycled ALP and the original material, facilitating a complete assessment of its performance and potential applications.

Additionally, potential areas for future research encompass the redesign of medical sterilization pouches and the optimization of the collection process. The ALP collected from Ikazia Hospital frequently contained medical-grade paper adhered to the plastic part. Investigating alternative adhesives for securing the medical-grade paper to the ALP could lead to improvements in the recyclability of medical sterilization pouches. Also, the removal of labeling stickers from these pouches proved to be challenging, resulting in leftover residues. Investigating the use of easily removable stickers or exploring alternative methods for labeling medical sterilization pouches could offer a solution to this issue. To ensure a consistent supply of clean ALP, developing a standardized collection process that can be employed across diverse medical institutions is highly advantageous. Such a process would streamline the collection of uncontaminated ALP material, facilitating more effective recycling procedures.

Furthermore, future work can involve upscaling the spring high-pressure molding process and integrating it into an entire recycling process chain for ALP. While the current block of recycled ALP has a relatively small volume of approximately 110 cm^3, a larger setup can be developed to produce larger blocks of ALP. This would enable the utilization of the recycled ALP in further processing steps or in acoustic panels. Additionally, conducting a cost analysis can assess the economic viability of large-scale ALP recycling, while a lifecycle assessment can evaluate its environmental impact.

By addressing these future research areas, it is possible to further advance the field of ALP recycling and develop sustainable solutions for the recovery of this plastic material.

I. Study limitations

This study has demonstrated the feasibility of recycling ALP through spring high-pressure melting. However, it is important to acknowledge the limitations encountered throughout the research process.

One significant limitation of the study is the narrow focus on ALP material derived from medical sterilization pouches made specifically from polypropylene with carbon nanotubes. It is crucial to recognize that different types of plastics are utilized in medical sterilization pouches, and their recycling behavior may vary.

Additionally, the ALP collected from the Ikazia hospital contained pollutants such as stickers and paper, which may impact the quality of the final product and the tensile testing results, especially when the dogbones were not composed of 100% pure polypropylene with carbon nanotubes.

The next limitation was encountered during the creation of the dogbones. The laser-cutting process resulted in the re-melting of the edges, leading to hardened sides of the dogbones. This may have positively influenced the tensile test results by potentially achieving higher mechanical properties. For future dogbone production, techniques such as injection molding are preferred.

Another limitation of the study was associated with the use of the melting setups. The initial setup, which included a filter, was reused multiple times throughout the experiments. Consequently, the setup became contaminated and proved challenging to thoroughly clean between each recycling cycle. This contamination and cleaning difficulty may have resulted in residual material from previous melting cycles being present during subsequent recycling attempts. Furthermore, the high-pressure melting setup employed in the study could only be used for a single cycle. Once the melting process was completed, the setup had to be disassembled to retract the recycled ALP block. This limitation restricted the ability to conduct a larger number of recycling trials. However, this issue did not arise with the spring high-pressure melting setup, allowing for multiple melting trials to be conducted.

Lastly, a limitation encountered during the melting process was the oven's inability to consistently maintain the desired temperature. The temperature often exceeded the intended point, requiring the lid to be opened to cool down the oven. This fluctuation in oven temperature is also evident in Figure 20. This inconsistency in temperature control may potentially impact the melting process and the quality of the recycled material. Future research could benefit from employing a more precise and reliable oven to ensure better temperature regulation and more consistent melting results.

V. CONCLUSION

In conclusion, the research on recycling autoclave-laminated plastic (ALP) has provided valuable insights into the potential of mechanical recycling. Despite encountering challenges during the melting and injection molding processes, the study has demonstrated promising potential for spring high-pressure molding in the recycling of ALP. Through X-ray diffraction tests, it was determined that the components of most of the collected ALP material were polypropylene and carbon nanotubes. The first melting experiments revealed that while a temperature of $250°C$ was insufficient for complete melting, temperatures of $300°C$ showed better melting results but with charring. Additionally, the immediate grinding of compressed ALP sheets for injection molding resulted in ALP flakes with a low mass, rendering them unsuitable for complete processing in a Babyplast injection molding machine. However, the Babyplast injection molding machine showed that the ALP could be melted at $210°C$ with additional pressure. Hereafter, the high-pressure melting tests using ALP flakes at $210°C$ - $230°C$ produced various blocks of plastic, demonstrating the feasibility of this recycling method. Subsequently, a new high-pressure melting setup using a spring for continuous pressure was developed. With this setup, a block of ALP was created and eventually processed with milling and laser cutting to dogbones. Hereafter, the dogbones underwent a tensile test which showed that the recycled ALP had an average Young's Modulus of 269 Mpa. Ultimately, the chosen mechanical recycling approach for ALP comprised shredding and subsequent high-pressure melting, facilitated by a spring mechanism. A possible application for recycled ALP is acoustic panels, due to the low weight, smooth outer surface, and slight porosity. These findings constitute a meaningful step towards advancing sustainability in the medical recycling industry, providing a solid foundation for ongoing research and optimization of ALP recycling methods.

ACKNOWLEDGEMENT

I thank the following individuals for their expertise and assistance throughout all aspects of this study:

- B. van Straten, T.J. Horeman and H.F.J. Lieshout, for their overall assistance and guidance throughout the research process.
- B.I. Pluijmers, for their support in setting up the collection of ALP at the oral surgery department at Ikazia Hospital, Rotterdam.
- R.W.A. Hendrickx, for conducting the X-ray diffraction analysis.

– P.J.W. van Nimwegen from Model Engineering, for their expertise and assistance during the injection molding process.
– The team from the IWS at 3ME, TU Delft, for their support during the manufacturing of the various setups.
– The team from the Mechanical Behaviour lab at 3ME, TU Delft, for their support during the tensile test experiments.

REFERENCES

[1] M. R. Capoor and K. Bhowmik, "Current Perspectives on Biomedical Waste Management: Rules, Conventions and Treatment Technologies," *Indian Journal of Medical Microbiology*, vol. 35, pp. 157–164, 4 2017.
[2] S. Leissner and Y. Ryan-Fogarty, "Challenges and opportunities for reduction of single use plastics in healthcare: A case study of single use infant formula bottles in two Irish maternity hospitals," *Resources Conservation and Recycling*, vol. 151, p. 104462, 12 2019.
[3] S. Ing, "It's a Wrap! Everything You Need to Know About Sterilization Pouches - Medicom," 7 2021.
[4] T. P. Company, "All About Autoclave Bags - Types, Industries and Selection Criteria," 2023.
[5] E. F. De Ridder, H. J. Friedericy, A. C. Van Der Eijk, J. Dankelman, and F. W. Jansen, "A New Method to Improve the Environmental Sustainability of the Operating Room: Healthcare Sustainability Mode and Effect Analysis (HSMEA)," *Sustainability*, vol. 14, p. 13957, 10 2022.
[6] B. H. Van Straten, D. J. Van Der Heiden, P. Robertson, C. Riekwel, F. Jansen, M. Van Der Elst, and T. Horeman, "Surgical waste reprocessing: Injection molding using recycled blue wrapping paper from the operating room," *Journal of Cleaner Production*, vol. 322, p. 129121, 11 2021.
[7] R. Faesal and W. Bdaiwi, "Recycling of Waste Medical Plastic Syringes in Manufacturing Low-Cost Structural Sections," *Materials Science Forum*, vol. 1050, pp. 115–123, 1 2022.
[8] M. Hansen, "Recycling of Autoclave-Laminated Material: An Analysis of Waste Management Techniques and Recycling Processes," *MSc Thesis Literature Study*, pp. 1–29, 3 2023.
[9] A. Dehal, A. N. Vaidya, and A. R. Kumar, "Biomedical waste generation and management during COVID-19 pandemic in India: challenges and possible management strategies," *Environmental Science and Pollution Research*, vol. 29, pp. 14830–14845, 10 2021.
[10] Y. Q. Gill, M. Khurshid, U. Mehmood, M. Irfan, and F. Saeed, "Upscale recycling of nonwoven polypropylene waste using a novel blending method," *Journal of Applied Polymer Science*, vol. 139, 7 2022.
[11] J. Patil, H. H. Patil, R. Sankpal, D. Rathod, K. Patil, P. R. Kubade, and H. B. Kulkarni, "Studies on mechanical and thermal performance of carbon nanotubes/polypropylene nanocomposites," *Materials Today: Proceedings*, vol. 46, pp. 7182–7186, 1 2021.
[12] ASTM, "Standard Test Method for Tensile Properties of Plastics," 7 2022.
[13] RecyclingInside, "Ink removal in plastics recycling plants," 11 2021.
[14] N. C. f. C. D. P. Division of Oral Health and H. Promotion, "Sterilization: Packaging storage — FAQs — Infection control — Division of Oral Health — CDC," 2 2016.
[15] I. O. for Standardization, "Sterilization of health care products — Chemical indicators," Tech. Rep. ISO 11140-1:2005(E), 7 2005.
[16] N. Iwashita, C. R. Park, H. Fujimoto, M. Shiraishi, and M. Inagaki, "Specification for a standard procedure of x-ray diffraction measurements on carbon materials," *Carbon*, vol. 42, pp. 701–714, 1 2004.
[17] D.-K. Lee, J. Yoo, H. W. Kim, B.-H. Kang, and S. Park, "Electrical and thermal properties of carbon nanotube polymer composites with various aspect ratios," *Materials*, vol. 15, p. 1356, 2 2022.
[18] G. Suzuki, N. Uchida, L. H. Tuyn, K. Tanaka, H. Matsukami, T. Kunisue, S. Takahashi, H. V. Pham, H. Kuramochi, and M. Osako, "Mechanical recycling of plastic waste as a point source of microplastic pollution," *Environmental Pollution*, vol. 303, p. 119114, 6 2022.
[19] D. N. Bikiaris, "Microstructure and properties of Polypropylene/Carbon Nanotube Nanocomposites," *Materials*, vol. 3, pp. 2884–2946, 4 2010.
[20] "Plastic Recycling Technology — Waste Recycling Plant Equipment — NGR."

APPENDIX

Trial 1

Trial 2

Trial 3

Trial 4

Trial 5

Trial 6

Trial 7

Trial 8

Fig. 24. Images taken from the different melting trial (First picture of Trial 6 - ALP after 50 min in an oven at 250 degrees Celcius. Second picture of Trial 6 - Partially melted ALP after 50 min in an oven at 300 degrees Celcius.)