Ref. Ares(2025)8039170 - 24/09/2025

CIRCULAR FOAM

D 6.3 Demonstrator for one recycling technology with downstream in 5 kg/h scale

Author: Dr. Stefanie Eiden, Covestro Deutschland AG

Date: 31 May 2025

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

Project Acronym	CIRCULAR FOAM
Project Title	Systemic expansion of regional CIRCULAR Ecosystems of End-of-Life FOAM
Project Coordinator	Dorota Pawlucka, Covestro Deutschland AG dorota.pawlucka@covestro.com
Project Duration	October 2021 – March 2025 (48 months)

Deliverable No.	D6.3
Dissemination Level[1]	PU
Lead Beneficiary	COV
Contributing Beneficiaries	Covestro Deutschland AG, Fraunhofer UMSICHT, Sulzer Chemtech AG
Issue Date of Deliverable	31.05.2025

[1]PU-public, CO-confidential, only for members of the consortium (including the Commission Services), EU-SEC-classified information: SECRET UE (Commission Decision 200/444/EC)

Disclaimer

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

Executive Summary

The recycling technology catalytic pyrolysis to recycle aniline was developed earlier in the project in g scale. The main objective of the task leading to this deliverable and part of demonstration, was to upscale the recycling process to produce new raw materials which would fulfil the specifications to produce new foams.

The successful upscaling of the depolymerisation of rigid PUR foam via smart catalytic pyrolysis and the downstream steps to separate the resulting re-aniline from the pyrolysis oil at the scale of 10 kg/h are described in this document.

The pyrolysis step was performed in a 10kg/h pyrolysis screw reactor at UMSICH. Here more than 400 kg end-of-life polyurethane pellets were successfully processed in this plant with a catalyst. Oil yields from above 50 % have been achieved. Also, more than 20 % of aniline was found in the oil. Around 300 kg pyrolysis oil could be produced successfully without any clogging issues.

The distillation process of the pyrolysis oil to r-aniline was upscaled over two steps. The first step to a 1 kg/h set up was performed at UMSICHT based on the simulation which were carried out at Covestro. Consequently, 1,9 l aniline could be produced. The second step will be done at Sulzer in a 10 kg/h set up in July 2025. Therefore, all necessary tasks were already performed which are required to insure a safe campaign.

This progress demonstrates that the upscaling of the recycling process of end-of-life appliance foam to r-aniline to 10 kg/h was successful and offers a partly closed loop recycling solution.

Table of Contents

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

1 State of the art

Catalytic pyrolysis

Pyrolysis is a process in which the thermal depolymerisation of long-chain organic materials takes place in an inert atmosphere with a catalyst (catalytic pyrolysis) or without (standard pyrolysis). Gaseous, liquid and solid products are obtained, whereby the pyrolysis of condensation polymers such as PU results in mixtures of defined compounds (fragments) due to the reaction behavior of the functional groups present. By contrast, other pyrolysis processes are focused on the production of raw materials (via pyrolysis oils) like those raw materials produced by fractional distillation of fossil fuels (refinery gases, petrol/naphtha or diesel).

However, it would be ecologically more efficient to produce directly high-value molecules (monomers) from pyrolysis. This approach has already been successfully demonstrated for polystyrene and polymethyl methacrylate, where monomers could be recovered with high yield in pilot plant size. BioBTX has also demonstrated successfully the pyrolysis of mixed PP/PE/PS to BTX (Benzene/Toluene/Xylene).

Pyrolysis of PU foam

Pyrolysis is considered a promising process for the recycling of polyurethane-based materials as other recycling approaches come with disadvantages or are not suitable for PUR. Recently, the general interest in pyrolysis of waste materials such as polyurethane has grown and there are already studies concerning the pyrolysis potential of PU-based foams. The goal is to pyrolyze the PU foam in such a way as to obtain fragments from the pMDI such as amines (aniline and toluidine) or fragments of the polyol part such as glycols, ethene or propene. The main goal for Covestro is to produce high amount of aniline because it is the direct precursor for the production of pMDA. The main focus in the CIRCULAR FOAM project is on polyether-based PUR foams used in the appliance industry. Results of the development of the catalytic pyrolysis process in g scale was described in detail in deliverable 4.2.

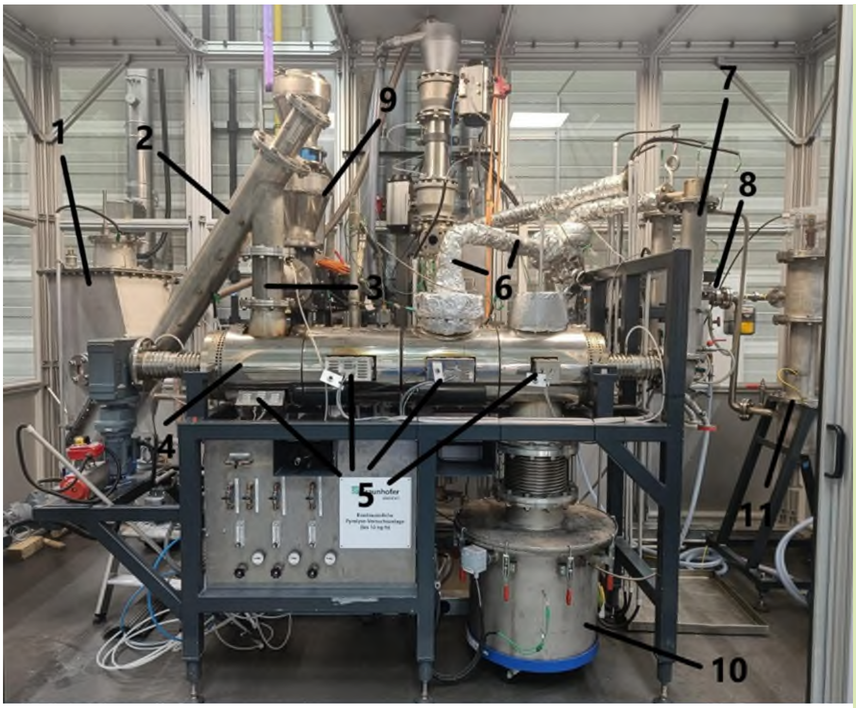
This report focuses on the demonstration of the pyrolysis process, including distillation, of the polyurethane end-of-life appliance form to r-aniline which can be used again for the production of new foams.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

2 Demonstration

2.1 Pyrolysis 10 kg/h

The 10kg/h pyrolysis plant used for the conversion of polyurethane can be seen in Figure 2. Over the course of the project the plant got adapted by adding a second feeding unit (No. 3 and 9). The first feeding unit (No. 1 and 2) was used to transport the PU into the reactor. With the help of the second feeding unit a given amount of catalyst can be added to the process.



Legend

1. Feed storage
2. Feeding screw
3. Inlet catalyst
4. Screw reactor
5. Electrical Heaters
6. Gas line
7. Condensers
8. Washing unit
9. Catalyst feed unit
10. Char container
11. ESP

FIGURE 1: 10 KG/H PYROLYSIS PLANT AT UMSICHT

The main part of the plant, the pyrolysis reactor (4), consists of a screw and is heated by external electrical heaters (5). By defining the screw rotation speed the retention time of the polyurethane and catalyst can be adjusted. After its retention time, the char and catalyst falls into a storage container (10). Vapours and gases from the pyrolysis process leave the reactor over heated pipes (6) and reach a condensing unit (7). Here, condensable parts (the pyrolysis oil) get separated from the stream and collected in bottles. Non-condensable gases (like CO, CO_2 or H_2) bypass a washing unit (8) to remove halogens (HCl, HBr) and afterwards an electrostatic separator (11). Its job is to remove remaining dust or liquid particles from the gas stream. The remaining gas stream gets incinerated by using a flare.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

By processing more than 400 kg end-of-life polyurethane pellets with a catalyst in this plant, 300 kg oil yields have been achieved. Also, more than 20 % of aniline was found in the oil. Besides, other valuable components, such as toluidine, benzene and toluene were found in reasonable quantities. So around 300 kg pyrolysis oil could be successfully produced without any clogging issues which will be used in the distillation trials in a 10kg/scale at Sulzer.

FIGURE 2: HOBOCKS WHICH CONTAIN THE 300 L PYROLYSIS OIL.

2.2 Distillation in 1 kg/h scale at Covestro/Fraunhofer UMSICHT

Experimental trials for the continuous distillation of pyrolysis oil are being conducted at Fraunhofer UMSICHT. The necessary parameters are provided by Covestro. The goal is to produce 99.5% aniline for further MDI manufacturing. Based on insights from batch experiments at RWTH and the pyrolysis oil composition, a simulation is set up in AVEVA™. A total of two trials were conducted with different settings.

This requires creating a simplified component list that serves as the foundation for the simulation. Substance component groups are formed based on retention times, and critical components are mapped. It should be noted that water is a component of the oil and must be considered, which was only incorporated in the second run. Generally, the separation sequence consists of two stages: in the first stage, low boilers are separated and in the second stage, pure aniline is purified at the top. Parameters such as reflux ratio and evaporation rate are defined based on the specifications to be maintained. The simulation results are then scaled to the distillation column at UMSICHT. The throughput is determined by the plant's boundary conditions, like diameter. This creates a dataset that can be used for the trials. The continuous distillation column with a diameter of 60 mm is shown in Figure 4.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

FIGURE 4 CONTINUOS DISTILLATION COLUMN AT FRAUNHOFE UMSICH

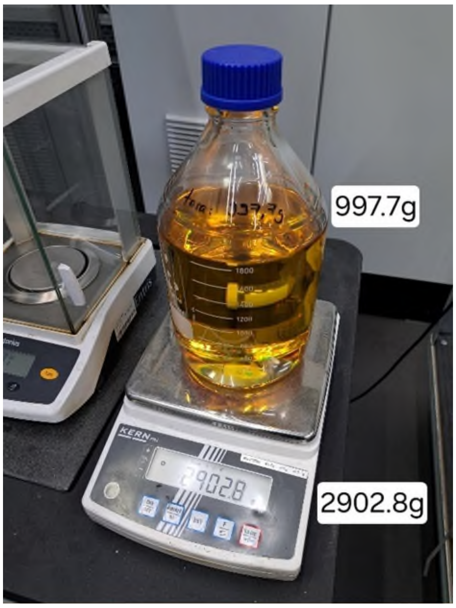


FIGURE 3: : 1,9 L SEPARATED R ANILINE.

The distillation trials confirm that the general mass balance of simulation and experiments align. Thus, 1,9 l r-aniline could be separated out of the pyrolysis oil.

2.3 Sulzer: Purification of aniline from aniline-rich pyrolysis oil through distillation process in 10 kg/h scale

Pyrolysis oil generated from Fraunhofer UMSICHT facilities will be processed in a distillation section to recover and purify aniline. The pilot-scale purification trial will be performed as a proof of concept in Sulzer test center facilities in K100 column (107 mm inner diameter sections). It will be based on the lab-scale feasibility test results carried out by Fraunhofer UMSICHT and the thermodynamic equilibrium data collected from ETH, based on simulation models. The process will be carried out in Allschwil pilot test center in a 3-stage distillation process, starting with a feed input of 4.3 kg/h in the first stage. The total quantity of pyrolysis oil which will be processed is approximately 200L. The aniline is expected to meet a purity of 99.5 %, which will be verified through analytical quality testing. Side products recovered from various stages of the process will also be analyzed to estimate the overall composition and the detailed mass balance.

The trails in the large set up will run in July. Up front several tasks had to be carried out to ensure a safe and successful campaign.

A small quantity of the pyrolysis oil generated at Fraunhofer UMSICHT has been delivered to Sulzer, including 2 l of filtered oil and 2 l of unfiltered oil. Based on the visual observation, unfiltered oil contains some solid particles which might affect the proper function of the gear pumps during the pilot-trial. Although the filtered oil appears to be free of solid particles, Sulzer team had to

This project has received funding from the European Union's Horizon 2020
research and innovation programme under grant agreement 101036854

confirm that the filtered oil can be easily processed through the pumps in the pilot-plant set-up, avoiding any potential blockages.

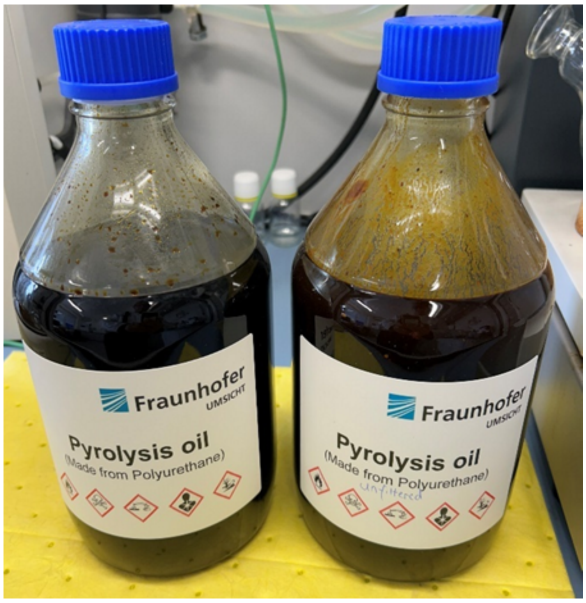


FIGURE 5: FILTERED (LEFT) & UNFILTERED (RIGHT) PYROLYSIS OIL DELIVERED FROM FRAUNHOFER UMSICHT TO SULZER

As a result, a rotary evaporation process took place in Sulzer, in Allschwil test center facilities, to investigate if the filtration stage performed upstream (at Fraunhofer UMSICHT) is sufficient to ensure removal of undesired solid particles and to avoid any issues related to mechanical failure of the pumps or blockages on the structured packing and/or distributor in the distillation column.

For the rotary evaporation process, 436 g of filtered pyrolysis oil is introduced into a round-bottom flask and subjected to reduced pressure starting at 200 mbar and gentle heating. The process conditions are adjusted to collect the necessary fractions during specific intervals. The evaporated components pass through a condenser and are collected in a receiving flask as the distillate. The distillate is collected in fractions over time which represent the expected fractions from the distillation process (based on results from the simulation model). The operation continues until approximately 58 % wt of the initial oil mass is recovered in the distillate. Throughout the evaporation, key parameters such as oil bath temperature, vacuum level, distillate and bottom temperatures are carefully controlled and documented.

The collected heavy fraction equals 177 g and allows for the evaluation of the flow behaviour. It has been observed that the collected heavy fraction is more viscous compared to pyrolysis oil, however, it remains within the pump's operational limits. Additionally, no solids, or other residues are detected that may affect the distillation process. Therefore, it can be concluded that the filtration stage performed by Fraunhofer UMSICHT is sufficient and pyrolysis oil can be processed directly in the distillation process without any pre-separation stage required. This conclusion is based on the confirmation from Fraunhofer UMSICHT that the composition and quality of the pyrolysis oil which will be delivered to Sulzer for the trial campaign will match that of the oil used in the rotary evaporation process.

In addition, in the distillate fractions, a yellow colour is observed, with an increased intensity in the final ones. Phase separation was observed in the first two fractions (D1 & D2), between water and light pyrolysis oil components which took place in less than a minute. The remaining fractions (D3 – D6) appeared as one single phase. The analysis of the samples will provide more details on the composition of the fractions.

The collected samples, including the feed, the distillate fractions and the heavy residue are shown in the picture below.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854

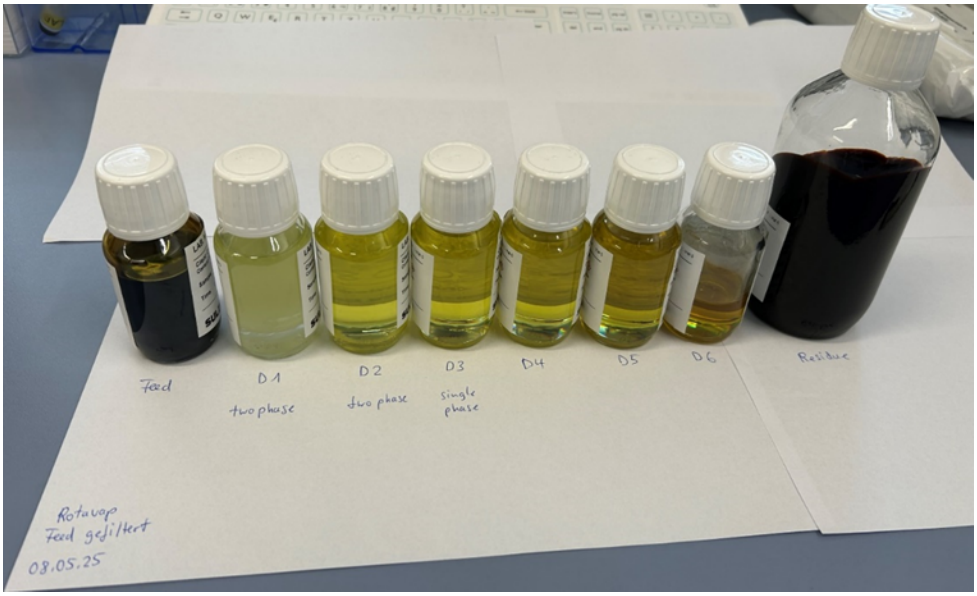


FIGURE 6 FEED, DISTILLATE & HEAVY FRACTION SAMPLES COLLECTED FROM ROTARY EVAPORATION TRIAL

Other main tasks which have been performed by Sulzer during the last months are summarized below:

- Execution of a second round of HAZOP study based on the updated pyrolysis oil composition and the updated modifications on the pilot-plant set-up. The purpose of the HAZOP is to identify and evaluate potential risks and ensure safety and operational efficiency during the trial campaign.
- Covestro and Sulzer are optimizing the simulation work which has been developed by ETH, aiming to determine the optimal operating conditions and estimate process efficiency. Sulzer also investigates through simulation work expected potential emissions in the exhaust streams.
- In parallel, analytical methods are being prepared and validated, based on the exchange with Covestro, to enable consistent and accurate characterization of samples during the trial campaign.

3 Conclusion

In this report, the successful upscaling of the depolymerisation of rigid polyurethane foam via pyrolysis and the downstream steps to separate the resulting re-aniline from the pyrolysis oil at scales of 10 kg/h L are described.

The pyrolysis step was performed in a 10kg/h pyrolysis screw reactor at UMSICHT. Here more than 400 kg end-of-life polyurethane pellets with a catalyst were processed in this plant, oil yields from above 50 % have been achieved. Also, more than 20 % of aniline was found in the oil. Around 300 kg pyrolysis oil could be produced successfully without any clogging issues.

The distillation process of the pyrolysis oil to r-aniline was upscaled over two steps. The first step to a 1 kg/h set up was performed at UMSICHT based on the simulation which were carried out at Covestro. So, 1,9 l aniline could be produced. The second step will be done at Sulzer in a 10 kg/h set up in July 2025. Therefore, all necessary tasks were already performed to insure a safe campaign.

This progress demonstrates that the up scaling of the recycling process of end-of-life appliance foam to r Aniline to 10 kg/h was successful and offers a partly closed loop recycling solution.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement 101036854