

Contaminant removal from plastic waste pyrolysis oil via depth filtration and the impact on chemical recycling: A simple solution with significant impact

Chemical recycling of plastic waste offers a promising solution to the limitations of mechanical recycling, particularly for mixed and contaminated waste streams like end-of-life plastic packaging.

Mechanical recycling often produces lower quality products unsuitable for original applications. Chemical recycling, especially for polyolefins, converts waste into virgin-grade polymers through pyrolysis and steam cracking, reducing emissions. However, it faces challenges due to contaminants in plastic waste, causing operational risks like equipment corrosion and coke formation.

To address these challenges, the study investigated depth filtration as a method to remove particles and metal contaminants from crude plastic waste pyrolysis oils. The effectiveness of three different filter media was assessed using various analytical techniques. The results outlined in this paper showed that depth filtration could significantly reduce contaminants, improving the quality of pyrolysis oils and their suitability for steam cracking. This advancement

Feedstocks

The pyrolysis oil used in this study was produced from a continuous pilot-scale pyrolysis unit, which included a single-screw extruder and a stirred-tank reactor. The pyrolysis process was conducted at 450°C, resulting in a heavy, waxy oil solid at room temperature. The starting material was a contaminated post-consumer mixed polyolefin (MPO) fraction, primarily composed of polyethylene (PE) and polypropylene (PP), with smaller amounts of other polymers like PET, PA, PS, and PVC.

Apparatus and Experimental Procedure

Filtration experiments used three depth filter media (coarse F1, medium F2, fine F3) with retention rates between 2.5 µm and 20 µm. Filters were tested at 80°C with a flow rate of 50 mL/min, processing 3000 mL of pyrolysis oil per filter type. The first 1000 mL of filtrate was discarded to minimize leaching.

Filtration Performance

The study analyzed filtrates based on filtration grade, observing increased differential pressure values due to filter media parameters like porosity and pore size, which are crucial for continuous process engineering.

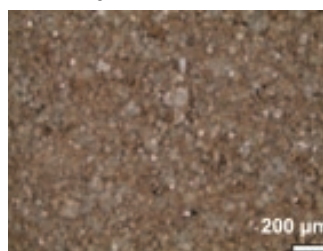
Bench-Scale Steam Cracking (BSSC) Unit

Steam cracking experiments were conducted using a bench-scale unit designed for industrial scale-up, including on-line analysis for light products using a Refinery Gas Analyzer (RGA) and GC × GC-FID. Conditions matched previous work, with two temperature profiles (820°C and 850°C). Coke deposition was measured using an infrared analyzer, with thorough cleaning to minimize uncertainty.

Results and Discussion

Microphotographs showed that the original pyrolysis oil contained brown fines (5-10 µm) and white crystal-like particles (20-50 µm).

Crude Py-oil



Coarse Filtration



Medium Filtration



Fine Filtration



Filtrate test membranes with retained contamination

The unfiltered oil had a total contamination of 69 mg/L, mostly fine particles. All three filter media effectively removed over 97% of particles, with the fine (F3) and medium (F2) filters reducing TSS to below 1 mg/L. Even the finest particles (>5 µm) were removed by over 99% with all filters.

After 2 days of storage, TSS in the filtrates increased, indicating re-agglomeration of finer particles. TSS rose from 2 mg/L to 7 mg/L for the coarse filter (F1), from <1 mg/L to 4 mg/L for the medium filter (F2), and from <1 mg/L to 2 mg/L for the fine filter (F3). This suggests that continuous filtration is advisable to prevent process issues due to larger particles.

Analysis of Retained Particles

For designing continuous filtration processes, understanding particle size, shape, and composition is crucial. SEM-EDX analysis showed unfiltered pyrolysis oil contained particles mostly smaller than 50µm, primarily composed of calcium, aluminum, chlorides, and carbon-based species. Additives are major contaminants, with calcium and silicon being dominant. Filtration removes most silicon particles, but small calcium, iron, and other contaminants may pass through. Calcium particles can cause fouling in steam cracking. After 2 days, re-agglomeration increased oxygen and silicon concentrations. Immediate processing or inert storage is needed to maintain filtrate stability.

Chemical Analysis of the filtered pyrolysis oils were analyzed using ICP-OES and GC × GC-FID to assess their suitability for light olefin production via steam cracking, focusing on contaminants and hydrocarbons. ICP-OES provided detection limits down to ppb levels, offering quantitative results.

Contaminants

Tested medium	Particle count per 1mL of sample					
	<5 µm	<10 µm	<20 µm	<50 µm	<100 µm	TSS (mg/L)
Unfiltered pyrolysis oil	2595	1420	599	91	14	69
Filtrate of coarse filter medium (F1)	7	4	1	0.8	0	2
Filtrate of F1 after 2 days storage	18	7	2	2	0.9	7
Filtrate of medium filter medium (F2)	5	3	1	0.4	0	<1
Filtrate of F2 after 2 days storage	17	6	1	15	3	4
Filtrate of fine filter medium (F3)	1	0.9	0.5	0.1	0	<1
Filtrate of F3 after 2 days storage	3	1	0.5	0	0	2

Particle count per 1 mL of sample and TSS of the crude oil and filtrates

Filtration significantly reduced particulate contamination in pyrolysis oils, with the fine filter (F3) removing 81.5% of total metals. Iron, sodium, and calcium were substantially reduced, though some organometallic species and small particles remained. Silicon was challenging to filter due to re-agglomeration. Combining different filter grades could improve efficiency. Most trace elements were reduced below detection limits, but arsenic, chromium, and titanium persisted.

Importantly filtration was proven to effectively remove metals and particles from pyrolysis oils, but heteroatoms like oxygen, nitrogen, sulfur, and chlorine can still cause issues. Hydrogen-based techniques (e.g., hydrodeoxygenation, hydrosulfurization) can remove these heteroatoms, reducing the risk of catalyst deactivation in an integrated industrial system.

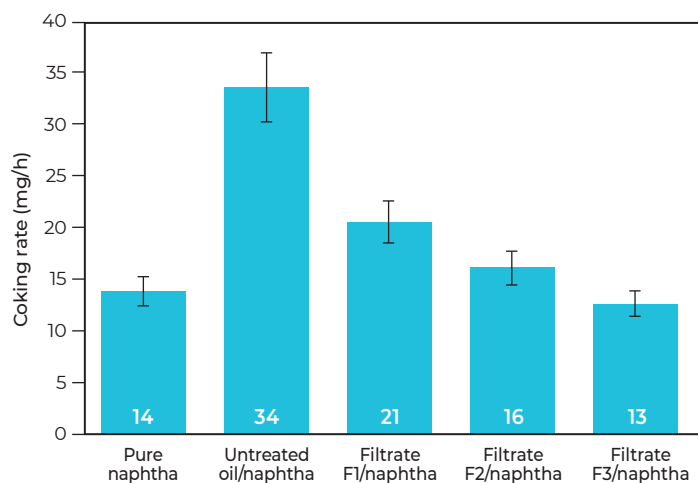
The hydrocarbon composition of the filtered pyrolysis oils remained similar to the crude oil, indicating that filtration did not significantly affect it. Therefore, steam cracking yields should be similar across samples, though technical issues like coke formation and fouling will depend on contaminant levels.

Steam Cracking Performance

A 25 wt% pyrolysis oil and 75 wt% fossil naphtha mixture was used for steam cracking. The hydrocarbon composition of the blends showed reduced olefin concentrations and slightly lower amounts of n-paraffins, iso-paraffins, naphthenes, and aromatics compared to fossil naphtha. Filtration did not significantly affect product yields, but adding pyrolysis oil enhanced ethylene yields, demonstrating its potential as a steam cracker feedstock.

Radiant Coil Coke Formation

Coke formation impacts the run-length of steam cracking furnaces. Metals like iron, nickel, and sodium catalyze coke formation. Filtration reduced metal contaminants, decreasing coke formation by 39% (F1), 52% (F2), and 62% (F3) compared to unfiltered blends. The fine filtrate/naphtha blend had a coking rate similar to or lower than pure naphtha. However, contaminants like silicon and calcium caused fouling in other sections of the cracker. Depth filtration proved effective in reducing coke formation, but further research is needed to evaluate long-term feasibility and address fouling issues.



Note: Radiant coil coke formation results in mg/h of cracking of the filtered samples/naphtha blends.

Conclusions and Outlook

This detailed study on depth filtration of contaminated post-consumer plastic waste pyrolysis oil showed that filtration effectively reduces particulate contamination while the hydrocarbon composition remained unchanged. Particle analysis revealed that calcium dominated smaller particles, while oxygen and silicon were prevalent in larger ones. Re-agglomeration of particles occurred after 2 days of storage, highlighting the need for immediate processing.

Steam cracking performance of the filtered oils was like unfiltered oil in terms of product yields, but coke formation was significantly reduced by 40-60%. This indicates that depth filtration can enhance the suitability of pyrolysis oils for closed-loop recycling.

For larger-scale applications, a continuous multi-stage filtration process is recommended to effectively remove particles without high pressure drops. Immediate use of filtered products is advised to prevent re-agglomeration. The study also identified calcium, silicon, iron, and oxygen as major contaminants, suggesting that plastic additives are a key source of metal contamination. This insight can guide manufacturers in designing for chemical recycling.

Contact Information:

For further information, you can contact Marvin Kusenbergs at [marvin.kusenbergs@ugent.be] or Emmanuelle Biadi at [emmanuelle_biadi@europe.pall.com]

Link to original paper:

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Corporate Headquarters

Port Washington, NY, USA
+1-800-717-7255 toll free (USA)
+1-516-484-5400 phone

European Headquarters

Fribourg, Switzerland
+41 (0)26 350 53 00 phone

Asia-Pacific Headquarters

Singapore
+65 6389 6500 phone


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