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THE INVESTIGATION OF CATALYTIC DECOMPOSITION OF WASTE FISHING NETS FOR ENERGY PRODUCTS RECOVERY

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ABSTRACT: The ongoing issue of marine plastic waste causing eutrophication and water pollution underscores the need for eco-friendly solutions. Collected waste materials can potentially serve as an alternative feedstock, generating higher-value energy products. Additionally, the recovery of caprolactam from nylon 6 waste fishing nets has the potential to conserve resources, enhance waste economics, and establish a closed loop in the fishing net industry's circular economy.

This study focuses on investigating the pyrolysis process and the impact of catalyst interactions at varying temperatures on resulting products, contributing to circular economy efforts. We examine used fishing nets through TGA-DTG-FTIR analysis and a mini pyrolysis plant. Y-type zeolite and ZSM-5 were used as catalysts, with ratios of 1:3 and 1:8, respectively. Initial micro-thermal analysis characterized the feedstock, revealing minor decomposition at around 200 °C and a significant peak at 450 °C, resulting in an 88 wt.% weight loss. Analysis of pyro-oils revealed fractions rich in aromatic and aliphatic hydrocarbons like naphthalene, styrene, and toluene.

Furthermore, caprolactam recovery yields not only energy products but also higher-value chemical derivatives. The combined influence of temperature and catalysts indicated that the optimal condition for generating the purest, highest-quality products is 700 °C. Based on the findings, catalytic thermal treatment at 700 °C with Y-Type catalyst holds promise as an effective method for caprolactam extraction with a substantial yield (96%).

Keywords: Pyrolysis, Fishing nets, Waste-to-energy, Caprolactam, Catalytical-conversion.

1. INTRODUCTION

In an era of growing environmental concern and diminishing fossil fuel reserves, innovative solutions are imperative to tackle both waste management and energy generation challenges. Coal, natural gas, and petroleum constitute the central fossil fuel reservoirs that fulfill the global energy needs. These resources play a pivotal role in satisfying a diverse array of energy demands, collectively accounting for 85% of the world's energy consumption. Furthermore, the combustion mechanism inherent in fossil fuel utilization prominently escalates the discharge of carbon dioxide, nitrogen oxides, and sulfides. This phenomenon engenders substantial ecological perturbations and imparts a profound jeopardy to human health and the overall biosphere.

Plastic waste found along coastlines is recognized as a profound contributor to oceanic and freshwater contamination, constituting a pivotal factor in the disruption of marine ecosystems and the vitality of aquatic organisms (Ciriminna & Pagliaro, 2020). Recent investigations reveal that the cumulative plastic load in aquatic environments encompasses a staggering 5.2 trillion fragments, corresponding to an aggregate mass surpassing 260,000 tons (Bandini et al., 2021). These fragments emanate from diverse sources, often intertwined with improper daily practices. Notably, fishing-related operations emerge as a substantial generator of plastic waste and pollution, with an estimated annual disposal of approximately 700,000 tons of fishing equipment into aquatic habitats. Alarmingly, this discarded gear annually entangles and endangers an average of 130,000 whales, seals, and sea lions, underscoring the substantial ecological consequences of such activities.

Energy reclamation from plastic waste offers an alternative avenue, characterized by its straightforward approach that demands minimal exertion and treatment, coupled with modest expenses in contrast to the retrieval costs (Yousef et al., 2021). This technique engenders a trio of outputs—namely, pyro-oil, gas, and char—subject to the prevailing operational conditions (Brebu & Vasile, 2010). As a result of these premises, a series of inquiries have been undertaken to scrutinize the pyrolytic behavior of discarded fishing nets, employing micro-scale thermogravimetric analysis to facilitate polymer reclamation (Jang et al., 2021). Evidently, these investigations demonstrate that fishing nets possess the potential to degrade into supplementary energy derivatives alongside beta-caprolactam monomers, a pivotal compound extensively utilized in the synthesis of nylon-6 polymeric fibers (Skvorčinskienė et al., 2019). The reclamation of these monomeric constituents stands to contribute towards resource conservation, waste reduction, and alleviation of environmental encumbrances, thus fostering the exploration of alternative, more cost-effective avenues for their production (Eimontas et al., 2021).

Chaidan et al. (Chaihad et al., 2020) extensively detailed the catalytic thermal conversion of fishing nets into valuable and premium-grade energy commodities. In their work, the researchers explored the thermal treatment of used fishing nets in conjunction with calcined scallop shells to facilitate the recovery of ε-caprolactam monomer. The outcomes revealed that the catalyst significantly augmented the output of targeted products, specifically ε-caprolactam, yielding up to 66 wt.% at a temperature of 410 °C and a residence time of 2 minutes. While sourcing an alternative catalyst for enhancing the thermal decomposition of fishing nets and achieving superior product quality remains a challenge, the assessment underscores the pertinence of fishing nets pyrolysis. It confirms the suitability of fishing nets as a feedstock within pyrolytic technologies, warranting further comprehensive investigations for subsequent elucidation.

Within this investigation, fishing nets were employed as the primary substrate for conducting pyrolysis trials, during which the chemical composition of resulting volatile substances was meticulously analyzed. The modus operandi behind weight reduction and the corresponding temperature spectrum were subjected to scrutiny via thermogravimetric analysis. Additionally, the composition of the generated products underwent evaluation through the utilization of a combined thermogravimetry and Fourier-transform infrared spectroscopy (FTIR) system. Furthermore, the examination encompassed the study of the pyrolytic process utilizing distinct ZSM-5 and Y-Type catalysts.

2. METHODOLOGY

2.1 Material

Recovered fishing nets were procured from Melnragė beach in Klaipėda, Lithuania, to serve as the primary material for all experimental procedures. Initial preparatory steps involved subjecting the feedstock intended for thermal treatment to a washing process to eliminate predominant sand and abrasive debris. Subsequent to this cleansing, the fishing nets underwent a drying regimen in accordance with ISO 579 standards, followed by a process of segmentation to achieve a uniform fraction. procedures to achieve a state of immediate usability. Additionally, the ZSM-5 catalyst exhibited the subsequent specifications: SiO2/Al2O3 molar ratio (38), physical configuration (column pelletized), pore volume (≥0.25 mL/g), dimensions (Φ2 × 2–10 mm), bulk density (0.72 kg/l), specific surface area (≥250 m2/g), compressive strength (≥98 N/cm2), attrition rate (<1 wt%), pore size (~5 Å), binder composition (Pseudo-Boehmite, 30 wt.%). On the other hand, the Y-Type-RE2 catalyst possessed the subsequent attributes: SiO2/Al2O3 molar ratio (>5), Unit Cell size (<2.453 nm), specific surface area (≥578 m2/g), Particle Size Distribution D50 (<6 μm), and bulk density (0.68 kg/l).

2.2 Thermal decomposition at micro-scale using TGA-FTIR system

The investigation of thermal degradation of the designated feedstock was executed employing a thermogravimetric analyzer (Netzsch Jupiter F3, Germany) coupled with a Fourier transform infrared spectrometer (Bruker Tensor 27, Germany) system, denoted as TG-FTIR. All experimental trials were conducted within a nitrogen-rich anoxic environment, maintaining each batch at 15-20 mg and a nitrogen flow rate of 60 ml/min. Consistent with the findings from prior literature, catalytic pyrolysis experiments were conducted employing Y-type Zeolite and ZSM-5 catalysts, using catalyst-to-feedstock ratios of 1:3 and 1:8, respectively, while adhering to identical conditions (Eimontas et al., 2021). The experimental parameters spanned a temperature range of 40-850 °C. Differentiation of thermogravimetric analysis (DTG) curves were performed through the derivation of thermogravimetric analysis (TGA) curves, facilitating the identification of the peak thermal decomposition zone for each analyzed sample.

2.3 Pyrolysis test at laboratory scale bench

The experimental assessments involving the pyrolysis of fishing nets samples were conducted employing a laboratory-scale pyrolysis apparatus. In response to discrepancies noted among diverse authors' results within the introductory context, a decision was made to investigate product distribution up to maximum temperatures of 500, 700, and 900 °C, employing a 20-minute isothermal hold at the peak temperature. The heating rate was set at approximately 20 °C/min, aligning with the specifications of a previous publication (Eimontas et al., 2021). A comprehensive illustration of the laboratory-scale bench setup is presented in Figure 1. Each pyrolysis trial utilized a sample mass of 100 g. A nitrogen atmosphere, with a flow rate of 3 L/min, was established for gas creation. The pyrolysis apparatus encompassed a principal reactor chamber, connected to a condenser at its terminus, characterized by small apertures (up to 2-3 mm) for the egress of formulated gaseous and liquid products, while retaining the char fraction within the reactor.

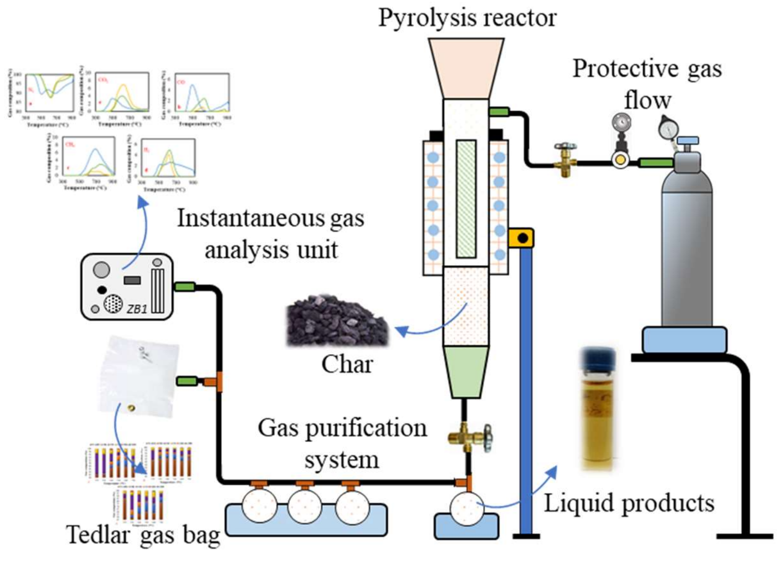

Figure 1. Laboratory scale pyrolysis bench

Liquid products were collected in a reservoir positioned at the reactor's terminus, while gaseous products were routed through a conduit network, initiating a sequence involving cooling, purification, analysis, and collection. The process of purifying the formulated gaseous products was executed concurrently via passage through five isopropanol-filled scrubbing vessels. Continuous analysis of the composition of purified gases was performed utilizing a VISIT 03H gas analyzer.

For the assessment of certain lighter C_nH_m species present, the accuracy of CH4 concentration values could potentially be compromised due to the inherent limitations of the infrared (IR) method. To address this, periodic measurements using gas chromatography (GC) were intermittently integrated. Subsequently, the liquid and gaseous products obtained were subjected to analysis via GC/MS (Agilent 7890A) system, with identification of predominant compounds facilitated through the NIST database. The carrier gas flow rate within the GC/MS system was maintained at 1.5 ml/min, and the principal analytical range spanned from 30 to 600 m/z. The utilized column for this system is the HP-5MS, characterized by a (5%-phenyl)-methylpolysiloxane stationary phase.

3. RESULTS AND DISCUSSION

3.1 Micro-thermal analysis by TGA

Figure 2 depicts the outcomes of TGA-DTG analyses conducted during the pyrolysis of the designated feedstock. Notably, the fishing nets exhibit a minor decomposition region near 200 °C, attributable to the volatilization of impurities and chemical remnants from seawater. This process could potentially lead to the degradation of auxiliary polymeric chains alongside the primary nylon 6 polymer, including substances like PP and PE. Furthermore, a substantial degradation phase transpires around 440 °C, the mechanism of which can be attributed to the linear condensation and thermal penetration of the polymeric chains within nylon 6 (marked by recurring amide bonds, -NH-CO-). Consequently, a substantial proportion of extended polymeric chains undergo decomposition, resulting in the generation of pyrolysis byproducts encompassing volatile liquids (paraffins, aromatic hydrocarbons, etc.), gaseous constituents (H_2, CH_4, monomers), and solid char particulates. Extant literature supports a variance in the upper degradation temperature threshold for these polymers, ranging between 400 °C and 460 °C, contingent upon the rate of heating (Kim et al., 2005).

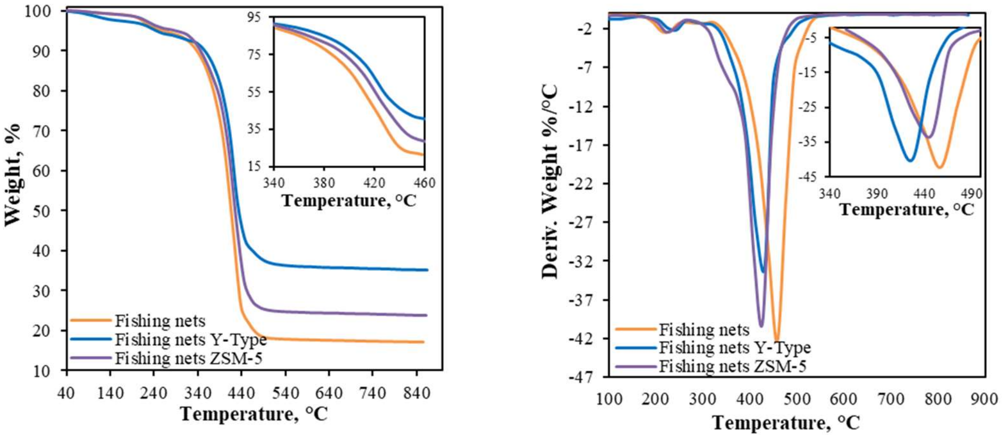

Figure 2. TGA (A) and DTG (B) curves of fishing nets decomposition

Furthermore, the catalytic micro-thermal analysis distinctly reveals that the introduction of catalyst engenders a notable elevation in the thermal resistance of sample decomposition, as evinced by the total weight loss pattern. This phenomenon stems from the inherent resistance of ZSM-5 and Y-type catalysts to decomposition, thus resulting in their residual presence throughout the pyrolysis process.

Consequently, recalibration of the residual mass within the TGA data becomes imperative to ensure the precision of result evaluation (Bridgwater, 2012). Figure 4 A offers recalibrated results predicated on the catalyst content. Examination of these recalibrated results establishes that the incorporation of catalyst does not exert any discernible influence upon weight loss or, by extension, the overall efficacy of thermal decomposition within the principal degradation zones. The DTG profiles (Figure 4 B) reveal a distinct, well-defined peak within the decomposition range of fishing nets (approximately 440 °C), thereby confirming the limited modification in fundamental characteristics, but rather accentuating the nuances of resultant chemical products.

3.2 TGA-FTIR data analysis

To assess the profile of volatile constituents during micro-thermal analysis, a coupled TGA-FTIR system was employed. Figure 5 presents two- and three-dimensional FTIR evolved gas spectra captured at the apex of maximum decomposition during pyrolysis, comparing fishing nets samples with and without the catalyst. In the absence of a catalyst, the fishing nets spectrum reveals a minor peak around 2300 cm^{-1}, attributed to the presence of CO_2 (Wang et al., 2014), a feature mitigated with catalyst incorporation. Moreover, discernible oscillations at approximately 1300 and 1500 cm^{-1} signify the existence of the N-O functional group (Wang et al., 2014), while valence carbonyl group oscillations at 1700 cm^{-1} point to the presence of aldehydes and ketones. A minor overtone oscillation at 3600 cm^{-1} signifies the amine (N-H) group originating from nylon-6 thermal decomposition.

The analysis of Fig. 3 reveals a distinct and intense peak around 3000 cm^{-1} within the fishing nets spectrum, attributed to C-H bonds characteristic of methane and aromatic-like compounds (Shen et al., 2010). The absorbance of hydrocarbons (C-H) and aromatic compounds undergoes a slight enhancement, attributed to the aggregation of non-stable hydrocarbons leading to polyolefin formation. This process, in turn, can yield a higher abundance of flammable compounds and oils (Bu et al., 2021). This conjecture is supported by the discernible presence of peaks within the fingerprint region spanning 600 – 1000 cm^{-1}, primarily originating from aromatic compounds. Furthermore, prior studies have demonstrated a direct correlation between the quantity of flammable derivatives and the heating rates (Pannase et al., 2020). Notably, the catalyst's inclusion demonstrated minimal influence on the indicated FTIR spectra peaks, as substantiated by previous studies.

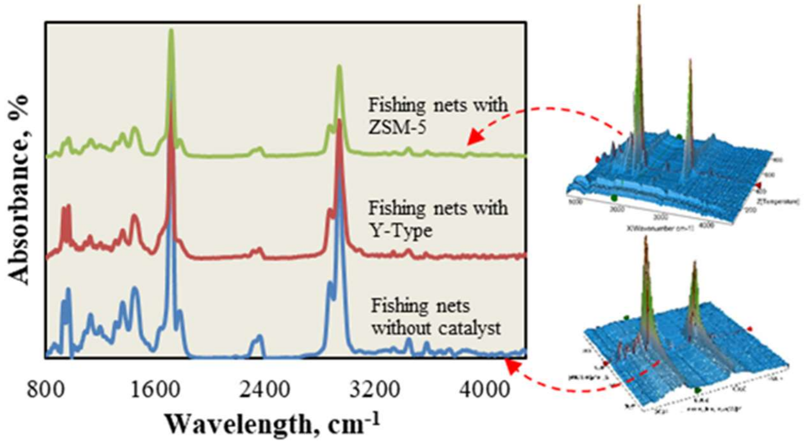

Figure 3. FTIR spectra curve of the fishing nets with and without catalyst

3.3 Gaseous products analysis

To attain insights into the thermal decomposition of the feedstock during the pyrolysis procedure and the gradual release of gaseous byproducts as the process temperature incrementally escalates, the semionline gas analyzer VISIT 03H was employed. Typically, following the thermal treatment of plastic waste derived from fishing activities, the resultant gaseous products encompass a composition inclusive of carbon monoxide (CO) and dioxide (CO_2), hydrogen (H_2), methane (CH_4), and additional light hydrocarbons like C_2H_2, C_2H_4, C_2H_6, and others. The temporal progression of gas evolution within the context of seaweed pyrolysis is visually elucidated in Figure 4.

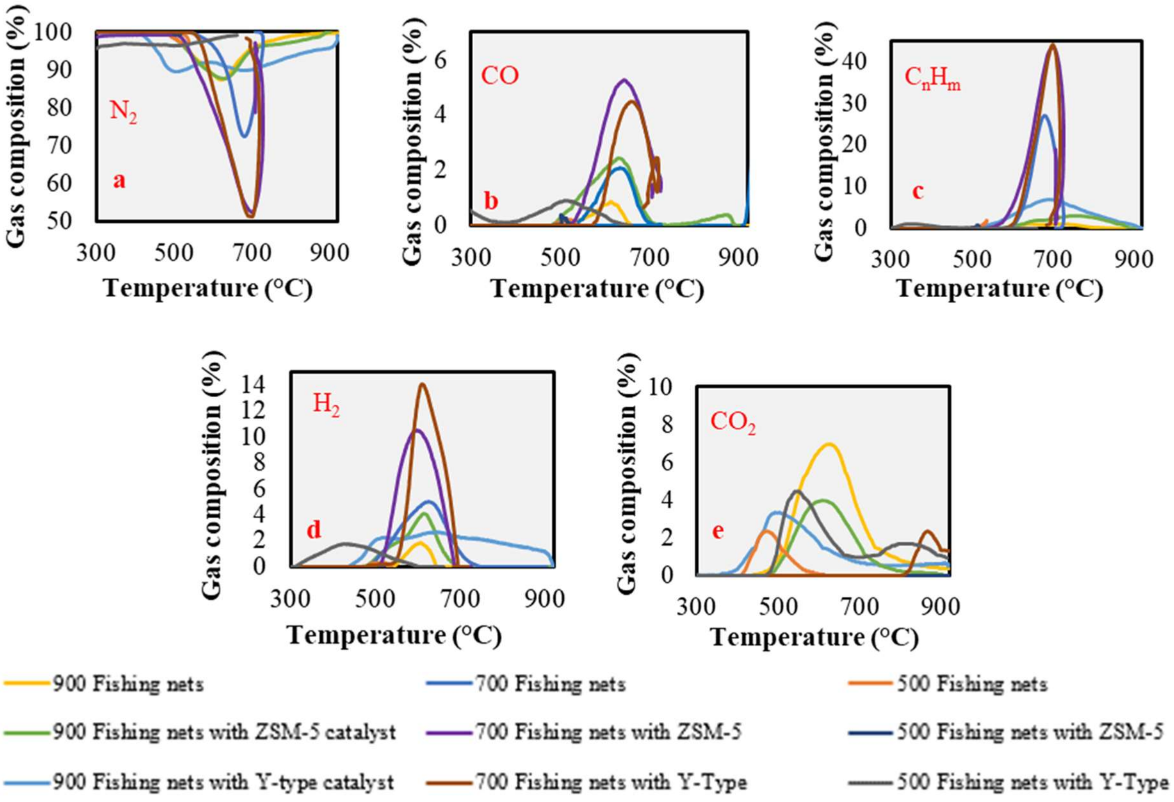

Figure 4. Gaseous products analysis by the Online measurement device

Primarily, a comprehensive elucidation of the impact of temperature on the formation of pyrolysis products becomes a fundamental pursuit. The entirety of experimental trials transpired within a temperature range spanning 500 to 900 °C, a regime that conspicuously unveils distinct and discernible trends. As evidenced in Figure 4-a, the nitrogen content within the gas composition undergoes a marked reduction as the thermal treatment culminates at each of the targeted temperatures (500, 700, and 900 °C), signifying the commencement of product formation. Notably, temperature elevation exhibits a pronounced influence on the generation of gaseous products. Specifically, an increment in temperature propels the escalation of certain constituents: the proportion of CO escalates from 0.3 to 4.9%, C_nH_m content surges from 1.4 to 27.4%, and H_2 increases from 0.3 to 4.6%. Simultaneously, the concentration of CO_2 experiences a decline from 2.3 to 0.1% upon transitioning from 500 to 700 °C. A subsequent augmentation (from 700 to 900 °C) elicits a transformative impact on the composition of all components. Concretely, CO content diminishes from 4.9 to 0.8%, C_nH_m content diminishes from 27.4 to 2.4%, H_2 content diminishes from 4.6 to 1.9%, while CO_2 content ascends from 0.1 to 6.7%.

The investigation established the optimal temperature range for the generation of valuable gaseous products to be approximately 700 °C. However, these findings notably emphasize the substantial role of temperature in shaping the composition of the resulting pyrolysis gaseous products, particularly concerning the combustible gases (C_nH_m and H_2). These trends underscore that the elevated temperature regime contributes to an augmented quantity of these derivatives, indicative of the decomposition of residual organic constituents within the remnant fraction (Zhang et al., 2020).

The catalytic impact on the pyrolysis process holds substantial significance, as exemplified by the data presented in Figure 6. The illustrated information vividly illustrates the pronounced enhancement of flammable gaseous product production facilitated by both catalysts, namely ZSM-5 and Y-Type. The catalytic intervention distinctly prompts the release of C_nH_m derivatives, precipitating a notable rise in their content from 27.4 to 43.5% with ZSM-5 catalyst and 43.4% with Y-Type catalyst. Similarly, the presence of H_2 also undergoes a considerable elevation, surging from 4.6 to 10.2% with ZSM-5 catalyst and further to 14.1% with Y-Type catalyst. The catalytic influence engenders a decrement in CO_2 yield, a consequence attributed to the propensity of CO_2 to interact with H_2 under pyrolysis temperatures of 700 and 900 °C, giving rise to the formation of C_nH_m molecules. This, in turn, contributes to a pronounced escalation in the production of these derivatives. Notably, the outcomes align harmoniously with the findings gleaned from a review of pertinent literature, wherein analogous tendencies were observed in the decomposition of polyamide plastic wastes through catalyzed pyrolysis processes (Kim et al., 2005).

3.4 Liquid products analysis

Figure 5 portrays the GC/MS spectra encompassing the tars ensuing from the pyrolysis of fishing nets, both in the presence and absence of a catalyst, conducted at varied temperatures. The spectra unveil a conspicuous proliferation of high-molecular-mass tars, a phenomenon particularly pronounced in the absence of catalysts. Upon examination, it was ascertained that under the conditions of 900 °C pyrolysis, the composition of the heavy liquid fraction chiefly comprises polysubstituted benzene derivatives, exemplified by compounds like toluene, ethylbenzene, o-xylene, naphthalene, as well as variously substituted phenolic compounds.

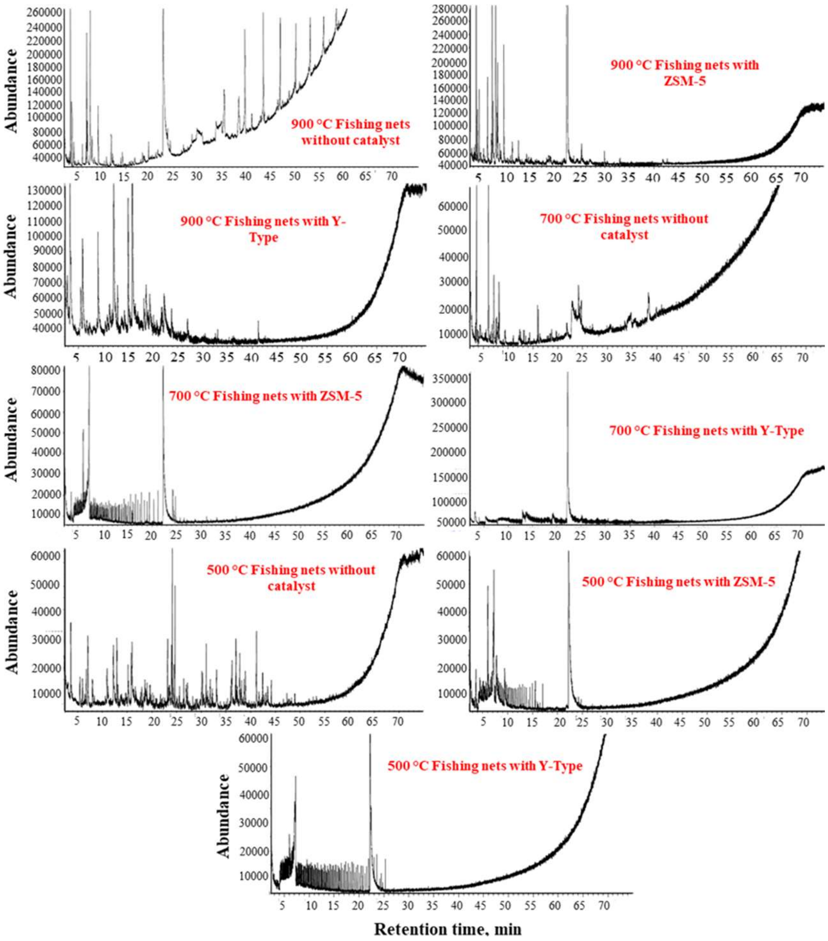

Figure 5. GC/Ms spectra curves of fishing nets liquid pyrolysis products

The introduction of a catalyst manifests a distinct effect, diminishing the presence of higher molecular mass derivatives while transforming them into lighter hydrocarbons. Moreover, the quantity of caprolactam exhibits a reduction due to the influence of decomposition reactions and the formation of lighter compounds. Remarkable trends emerge with the synergistic interaction of the Y-Type catalyst and elevated temperatures. In this context, a substantial portion of caprolactam experiences disintegration into the lighter molecular mass compound, 5-hexaneamide, via the aforementioned mechanism (Draye et al., 2001).

Noteworthy observations arise concerning caprolactam recovery as the temperature is modulated. The findings indicate that over 80% of this compound can be recuperated at 500 (83%) and 700 °C (96%) in conjunction with the Y-Type catalyst. Analogous trends, as observed in light tars analysis, hold sway: the catalyst curtails the abundance of heavier molecular mass compounds, with additional catalyst content accentuating this effect in conjunction with decreasing temperatures.

3.5 Pyrolysis conversion efficiency

To assess product yields and the influence of catalysts on product formation, the calculation of conversion efficiencies was conducted. A comprehensive mass balance was established to account for pyrolysis products, encompassing the initial feedstock mass and the terminal mass of resulting pyrolysis oil, pyro-gas, and biochar. Furthermore, the proportion of selected feedstock converted into liquid and gaseous products was quantified, with the outcomes documented in Table 1.

Table 1. Product yields

Sample	Pyrolysis oil, wt.%	Pyrolysis gas, wt.%	Pyrolysis char, wt.%
Fishing nets 500 °C	73.11	26.58	0.31
Fishing nets 700 °C	71.21	28.38	0.41
Fishing nets 900 °C	67.63	32.02	0.35
Fishing nets 500 °C Y-Type	76.41	23.17	0.42
Fishing nets 700 °C Y-Type	73.22	26.30	0.48
Fishing nets 900 °C Y-Type	69.63	29.98	0.39
Fishing nets 500 °C ZSM-5	77.22	22.23	0.55
Fishing nets 700 °C ZSM-5	74.68	24.69	0.63
Fishing nets 900 °C ZSM-5	70.15	29.24	0.61

The discernible trends characterizing the generated products align with established patterns intrinsic to thermal decomposition processes. A decrease in temperature induces heightened formation of liquid products, concomitant with a reduction in the emission of gaseous products. The apex of liquid product generation was observed at 500 °C with ZSM-5 catalyst, whereas the maximum of gaseous product yield was attained at 900 °C in the absence of a catalyst. This divergence can be ascribed to the distinctive catalytic mechanisms operating during the pyrolysis process. Specifically, the Y-Type catalyst curbs the development of aldehydes, acids, and ethers, concurrently promoting the production of light aromatic derivatives. In contrast, the ZSM-5 catalyst augments the production of aromatic derivatives while exerting an impact on the abundance of oxygenates through favoring deoxygenation reactions (Cai et al., 2016).

4. CONCLUSIONS

In this study, we conducted an investigation focused on the catalytic pyrolysis of discarded fishing nets, employing Y-Type and ZSM-5 catalysts across a range of temperatures (900-700-500 °C). Initial evaluation of the feedstock was undertaken using TGA-DTG-FTIR analysis, which unveiled a prominent peak at 440 °C accompanied by an 88 wt.% mass loss—attributed to the thermal degradation of the primary polymer, Nylon-6. Gaseous product assessment unveiled significant functional groups (C-H, C=O, O-H, and N-H), indicative of aliphatic and aromatic hydrocarbons, aldehydes, ketones, alcohols, and amines.

At the laboratory scale, our experiments yielded gaseous, liquid, and solid fractions. Variations of temperatures highlighted an optimal yield of gaseous products at 700 °C, containing 27.4% C_nH_m and 4.6% H_2. The introduction of catalysts notably enhanced these compounds, with the Y-Type catalyst at 700 °C yielding up to 43.4% C_nH_m and 14.1% H_2. The promising recovery of caprolactam was particularly notable, especially apparent with the Y-Type catalyst at 500 °C (83%) and 700 °C (96%), signifying the potential for monomer and energy product reclamation.

Furthermore, our mass balance computations aligned with observable trends: decreasing temperatures led to a heightened formation of liquid products, peaking at 73.1%, while the gaseous product content decreased to 26.6%. Introduction of the catalyst slightly increased the liquid product content, reaching a zenith of 77.2%, inducing significant alterations in derivative composition.

This inquiry underscores the feasibility of repurposing waste fishing nets via pyrolytic processes to generate supplementary energy products. The substantial recovery of caprolactam from these nets not only presents a path to extract higher-value products but also offers avenues for its utilization in Nylon-6 synthesis. Ultimately, this study underscores the material's potential as a feedstock for synthesizing high-value energy products, thereby augmenting contributions to the circular economy.

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