Membrane-Based Treatment of Hydrothermal Carbonization Effluent from Municipal Biowaste

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Abstract The treatment of hydrothermal carbonization (HTC) liquor derived from municipal solid waste (MSW) remains a key challenge due to its high organic load. This work compares, for the first time, the direct application of polymeric microfiltration (MF) and ultrafiltration (UF) with a multi-stage cascade configuration (MF → UF → nanofiltration, NF) for MSW depuration. Single-stage UF proved effective for turbidity removal (>97%) but achieved limited reductions in organic content. By contrast, the cascade system significantly enhanced performance, with COD, TOC, and TN removals reaching 89%, and >93% when optimized.

Keywords: HTC, Anaerobic digestion, Digestate, Ultrafiltration, Nanofiltration

1. Introduction

Addressing environmental sustainability has become a priority, fostering the adoption of circular economy frameworks that repurpose waste into valuable resources. Anaerobic digestion (AD) plays a central role by converting organic waste into biogas, a renewable energy source, but produces digestate as a byproduct. Managing digestate requires substantial land and poses environmental challenges such as eutrophication and water loss through evaporation (1).

Hydrothermal carbonization (HTC) offers a complementary solution by converting wet biodegradable residues, such as food waste and sewage sludge, into hydrochars for pollutant adsorption, soil enhancement, and renewable fuels (2). HTC also generates a liquid byproduct (HTC liquor) rich in organics and nutrients, but requires effective treatment due to its high total organic carbon (TOC) and chemical oxygen demand (COD) (3). Untreated, this liquor poses pollution risks; however, its nutrient-rich composition suggests potential for resource recovery. Strategies like recirculation into AD systems have shown promise in enhancing biogas yields (4), while direct treatments such as activated carbon-catalyzed HTC and advanced oxidation processes like Fenton reaction have achieved COD reductions of up to 90% and 73%, respectively (5,6).

Membrane separation technologies, including ultrafiltration (UF) and nanofiltration (NF), present a scalable, modular approach to HTC liquor treatment. Previous studies on HTC liquor from agricultural and sewage sludge have demonstrated significant COD reductions using membrane cascades (3,7). However, research on HTC liquor derived from municipal solid waste (MSW) remains limited. This study evaluates membrane-based treatments for HTC liquor from MSW, focusing on optimizing pollutant removal. As water scarcity intensifies, such solutions align with Sustainable Development Goal 6 (clean water and sanitation), supporting resource efficiency and sustainable water management in biogas systems.

2. Materials and methods

The HTC liquor, derived from biowaste mixed with plastic bags, was pre-filtered (12–15 µm, Whatman™) to prevent membrane clogging. A high-pressure stirred cell (HP4750, Sterlitech®) operated in dead-end mode was used with N_2 gas to apply transmembrane pressure (TMP, bar). Batch volumes of 50 mL were processed through membrane area (A_M) ranging 4.1–14.6 cm^2 and permeates were reused in multi-step trials.

MF (0.2–0.45 µm), UF (10 kDa), and NF (200 Da) membranes made of different polymers (RC – regenerated cellulose, CA – cellulose acetate, PES – polyethersulphone, PA – polyamide) were selected and operated at pressures of 2.0 (MF), 4.8 (UF), and 8.0 bar (NF). Permeability (L_P, mL·min^{-1}·cm^{-2}·bar^{-1}) and rejection efficiency (R_i, %) of key parameters (i = TOC, COD, TN) were evaluated over time (t, min) according to **Eqs. (1)** and **(2)**, respectively. The best performing membranes were selected for cascade trials based on flux and rejection efficiency.

$$L_P = \frac{V}{A_M \cdot t \cdot TMP} \quad (1) \qquad R_i = \frac{C_{i,Feed} - C_{i,Permeate}}{C_{i,Feed}} \times 100 \quad (2)$$

3. Results and discussion

The pre-filtered HTC liquor exhibited very high organic loads (COD 47.5 g·L^{-1}, TOC 19.2 g·L^{-1}), substantial total nitrogen (TN 817 mg·L^{-1}, 25% as NH_4^+) and high phenolic content (476 mg·L^{-1}). Turbidity (70 NTU) and total solids (49.4 g·L^{-1}) pointed to the necessity of pre-filtration.

Single-stage MF and UF achieved >97% turbidity removal but only ~13–32% COD/TOC/TN rejections (**Fig. 1**), underscoring their inadequacy for complex HTC liquor treatment. By contrast, all three cascades markedly outperformed single-stage runs: global COD rejections rose to 88–93%, TOC to 84–93%, and TN to 82–93%.

Cascade #1 (MF → UF_{10kDa}_RC → NF_{200Da}_PA) and #2 (MF → UF_{10kDa}_PES → NF_{200Da}_PA) (**Fig. 1**) delivered nearly identical overall rejections (~89% COD), indicating that once MF membranes remove colloids and macromolecules, the choice of UF material has minimal impact on final performance. Cascade #3 (MF → UF_{10kDa}_PES → UF_{10kDa}_RC → NF_{200Da}_PA) achieved the highest rejections (≥93% across COD, TOC, TN), validating the benefit of sequential sieving – even with identical MWCOs – to sharpen effective cut-offs and mitigate fouling (e.g., L_P = 7x10^{-2} mL·min^{-1}·cm^{-2}·bar^{-1} for the second UF step, one order of magnitude higher than for UF in cascade #2).

Despite these gains, final COD (~3.2 g·L^{-1}) and the persistent yellow coloration indicate the presence of low-molecular-weight organics (namely phenols) beyond NF retention. Such levels exceed typical industrial discharge limits and demand complementary polishing steps.

4. Conclusions

Single-stage MF and UF showed limited removal, whereas multi-stage cascades – especially double UF followed by NF (cascade #3) – achieved the highest contaminant rejection by sequentially sieving and minimizing fouling. However, the permeate remained yellow from soluble phenolics and contained residual COD above discharge limits, underscoring the need for post-treatment (e.g., adsorption or advanced oxidation) and further optimization to address low-molecular-weight organics and fouling.

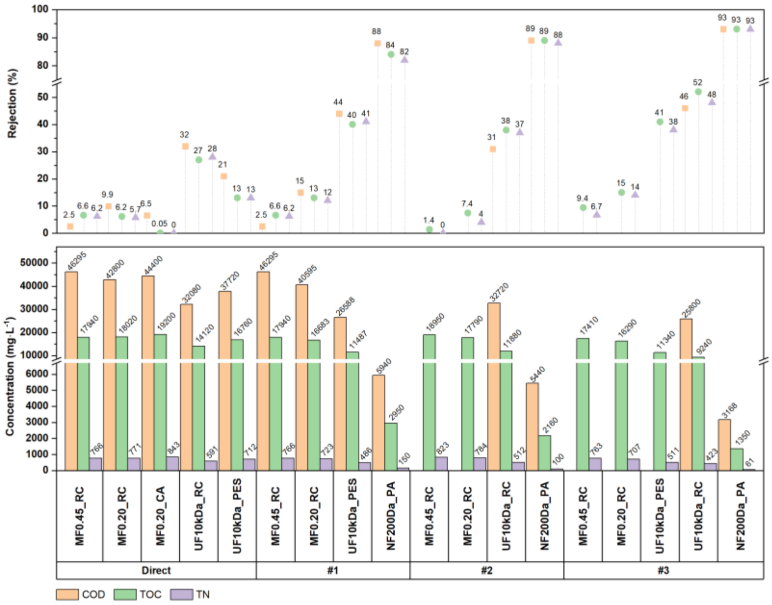

Figure 1. COD, TOC, and TN concentrations in the permeate after each step of cascades #1–3 and their overall rejections from the initial HTC liquor. Single-stage ("Direct") filtration results included for comparison.

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