



Recent Advances in the use of Catalysts to Improve the Efficiency of Oil Sludge Pyrolysis

N. U. Nurgaliyev,^{1,2} Ye. K. Aibuldinov,^{1,*} E. E. Kopishev,³ Zh. B. Iskakova,¹ A. Kolpek,³ A. S. Sabitov,^{3,*} G. Zh. Alzhanova,¹ G. G. Abdiyussupov¹ and M. T. Omirzak¹

Abstract

Oil sludge (OS) remains challenging to manage due to toxic metals and recalcitrant hydrocarbons. Among available treatments, pyrolysis enables energy recovery while limiting formation of dioxins and particulates; however, redistribution of metals and heteroatoms during conversion necessitates stringent emissions control and product upgrading. This review synthesizes recent advances in catalytic co-pyrolysis of OS using metal oxides (including nanostructured), mesoporous zeolites, and molecular sieves. This review evaluate how catalyst composition, acidity, and textural properties shape product yields and composition; suppress sulfur- and nitrogen-bearing species (*e.g.*, H₂S); enhance char adsorption capacity; and influence kinetics (activation energy, rates), conversion, liquid-phase viscosity, and overall process efficiency. The analysis identifies catalyst systems and operating windows that improve oil quality and reduce environmental risks, and outlines remaining barriers and priorities for scale-up.

Keywords: Oil sludge; Catalytic pyrolysis; Zeolites; Molecular sieves; Metal oxides; Co-pyrolysis; Pyrolysis oil quality; Hazardous waste valorization; Heavy metal immobilization; Energy recovery.

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1. Introduction

In the contemporary era, social, scientific and technological progress has given rise to environmental problems relating to environmental conservation and human health. A significant contributing factor to this phenomenon is the petroleum industry's production of substantial quantities of petroleum waste during the extraction, transport, storage and refining of petroleum resources. This process invariably generates a considerable amount of solid organic waste, commonly referred to as oil sludge (OS).^[1] This has been formally recognised by both the US Environmental Protection Agency (USEPA 1986) and the Directory of National Hazardous

Wastes (DNHW 2021).^[2] Prolonged storage or improper disposal of OS leads to loss of resources and serious environmental pollution.^[3]

Recent reviews have summarized (I) general OS management routes (separation, solvent/chemical cleaning, bioremediation, thermal methods), (II) non-catalytic pyrolysis performance and kinetics, and (III) selected catalyst classes in specific reactor types. However, cross-comparable evidence remains fragmented. In particular, prior syntheses rarely harmonize oil-quality descriptors (LHV/HHV, viscosity, S/N speciation), seldom link OS feed composition to catalyst acidity/redox and pore architecture at common operating windows, and under-report heavy-metal immobilization and catalyst regeneration. This review addresses these gaps by standardizing outcome metrics and mapping catalyst attributes to product slates and environmental performance across fixed-bed and microwave configurations.

OS is a complex mixture of water-oil emulsions and solids, typically composed of 30-80% oil, 30-50% water, and 10-20% solids.^[4] It exists as an emulsion stabilised by resins, asphaltenes, chemical surfactants, and solids.^[5] OS is regarded

¹Research Institute of New Chemical Technologies, L.N. Gumilyov Eurasian National University, Astana, 010008, Kazakhstan

²Department of Chemistry, Chemical Technology and Ecology, Kazakh University of Technology and Business, Astana, 010000, Kazakhstan

³Department of Chemistry, Faculty of Natural Sciences, L.N. Gumilyov Eurasian National University, Astana, 010008, Kazakhstan

*Email : aibuldinov_yek@enu.kz (Ye. K. Aibuldinov),

sabitov_as_1@enu.kz (A. S. Sabitov)

as a toxic and hazardous substance due to its stability as an emulsion, which contains numerous chemical species, including benzene, toluene, ethylbenzene, xylene, polycyclic aromatic hydrocarbons and heavy metals.^[4,6] Some mineral impurities may also be present in OS, consisting of surface soil, drilling mud residues and fine suspended particles.^[7] In addition to these constituents, OS contains hydrocarbons, which vaporise at high temperatures and generate a pungent odour, in addition to heavy metal elements, and pathogenic bacteria and microorganisms.^[8] Consequently, OS poses a grave threat to the ecological environment and human health, and due to the ease with which the oil and organic compounds it contains can contaminate soil and water bodies.^[9]

However, OS contains organic hydrocarbon components^[1] from which significant quantities of crude oil can be recovered, emphasising its significant potential for resource recovery. OS contains many hydrocarbons that can be reused as fuels and chemicals.^[10] Consequently, the development of effective methods to manage oil sludge and mitigate its environmental impact is a critical and urgent task. The treatment and recovery of oil sludge is therefore of great importance.^[11]

The disposal, separation and treatment of OS is a subject that has been extensively researched, with a range of methods being employed. These include burial,^[12] incineration,^[13–15] and thermochemical cleaning, ^s^[16,17] solvent extraction,^[18,19] ultrasound,^[20–22] microwave irradiation,^[23] and pyrolysis,^[24,25] freeze-thawing,^[26,27] centrifugal separation,^[28,29] froth flotation,^[30] mechanical separation,^[8] surfactants,^[31,32] biodegradation^[33] and combination of biodegradation and subcritical hydrothermal treatment,^[34] as well as other combined and emerging methods.^[26] However, it should be noted that these methods have both unique advantages and disadvantages. As a case in point, the incineration method is both costly and wasteful of resources. Solvent extraction is widely applied at pilot and industrial scale. Typical schemes include solvent selection, phase separation, and recycle. Reported benefits are high oil recovery and moderate operating temperatures; however, large solvent inventories and secondary emissions remain critical drawbacks requiring closed-loop handling and abatement. The utilisation of chemical surfactants, recognised for their toxicity, poses a significant environmental concern, while the availability and cost of lower environmental impact biological surfactants remains a challenge.^[35] In comparison with conventional oil sludge utilisation methodologies, OS pyrolysis emerges as a promising technology, demonstrating the capacity to recover resources and energy with enhanced efficiency and reduced environmental impact, thereby exhibiting considerable promise for industrial application.^[36] For instance, in

comparison with direct combustion, the pyrolysis process emits reduced levels of NO_x and SO_x pollutants, and the heavy metals present in OS are concentrated in the final solid product, thereby reducing the risk of secondary pollution. In contrast, the heavy metals present in fly ash (obtained from the combustion process) are easily carried away into the environment.^[14]

Pyrolysis is defined as the thermal decomposition of OS in an oxygen-free or inert atmosphere, and it is an effective method of fuel recovery from OS.^[37] The heating of oil sludge in a pyrolytic process at temperatures ranging from 400 to 800°C results in the thermal cracking of organic compounds, the condensation of low molecular weight oil products, and the formation of small molecules of non-condensable gas products. This process also yields solid coke.^[38] For instance, the predominant gaseous pyrolysis products of oil refinery sludge have been identified as CO₂, CO, and C₁–C₂ hydrocarbons, with the distillation characteristics of the petroleum product exhibiting a close resemblance to those of diesel.^[39] It has been determined that the pyrolysis products of oil sludge can possess a higher value than the original material itself,^[9] and that the liquid and gaseous by-products can be converted into chemicals and fuels.^[40] In particular, pyrolysis gas has been identified as a potential energy fuel and synthesis gas (CO, H₂).^[41] The final solid product, which typically comprises 30 to 50% by weight of the main oil sludge, can be used as adsorbents, catalysts, soil remediation agents and semi-coke.^[8,42–45] For instance, the solid product can be used to remove various pollutants, such as H₂S and NO_x, in gas streams.^[46] Furthermore, the high oil content and high calorific value of OS indicate that it has a significant energy potential.^[47,48] Considering the energy demand and environmental protection, the ultimate goal of OS treatment is to immobilise hazardous elements in the solid residue and to improve crude oil utilisation.^[49]

However, the pyrolysis process is associated with several disadvantages, including high ash content, high energy consumption, and low added value of the products, attributable to the influence of source and sludge characteristics.^[24] The presence of high oxygen concentrations in pyrolysis products results in a decrease in their energy densities, accompanied by an increase in their acidity and instability, rendering them unsuitable as fuels for use in most internal combustion engines. Furthermore, the presence of asphaltenes and carboxylic acids in heavy oil fractions has been shown to have a detrimental effect on the quality of pyrolysis oil,^[49] which is generally of a low quality.^[50] This regard, a combination of the pyrolysis process with catalytic upgrading processes has been shown to not only significantly

improve the quality of the product (by reducing its oxygen content), but also reduce the reaction time and lower the temperature.^[46] The employment of catalysts has become a common practice to enhance the quality of pyrolytic oil.^[51] Consequently, numerous studies have been dedicated to the field of catalytic pyrolysis and the impact of additives on product distribution and properties, utilising thermogravimetric analysis and laboratory reactors.^[52] The selection of the catalyst is of paramount importance, as it exerts a significant influence on the reaction pathways and the nature of the products formed.^[53] In recent times, a plethora of catalysts have been developed, encompassing metals, molecular sieves, natural ores and carbon catalysts.^[54-57]

This review article gives an overview of the recent advances in the field of co-pyrolysis of oil sludge with different types of catalysts as additives. The effect of different additives on the yield and quality of co-pyrolysis products with oil sludge, due to the synergistic effect between OS and catalysts, has also been analysed. In addition, the influence of catalysts on other factors such as adsorption characteristics of the resulting activated carbon materials, overall catalytic activity of the copyrolysis process, activation energy, carbon conversion of oil sludge, immobilisation of heavy metals in the solid residue, etc. was analysed.

2. Literature search methods

This review followed PRISMA 2020 for database search, screening, extraction, and synthesis. Web of Science and Scopus were primary sources (2010–May 2025), complemented by ScienceDirect and SpringerLink; Google Scholar was consulted for gray literature cross-checks. Core Boolean strings combined (“oil sludge” OR “oily sludge” OR “petroleum sludge”) AND (pyrolysis OR co-pyrolysis OR “microwave pyrolysis”) AND (catalyst OR additive* OR zeolite OR “metal oxide” OR CaO OR dolomite OR “red mud” OR slag). Eligibility included peer-reviewed English-language reports on catalytic (co-)pyrolysis of OS with defined additives or co-feeds under specified conditions. Exclusions: theses, abstracts, patents, non-technical reports, duplicates. Two independent reviewers screened records and extracted data into a structured template (units harmonized to wt%, kJ·mol⁻¹, mPa·s at stated T). Where necessary, values were digitized from plots (flagged in tables).

Extracted items encompassed OS origin/composition (incl. S/N and metals), additive/catalyst class (metal oxides and mixed oxides; zeolites/mesoporous silicas; basic sorbents such as CaO/dolomite/red mud/slag; minerals/clays; biomass or polymeric co-feeds), loading and particle size; reactor and operating conditions (fixed-bed or microwave, temperature

program, residence time, heating rate, carrier gas, pressure); outcomes (gas/liquid/char yields and composition; liquid LHV/HHV and viscosity; heteroatom removal *e.g.*, H₂S/N-species; gas composition is H₂/CO/CH₄; char surface area/adsorption; kinetic parameters such as activation energy; heavy-metal immobilization; and reported uncertainties). Units were harmonized (wt%, kJ·mol⁻¹, mPa·s at stated temperatures); when multiple conditions were reported, data were recorded per condition and converted to comparable metrics (*e.g.*, % change in viscosity or S removal).

Quantitative synthesis comprised parameter-based tabulation/plotting across catalyst classes and operating windows and, where ≥ 3 comparable studies existed, simple random-effects meta-aggregation of mean differences (or percentage changes); otherwise, cautious vote-counting by direction of effect was used. Qualitative synthesis linked acidity/redox and pore architecture to selectivity (paraffinic vs aromatic), de-S/de-N pathways, coke propensity, and metal immobilization, integrating evidence across fixed-bed and microwave modes. Risk of bias was appraised via a tailored checklist (feed characterization, reactor description/temperature control, replicates, calibration and standards, mass-balance closure, catalyst regeneration, statistical treatment), with domain ratings (low/unclear/high) informing narrative weighting. To structure the evidence, all included articles were classified by theme, research focus, catalyst class, and key assessed parameters as summarized in [Table 1](#).

3. Catalysts as additives for co-pyrolysis with oil sludge

With the development of research, the addition of catalyst to increase the reaction efficiency has become a common method.^[58] The initial aim of catalyst addition is to increase the pyrolysis efficiency at a lower temperature and in a shorter time. In doing so, the main results are:

- increasing the yield of pyrolysis oil and gas, especially the yield of light hydrocarbons;
- improving the quality of pyrolysis oil, such as reducing the distribution of S and N in the oil phase;
- controlling the formation and emission of pollutants, especially gas products such as ammonia and hydrogen sulfide.

Catalytic pyrolysis can be divided into in-situ catalysis (*i.e.* catalysts in the pyrolyser) and ex-situ catalysis (*i.e.* catalysts in the post-reformer). There are major types of additives used in catalytic pyrolysis such as, soluble inorganic salts, molecular sieves, metals and their oxides. Soluble inorganic salts can catalyse an activated hydration reaction that can synthesise alcohols, ketones, esters and other organic compounds. Soluble inorganic salts are also alkaline

Table 1: Classification of Scientific Articles from Bibliographic Databases.

Theme	Influence area	Additive types (examples)	Assessed metrics
Catalytic pyrolysis of OS	Product yield & quality	Zeolites (ZSM-5, Beta, Y), mesoporous silica (SBA-15, MCM-41), metal oxides (NiO, Fe ₂ O ₃ , MgO, Al ₂ O ₃ , CaO), CaO/dolomite, red mud, slag, activated carbon	Oil/gas/char (wt%), oil composition (paraffins/aromatics), LHV/HHV, viscosity, S/N in oil
Catalytic pyrolysis of OS	Process & environmental factors	FeMg-LDH, Fe ₃ O ₄ , ZnFe ₂ O ₄ , dolomite, Fe/Al-pillared clays, inert solids	H ₂ /CO/CH ₄ in gas, heavy-metal immobilization in char, H ₂ S/NH _x suppression, activation energy, emissions
Co-pyrolysis strategies	Synergy & selectivity	Biomass (sawdust, rice husk), polymers (PE, PP, PS), petroleum residues	Synergy index, light fraction yield, aromatics/olefins distribution, hydrogen transfer indicators
Reactor / operation window	Residence time & heating mode	Fixed-bed, microwave-assisted, auger; catalysts as above	Temperature window, vapor residence time, heating rate; oil stability, gas LHV
Catalyst preparation & loading	Acidity/redox & pore architecture	Impregnated/metallated zeolites (Ni/ZSM-5, Fe/ZSM-5), mixed oxides (Ni-Mg-Al), LDH-derived oxides	Acid site density (NH ₃ -TPD), redox (H ₂ -TPR), surface area/pore size (BET), optimal loading (wt% of OS)
Post-treatment & circularity	Emissions control & regeneration	ZnO beds, amine scrubbing, cyclones/filters; air/steam regeneration	H ₂ S removal efficiency, PAH/PM reduction, catalyst coke, activity recovery (%)

stimulants, which is favourable for improving the activity of the reactants and increasing the efficiency of the reaction. It has been reported that metal oxides, chlorides, sulfates and carbonates, zeolites, bentonite and some industrial wastes can also be used as efficient catalysts for pyrolysis of oil sludge.^[2, 59-61] In particular, zeolites have been used to facilitate the cracking of hydrocarbons contained in oil sludge.^[62] For example, among all catalysts, zeolite ZSM-5 is known for its excellent selectivity to monocyclic aromatic compounds, especially benzene, toluene and xylene. This is mainly due to its large number of active acid centres, high specific surface area and unique pore structure compared to other catalysts (e.g., metal oxide-based catalysts).^[63]

Molecular sieves (Molecular sieves) are a kind of aluminosilicate crystals with regular microporous structure, and they are widely used in types X, Y, ZSM-5, MCM-41, etc.^[64,65] Due to its advantages in pores and large specific surface area and unique topological structure, zeolite molecular sieve can be used as an efficient catalyst.^[63,66] The acidic Brønsted centre of zeolite can catalyse cyclisation and hydrogen transfer reactions. However, zeolites are prone to

slight deactivation due to coking phenomenon, resulting in the destruction of acidic active sites, which reduces their catalytic efficiency^[67] and this leads to a shortened catalyst lifetime.^[68,69]

Ex-situ catalysis is used to extend the lifetime of catalysts, which, compared to in-situ catalysis, can effectively mitigate catalyst deactivation, for example, by separating pyrolysis reactors and catalytic upgrading.^[70] Another disadvantage of many molecular sieves is their weak acidity and low surface density, resulting in low catalytic activity. Therefore, domestic and foreign researchers have used metals to improve their acidity and catalytic performance. Oxides and other compounds with catalytic effects are introduced into the molecular sieve, which can not only take advantage of the pores of the molecular sieve, but also take advantage of the special properties such as high activity of the oxide.^[71] The thus obtained modified molecular sieves possess metallic active sites. For example, when transition metal ions are introduced into zeolite catalysts, aromatisation can occur at the centre of the metallic Lewis acid via dehydrogenation, forming molecular hydrogen as a by-product and increasing the lifetime of the catalysts.^[72,73] Previous studies have shown

that metallic Zn can form sites in zeolite and promote aromatisation by dehydrogenating additional sites, thereby promoting biomass pyrolysis.^[66]

Metal-based catalysts are mainly transition metal elements (including Ni, Fe, Cu and Al) and are usually loaded on the carrier to improve the pyrolysis efficiency of oil sludge.^[74,75] Metal catalysts can increase the formation of pyrolysis gas^[76] and in comparison, catalysts based on metal compounds not only increase the yield of pyrolysis gas, but also increase the yield of hydrocarbons and their pyrolysis oil compounds. Such catalysts are easy to obtain and different metal compounds have different effects on pyrolysis of oil sludge, which should be selected according to specific needs.^[71]

Metal oxides have become widely used as effective catalysts in petroleum refining and catalytic cracking processes. These oxides exhibit excellent redox characteristics and high catalytic activity.^[77] In pyrolysis reactions, the catalytic action of metal oxides is primarily attributed to their specific redox properties, acidity, and basicity.^[78]

For instance, studies on the influence of additives such as CaO, K₂CO₃, Na₂CO₃, Fe₂O₃, and Al₂O₃ (as catalysts) on the properties and yield of pyrolysis products have demonstrated that they can contribute to obtaining higher-quality pyrolysis products from petroleum.^[79-83] The catalytic cracking of organic substances (OS) in the presence of aluminum compounds (Al, Al₂O₃, and AlCl₃) and iron compounds (Fe, Fe₂O₃, FeSO₄·7H₂O, FeCl₃, and Fe₂(SO₄)₃·nH₂O) has been extensively described in,^[83] where the addition of Fe₂O₃ and Fe₂(SO₄)₃·H₂O was found to enhance the quality of petroleum pyrolysis products.

Furthermore, cost-effective additives such as Fe₂O₃ and CaO can facilitate the cracking of macromolecular oxygen-containing compounds into smaller molecules and effectively promote keto-ylation and condensation of hydroxyl aldehydes. As a result, deoxygenation of acids, aldehydes, and ketones occurs.^[63,84] Additionally, these metal oxides have been shown to effectively capture heavy metals, leading to their immobilization in pyrolysis slag rather than being released into pyrolysis oil and gas, thereby reducing heavy metal emissions during the pyrolysis process.

Calcium-based compounds such as CaO, CaCO₃, CaCl₂, and Ca(OH)₂ have been widely used as pyrolysis catalysts, exhibiting both catalytic effects and sulfur stabilization.^[85] These compounds have been found to enhance the yield and quality of pyrolytic oil while increasing the reaction rate at 710 K in the following order: CaO > CaCO₃ > CaCl₂ > Ca(OH)₂.^[86] Thus, catalytic pyrolysis has demonstrated significant potential for development and research due to its high

purification efficiency and low pollution. The addition of a catalyst in the pyrolysis process can positively impact the products of petroleum sludge pyrolysis, resulting in increased oil and gas yields, reduced solid residue, and improved oil quality.^[87,88] Moreover, catalysts can modify the pyrolysis reaction conditions to shorten the pyrolysis time and lower the reaction temperature. Therefore, it is crucial to analyze experimental results and select an appropriate, cost-effective, and non-toxic catalyst while considering environmental protection.

4. Catalytic pyrolysis of oil sludge

4.1 Effect of catalysts on product yields

Pyrolysis of OS with three mesoporous zeolite catalysts (CBV 720, 760 and 780) was carried out in a fixed bed tubular reactor (stainless steel) at 450 °C to produce paraffin rich oil.^[62] Thermal pyrolysis of OS mainly produced oil (59%), gas (8%) and solid residue (33%). The higher acidity and larger total area of CBV 720 catalyst promoted higher gas (from 8 to 15 wt%), resulting in lower oil (59-47 wt%). The CBV-760 and CBV-780 catalysts showed 54% and 58% oil fraction and 10% and 5% gas fraction, respectively, and there was no significant increase in solid residue (all below 5%).

Al-MCM-41 molecular sieve as a catalyst (at dosage up to 3 wt%) was used to improve the efficiency of OS pyrolysis.^[71] The results showed that the optimum oil yield (83.48%) was observed when 1% catalyst was added at 430°C for 3 h (with Si/Al ratio equal to 60) and the oil yield did not change significantly with further increase in catalyst content (Fig. 1).

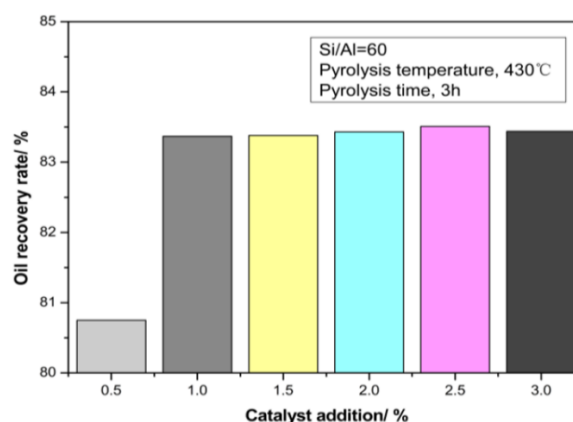


Fig. 1: Effect of catalyst concentration on the recovery of pyrolysis oil. Reproduced from.^[71]

Similar high oil recovery (up to 85.52%) was obtained by using a mesoporous molecular sieve (containing Fe) in the process of OS pyrolysis in a tube furnace reactor at a temperature of 723 K, with an optimal catalyst to OS ratio of 0.035.^[89]

New nickel ore based catalysts were used in the pyrolysis of OS in a self-made vertical electric heating furnace to produce H₂.^[90] After the addition of C-OL, C-XY, C-ID, C-OL(Ni), C-XY(Ni), C-ID(Ni) catalysts, the overall weight loss rate of OS increased by 6.03%, 2.81%, 1.85%, 11.58%, 10.05%, and 9.12%, respectively. Compared with the pyrolysis of OS alone at 900 °C, the H₂ production after the addition of C-OL and C-XY increased by 23.64% and 6.72%, respectively, while the addition of Ni-containing catalysts C-XY(Ni), C-OL(Ni) and C-ID(Ni) significantly increased the H₂ production by 68.44%, 61.80% and 34.89%, respectively. Such catalytic activity of Ni, but already in a ceramic membrane, was used in the OS pyrolysis process inside a spiral reactor to remove the generated solid particles, which are carried away from the reactor by the gas, causing clogging of downstream equipment and reducing the quality of pyrolysis oil and gas.^[91] The use of ceramic membrane (than without membrane) resulted in an increase in gas yield from 17.92 L/kg to 31.46 L/kg and a decrease in pyrolysis oil yield from 112.04 g/kg to 86.24 g/kg. In addition, the loading of Ni catalyst on ceramic membrane can effectively remove fine

particles.

The effect of KOH on the catalytic pyrolysis of 2 different OS samples was investigated in a horizontal quartz tube fixed-bed reactor at 600 °C.^[92] When KOH was added (up to 10 %), the yields of liquid products decreased for the 2 samples in the ranges of 60.1-52.8 % and 68.5-59.6 %, while the yields of gas (23.3-27.8 % and 25.6-30.9 %) and solid residues (16.6-19.4 % and 5.9-9.5 %) increased. Another study using KOH and with CaO and β -zeolite (as catalysts) in OS pyrolysis process was carried out in a tube furnace reactor to investigate the effect of catalysts on product distribution and properties.^[93] The maximum oil yield was 76.84 wt.% at 750 °C and a pyrolysis time of 60 min without catalyst (Fig. 2). Addition of KOH up to 10 wt.% (at a pyrolysis time of 30 min), as in the previous work, led to a similar result: a decrease in pyrolysis oil yield by 8.71 wt.% (from 76.45 to 67.74 wt.%) and an increase in gas by 9.85 wt.% (from 19.96 to 29.81 wt.%) (Fig. 2). The other catalysts had no significant effect on oil and gas yields.

In the next study with the same catalysts (KOH, CaO, and β -zeolite) as in previous studies, microwave pyrolysis of OS

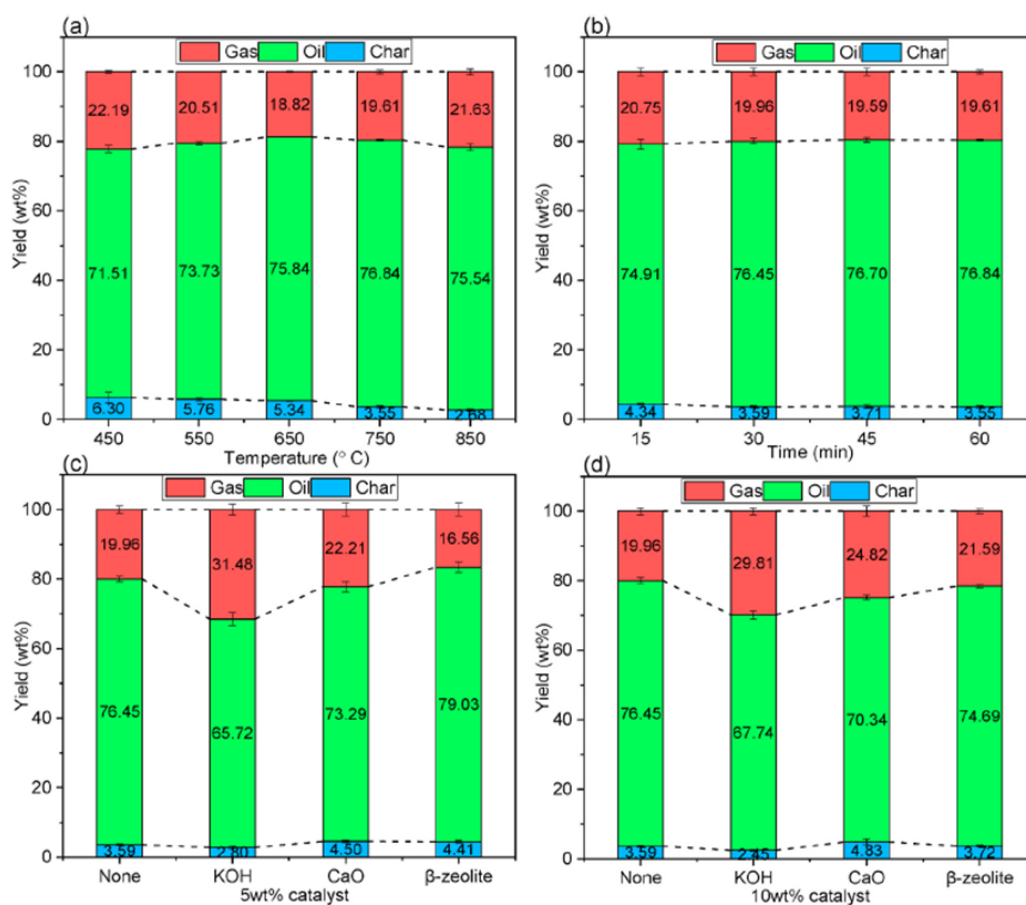


Fig. 2: Products yields under different pyrolysis conditions: (a) with different pyrolysis temperatures (pyrolysis time = 60 min); (b) with different pyrolysis time (pyrolysis temperature = 750 °C); (c) with different additives at 750 °C (pyrolysis time = 30 min, 5 wt% catalyst); (d) with different additives at 750 °C (pyrolysis time = 30 min, 10 wt% catalyst). Reproduced from.^[93]

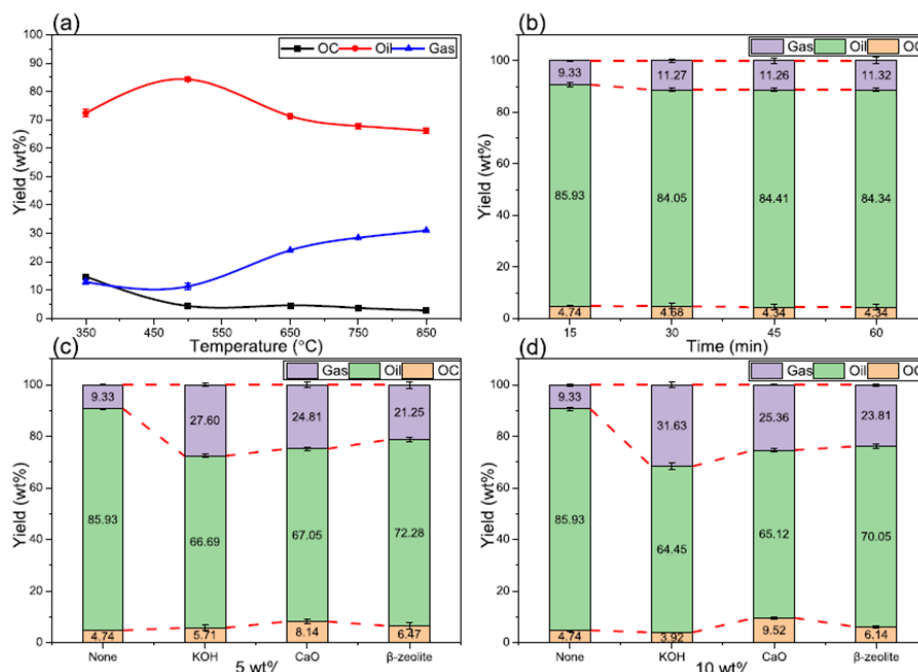


Fig. 3: Products yields under different pyrolysis conditions: (a) different pyrolysis temperatures (pyrolysis time of 60 min); (b) different pyrolysis time (pyrolysis temperature of 500 °C); (c) different additives at 500 °C (pyrolysis time of 15 min, 5 wt% catalyst); (d) different additives at 500 °C (pyrolysis time of 15 min, 10 wt % catalyst). Reproduced from.[94]

in a tubular reactor was carried out to study their influence on the characteristics of OS pyrolysis, yields, and product properties.[94] But compared to previous works, in this study, the addition of 10 wt.% KOH, CaO and β-zeolite (at 500 °C and pyrolysis time of 15 min) resulted in significantly higher decrease in pyrolysis oil yield (by 21.48, 20.81 and 15.88 %), respectively, and higher increase in gas yield (by 22.3, 16.03 and 14.48 %), respectively (Fig. 3). This seems to be due to the different operating parameters of OS pyrolysis, *i.e.* in this study the process itself was carried out in a microwave reactor and with lower values of pyrolysis temperature and time than in previous studies.

When nanometal oxides (10 wt%) were added to microwave pyrolysis OS (at 500 °C), the combustible gas (H₂ + CH₄ + CO) content increased by 18.824 wt% (MgO), 4.511 wt% (CuO), 9.28 wt% (NiO), and 16.164 wt% (γ-Al₂O₃) (Fig. 4).[38]

Catalytic pyrolysis of OS over dolomite was carried out in a U-shaped fixed bed reactor at 500 °C.[95] With increasing residence time in the range of 0-8.9 s, the oil content decreased from 76.6 to 44.2 %, while the gas and char contents increased in the ranges of 12.1-33.7 % and 11.3-22.1 %, respectively. Moreover, the increase in gas was mainly due to the increase in H₂.

The use of 2.0 wt% Fe/Al-pillared bentonite (at Fe/Al ratio = 0.5) as a catalyst in the OS pyrolysis process contributed to the optimum oil yield (increase from 29.23 %

to the highest value of 52.46 wt%), decrease in char (solid residue) yield and formation of CH₄ and CO, compared to pyrolysis in the absence of catalyst.[96] The authors attributed this enhanced oil recovery to the Fe/Al pillaring properties in bentonite, which produces a porous material with a large surface area and a Lewis acid centre on the inner surface that promotes the cracking of oil organic compounds in OS.

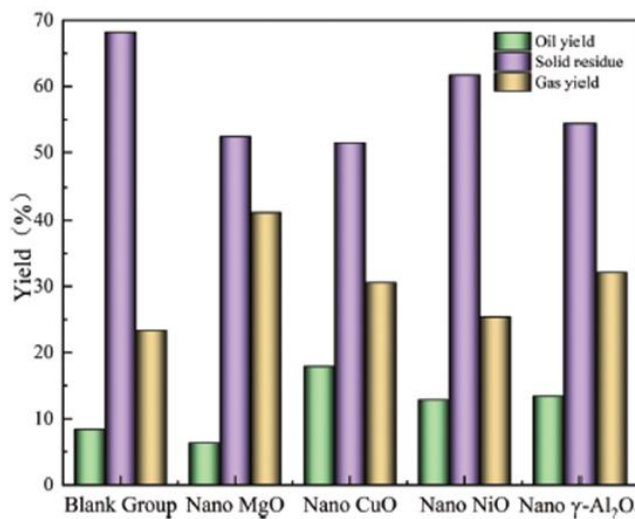


Fig. 4: Product distribution after microwave pyrolysis of oil sludge under different catalysts. Reproduced from.[38]

Another catalyst as coal gangue (CG) was used in copyrolysis which was carried out in a fixed bed reactor to

Table 2: Overview of studies on the influence of catalysts and operating parameters on the yield of copolymerization products from oil sludge.

Catalyst type	Catalyst content (wt%)	Type of reactor	Temperature (°C) and time (s or min.) of pyrolysis	Heating rate (°C/min)	Change in co-pyrolysis product yield (compared to pyrolysis without a catalyst), wt% (+ increase, – decrease)			Ref.
					Oil	Char	Gas	
Mesoporous zeolites: CBV 720 CBV 760 CBV 780	10	Fixed bed reactor	450(15 min.)	n.r.	– 12 % (59-47) – 5 % (59-54) – 1% (59-58)	+5(33-38**) +3(33-36*) +4(33-37*)	+7(8-15**) +2(8-10) –3(8-5)	[62]
KOH Sample 1	5	Fixed bed reactor	600 (n.r.)	n.r.	– 6.6(60.1-53.5)	+2.5(16.6-19.1)	+4.1 (23.3-27.4)	[92]
Sample 2	10				+2.8(16.6-19.4)	+4.5 (23.3-27.8)		
	5				– 7.3(60.1-52.8)	+4.6 (25.6-30.2)	+2.1 (5.9-8.0)	
	10				+3.6 (5.9-9.5)	30.9		
					– 6.7(68.5-61.8)			
					– 8.9(68.5-59.6)			
KOH CaO β-zeolite	5	Tube furnace reactor	750 (30 min.)	10	– 10.73(76.45-65.72) – 3.16(76.45-73.29) +2.58(76.45-79.03)	– 0.79(3.59-2.80) +0.91(3.59-4.50) +0.82(3.59-4.41)	+11.52(19.96-31.48) +2.25(19.96-22.21) –3.4(19.96-16.56)	[93]
KOH CaO β-zeolite	10				– 8.71(76.45-67.74) – 6.11(76.45-70.34) – 1.76(76.45-74.69)	– 1.14(3.59-2.45) +1.24(3.59-4.83) +0.13(3.59-3.72)	+9.85(19.96-29.81) +4.86(19.96-24.82) +1.63(19.96-21.59)	

Catalyst type	Catalyst content (wt%)	Type of reactor	Temperature (°C) and time (s or min.) of pyrolysis	Heating rate (°C/min)	Change in co-pyrolysis product yield (compared to pyrolysis without a catalyst), wt% (+ increase, - decrease)			Ref.			
					Oil	Char	Gas				
KOH CaO β-zecolite	5	Microwave reactor (1.5 kW, 2.45 GHz)	500 (15 min.)	n.r.	-	+0.97(4.74-5.71)	+18.27(9.33-27.60)	[94]			
					19.24(85.93-66.69)	+3.40(4.74-8.14)	+15.48(9.33-24.81)				
			10	500 (15 min.)	n.r.	-	18.88(85.93-67.05)		+1.73(4.74-6.47)	+11.92(9.33-21.25)	
						-	13.65(85.93-72.28)		+0.82(4.74-3.92)	+22.30(9.33-31.63)	
				800 (n.r.)	n.r.	-	-		+4.78(4.74-9.52)	+16.03(9.33-25.36)	
						21.48(85.93-64.45)	+1.40(4.74-6.14)		+14.48(9.33-23.81)		
	Nano-MgO Nano-CuO Nano-NiO Nano-γ-Al ₂ O ₃		10	Microwave reactor (2 kW, 2.45GHz)	800 (n.r.)	n.r.	-		-	+17.8(23.4-41.2)*	[38]
							2.0(8.6-6.6)**		15.8(68.0-52.2)**	+7.2(23.4-30.6)*	
							+9.3(8.6-17.9)**		16.5(68.0-51.5)**	+2.1(23.4-25.5)*	
							-		-	+8.7(23.4-32.1)*	
							+4.4(8.6-13.0)**		6.5(68.0-61.5)**	-	
							+4.9(8.6-13.5)**		13.6(68.0-54.4)**	-	
Dolomite	n.r.	U-shape fixed-bed reactor	500 (2.2 s)	n.r.	-	-	+5.2(12.1-17.3)	[95]			
					1.9(76.6-74.7)	3.3(11.3-8.0)	+18.0(12.1-30.1)				
			500 (4.4 s)		n.r.	-	-		+21.6(12.1-33.7)		
						14.6(76.6-62.0)	3.4(11.3-7.9)		+10.8(1.3-22.1)		
			500 (8.9 s)		n.r.	-	-		-		
						32.4(76.6-44.2)	-		-		

Catalyst type	Catalyst content (wt%)	Type of reactor	Temperature (°C) and time (s or min.) of pyrolysis	Heating rate (°C/min)	Change in co-pyrolysis product yield (compared to pyrolysis without a catalyst), wt% (+ increase, – decrease)			Ref.
					Oil	Char	Gas	
Fe/Al-pillared bentonites Ratio Fe/Al:	2	Fixed bed reactor	450 (180 min.)	10				[96]
0.05					+7.08	–(n.r.)	+(n.r.)	
0.1					+18.65	–(n.r.)	+(n.r.)	
0.5					+23.23	–(n.r.)	+(n.r.)	
1.0					+17.43	–(n.r.)	+(n.r.)	

n.r.: not reported

* Calculated based on weight difference

** Numerical data extracted using the PlotDigitizer software from graphical data, as these numerical values were not provided in the articles. Some values were extracted from published plots where tables were unavailable; the associated uncertainty is estimated at ± 5 –10% of the plotted value. Such entries are marked as digitized and interpreted with caution.

investigate the characteristics of pyrolysis and marine oil sludge (MOS) products.^[3] The results of catalytic pyrolysis showed that the highest oil and gas yields were 29.09 wt% and 35.18 wt% at 900 °C, respectively, while at lower temperature (700 °C), the oil and gas yields decreased to 27.51 wt% and 34.03 wt%, respectively.

Summarising the above, Table 2 summarises some of the studies on the effect of catalysts and process conditions on the yields of co-pyrolysis products with oil sludge. It can be seen that from the presented catalysts only nanometal oxides Cu, Ni, γ -Al₂O₃^[38] and Fe/Al-pillared bentonites^[96] contribute to the increase of oil and gas yields. Moreover, the effect of the latter catalyst is more significant in terms of oil yield, which is apparently related to the above-mentioned advantages (porosity, etc.). At the same time, other catalysts mainly reduce oil and gas yields. For several studies, numerical values in Table 2 were digitized from published graphs when tabulated data were unavailable. Graph-based extraction can introduce small imprecision due to figure resolution, axis scaling, and point selection, which may propagate to derived percentages or differences. Accordingly, these entries should be interpreted as approximate and used primarily to compare trends rather than exact magnitudes.

4.2 Effect of catalysts on product distribution and quality

Zeolite catalysts HZSM-5 and Zn/HZSM-5 were used in OS pyrolysis in a two-stage fixed bed tubular reactor to extract aromatic hydrocarbons.^[97] The results showed that the total aromatic hydrocarbons (TAH) yield increased from 48.7% to

92.2% when the residence time was increased from 1.0 s to 7.6 s. However, at 7.6 s, aromatic compounds were more concentrated on polyaromatics with three or more benzene rings, and 3.8 s was considered the optimum residence time for higher yield of aromatic compounds (84.8%) and highest selectivity (67.4%) of naphthalene yield. The inclusion of 3% Zn increased the TAH yield (58.7%–81.0%) and naphthalene yield (31.5%–67.5%), and also increased the H₂ and CO₂ yields and slightly decreased the coke deposit. Metal/HZSM-5 zeolite catalysts (Ni/HZSM-5, Co/HZSM-5 and Mo/HZSM-5) were also used in the catalytic pyrolysis of refinery oil sludge (ROS) in a fixed bed tubular reactor to obtain pure high quality petroleum products.^[46] Among the synthesised catalysts, the addition of 3 wt.% Ni/HZSM-5 contributed to the reduction of unfavourable oxygen, sulphur and nitrogen contents in the petroleum products and increased the calorific value of the improved product to 44.24 MJ/kg, which is within the range of natural crude oils (typically 42 to 47 MJ/kg), confirming the quality and value of the final product. The products obtained in the presence of mesoporous zeolites (CBV 720, 760 and 780) in OS pyrolysis showed selectivity for the conversion of light hydrocarbons with reduced aromatic compounds.^[62] And the low acidity of the zeolite (CBV-780) combined with its more homogeneous mesopore distribution influenced the selectivity of the secondary reactions to produce an oil product (rich in paraffins), which in turn influenced the best yield of light hydrocarbons (96%). In-situ OS pyrolysis also used HZSM-5 zeolite catalysts using different metals (Ni, Ga, Fe) already to produce aromatic

compounds (benzene, toluene, ethylbenzene and xylene).^[98] The Ni/HZSM-5 catalyst showed the highest yield of aromatic compounds (6.61 wt.%) due to better distribution of Ni on the carrier surface and higher acidity of this catalyst. Increasing the Ni loading in HZSM-5 (up to 10 wt.%) increased the BTEX yield to 13.48 wt.%. However, excessive Ni loading in HZSM-5 (15 wt.%) resulted in lower BTEXs yield due to the blockage (the blockage) of the zeolite channels. Increasing the temperature at 600-650 °C slightly decreased the BTEXs yield due to the stimulating secondary reactions at high temperatures. The highest yield of BTEXs (30.50 wt.%) was obtained by increasing the amount of catalyst

(OS/catalyst=1/3). When Al-MCM-41 molecular sieve (1% content, Si/Al ratio=60) was used in OS pyrolysis, the pyrolysis oil mainly consisted of C₁₆-C₂₀ hydrocarbons.^[71] Compared with the pyrolysis process without catalyst, the yield of the C₆-C₁₅ fraction increased significantly due to the action of the catalyst and the oil became lighter, and the saturated hydrocarbon content increased significantly.

The study of the effect of KOH on the improvement of the petroleum product quality during OS pyrolysis showed that increasing the KOH fraction up to 10 % produced more saturated hydrocarbons (C₁₄H₃₀-C₁₇H₃₆) and the concentration of asphaltenes halved (Table 3), so there was a

Table 3: Saturates, Aromatics, Resins and Asphaltenes (SARA) concentrations of oil products from pyrolysis of Guolian and Xingzhong oil sludge with and without KOH.

		Saturates (%)	Aromatics (%)	Resins (%)	Asphaltenes (%)
Guolian pyrolysis oil	Without KOH	33.8 (1.0) ^a	33.6 (1.0)	20.4 (0.8)	9.2 (1.1)
	5% KOH	32.2 (0.7)	37.9 (2.5)	22.8 (0.5)	7.1 (1.4)
	10% KOH	40.7 (0.7)	34.9 (2.2)	19.9 (1.0)	4.5 (0.9)
Xingzhong pyrolysis oil	Without KOH	38.7 (1.0)	35.0 (1.3)	10.7 (1.6)	15.6 (0.6)
	5% KOH	34.8 (0.5)	35.5 (1.0)	21.4 (0.1)	8.3 (0.7)
	10% KOH	33.6 (1.2)	44.0 (2.1)	15.5 (3.0)	6.9 (0.6)

^a Numbers in parentheses are standard deviations.

53.0 % decrease in the average molecular weight (due to the cracking reaction of heavy oil into light species).^[92] The addition of 10 % KOH also led to a significant decrease in the viscosity of the petroleum product and the greatest increase in its calorific value up to 41.1 MJ/kg, indicating an increase in the quality of the obtained petroleum products.

In another study,^[93] during the pyrolysis of OS using KOH, CaO, and β -zeolite, all catalysts significantly improved the quality of pyrolysis oil. Aromatic hydrocarbons dominated (10.01–52.69%) in the oil, and the catalysts notably reduced the presence of oxygen heterocycles. Similar results regarding the addition of KOH, CaO, and β -zeolite in OS pyrolysis were obtained in study,^[94] where the catalysts (especially KOH) enhanced the decomposition of oxygen heterocycles and increased the content of hydrocarbons and oxygenates (particularly aromatic hydrocarbons). The addition of β -zeolite resulted in the highest aromatic hydrocarbon content (64.31%) and a reduction in oxygenates in the oil, which positively affected the stability of petroleum products. The addition of KOH and CaO also reduced the content of halogenated compounds in the oil. Furthermore, the catalysts significantly increased the yields of CO, CH₄, and H₂, with KOH achieving the highest yields of CO (5.88 wt.%), H₂

(2.40 wt.%), and CH₄ (16.85 wt.%).

In the case of nano-metal oxides used in microwave catalytic pyrolysis of OS, it was found that after the addition of nano-MgO, nano-CuO, and γ -Al₂O₃ (10 wt.%), the content of heavy oil components (C₁₉₊) significantly decreased, while the content of light components (C₄-C₁₈) increased by 8.254%, 10.675%, and 11.473%, respectively.^[38] This effect is attributed to the ability of nano-metal oxides to reduce the degree of polycondensation of aromatic rings^[99] and increase the fraction of light oil components. However, after the addition of the nano-NiO catalyst, the content of light hydrocarbons decreased.^[38] Thus, nano-metallic oxide catalysts (especially γ -Al₂O₃) can facilitate the decomposition of OS, increase the proportion of straight-chain hydrocarbons and low-carbon alcohols, and improve the quality of pyrolysis oil products.

As a result of catalytic pyrolysis of OS over dolomite, a light petroleum product with 57.0% saturated hydrocarbons (an increase of 45.0%) was obtained due to the conversion of aromatic compounds and heavy hydrocarbons into light aliphatic hydrocarbons, such as straight-chain hydrocarbons.^[95] The asphaltene content decreased by 88.5%, reaching less than 5%. Consequently, the lower heating value of the petroleum

product reached a maximum of $37.2 \text{ MJ}\cdot\text{kg}^{-1}$. Calcium in dolomite reduced H_2S emissions and sulfur content in pyrolytic oil, likely due to the decomposition of thiophenes on dolomite. Additionally, dolomite deactivation occurred, caused by coke deposition and CaO carbonation.

The addition of 2.0 wt% Fe/Al-pillared bentonite (at a Fe/Al ratio of 0.5) in the OS pyrolysis process led to an increase in the content of aliphatic compounds (C_{13-19}) and a reduction in the content of heterocyclic compounds in the liquid products, which favored the use of pyrolysis oil as fuel, such as diesel fuel.^[96]

A 20.0 wt.% CaO was used as a neutralizing agent, solidificator, and sulfur-binding compound to stabilize OS during pyrolysis. CaO facilitated char gasification (via $\text{CaSO}_4 + 2\text{C}_{\text{char}} = \text{CaS} + 2\text{CO}_2$), resulting in a low carbonaceous residue (<2.0 wt.%).^[79] The liquid products consisted of paraffins (76.1 vol.%), olefins (10.8 vol.%), and aromatic compounds (13.1 vol.%). The relatively low aromatic content and low sulfur content (0.15 wt.%) indicated that the obtained oil exhibited properties similar to liquid fuel, with a composition corresponding to heavier petroleum fractions such as furnace fuel which aligned well with its density and higher heating value of $42.1 \pm 2.6 \text{ MJ/kg}$.

The addition of 5% CoCl_2 to the OS pyrolysis process (at $500 \text{ }^\circ\text{C}$) led to an increase in the heating value of liquid products (up to 42.31 MJ/kg).^[100] The authors attributed this to an increased amount of cyclic, aromatic, and unsaturated hydrocarbons. At the same time, the total combustion heat of gaseous products increased by 2.2 times due to the higher content of light hydrocarbons. Moreover, the catalyst remained entirely within the mineral fraction, consisting of silica particles coated with pyrolytic graphite, and could be utilized as a porous sorbent.

Nanopowders of Al_2O_3 (nano-alpha aluminum oxide and nano-gamma aluminum oxide) also enhanced the heating value of petroleum products during catalytic OS pyrolysis.^[54] The characteristics of the obtained liquid fuel corresponded to the diesel fraction, with a heating value of $46,073 \text{ kJ/kg}$ and a density of 845 kg/m^3 under optimal conditions (temperature of $400 \text{ }^\circ\text{C}$, heating rate of $30 \text{ }^\circ\text{C/min}$, holding time of 30 min). A total of 550 L of diesel fraction was obtained from 1 m^3 of OS. This catalyst improved the quality of the resulting fuel, although it slightly reduced its yield. The sulfur content in the fuel was also reduced by more than 94% compared to OS.

Aluminum (Al_2O_3 , AlCl_3) and iron (Fe , Fe_2O_3 , $\text{FeSO}_4\cdot 7\text{H}_2\text{O}$, FeCl_3 , $\text{Fe}_2(\text{SO}_4)_3\cdot n\text{H}_2\text{O}$) compounds were used as catalysts for OS pyrolysis, where only Fe_2O_3 and $\text{Fe}_2(\text{SO}_4)_3\cdot n\text{H}_2\text{O}$ improved the quality of the oil, although all catalysts increased OS conversion and reaction rates.^[101] In a

study on the effect of inherent minerals (SiO_2 , CaO , MgO , Fe_2O_3 , and Al_2O_3) on OS pyrolysis, it was found that the presence of SiO_2 in OS had an insignificant impact on OS pyrolysis, whereas CaO and MgO contributed to nearly unchanged liquid product quality, the lowest content of total petroleum hydrocarbons (TPHs) in solid residues, and the acceleration of TPH volatilization from OS.^[102] Similar effects were observed with Fe_2O_3 and Al_2O_3 , which also promoted the formation of more gases and long-chain liquid products, although their overall yield decreased. Additionally, natural minerals inhibited the release of pollutants (containing N/S/Cl) during the dehydration and devolatilization stages (< 358°C) but facilitated their release during thermal cracking and secondary reactions (> $358 \text{ }^\circ\text{C}$).

Magnetic ZnFe_2O_4 nanoparticles, due to their high dielectric permittivity, were used as a novel type of microwave-absorbing medium to accelerate the microwave pyrolysis process of OS.^[103] With the addition of ZnFe_2O_4 nanoparticles, the $\text{C}_4\text{--}\text{C}_{12}$ content increased by 3.5%, while the C_{18+} content decreased by 4.1%, indicating that more recombinant components participated in the pyrolysis reaction, leading to the formation of light gaseous components. ZnFe_2O_4 magnetic nanoparticles were also employed as catalysts for OS pyrolysis,^[104] where an increase in the mass concentration of ZnFe_2O_4 nanoparticles resulted in higher H_2 , CH_4 , and C_2H_6 content in the pyrolysis gas products.

In study,^[104] an innovative method for obtaining Fe-char from OS was proposed, which was subsequently applied in the catalytic cracking of heavy oil in OS. The oil conversion rate using Char900 (90.2%) at 900°C was higher than that using Char600 (80.1%) at 600°C . With Char900, polyaromatic compounds in the petroleum product were decomposed into monocyclic aromatic compounds, while the yields of H_2 and CO significantly increased. At 800°C , the oil conversion rate reached 95.8%, and the CO yield was 9.5 times higher than under non-catalytic conditions. The catalytic effect of Char900 can be attributed to the combined effect of char and Fe° , which plays a crucial role as a catalytic medium in facilitating hydrocarbon reforming and the water-gas shift reaction.

A study on the effects of different catalysts (walnut shell, Fe_2O_3 , K_2CO_3 , polyvinyl chloride (PVC), and OS pyrolysis char) on the efficiency of OS pyrolysis at low temperatures revealed that with the addition of catalysts at 450°C , the content of gasoline, diesel fuel, and jet fuel decreased, whereas distillates and heavy oil tended to increase.^[59]

The catalytic pyrolysis of marine oil sludge (MOS) with coal gangue contributed to the reduction of heteroatomic compounds in the pyrolysis oil.^[3] The yield and selectivity of aromatic compounds in the oil were 28.74 wt.% and 98.79%,

respectively, while the hydrogen content in the gas reached 46.93 vol.%. The highest CH₄, CO, and H₂ yields at 700°C were 140.63 g/kg (or 42.28 vol.%), 147.75 g/kg, and 22.32 g/kg, respectively. The concentration of heavy metals (HMs) in the pyrolysis char met standard requirements. Catalytic pyrolysis facilitated the migration of N and C₁ from the pyrolysis oil into the gas phase.

4.3 Influence of catalysts on other factors

During the catalytic pyrolysis of OS using novel nickel ore-based catalysts, it was found that their addition increased the overall weight loss rate of OS in the range of 6.03–11.58%.^[90] These catalysts can promote the cracking of tar, thereby producing more pyrolysis gas, with a significant increase in H₂ content.

In studies,^[93,94] the addition of KOH, CaO, and β-zeolite catalysts to OS pyrolysis enhanced the iodine and methylene blue adsorption values in char, indicating its potential use as an adsorbent.

Layered double hydroxide (LDH) FeMg (FeMg LDH) was used as a catalyst for OS pyrolysis to reduce emissions of pollutants such as amide and heterocyclic nitrogen species and aliphatic and disulfide sulfur compounds, which pose significant environmental challenges.^[105] It was found that FeMg LDH could effectively suppress the emissions of these pollutants.

The use of the microwave-absorbing material Fe₃O₄ in microwave OS pyrolysis led to an increase in the weight loss coefficient from 13.0% to 14.1% and a reduction in activation energy and enthalpy values.^[106] This suggests that Fe₃O₄ partially catalyzes the OS pyrolysis process. Moreover, the addition of Fe₃O₄ increased the diversity of gaseous products, with the number of molecular species rising from 5 to 46 at temperatures of 200–300°C.

When magnetic ZnFe₂O₄ nanoparticles were introduced into microwave OS pyrolysis,^[103] the activation energy of OS decreased by 36.49%, and the pre-exponential factor was reduced by 91.39% in Stage III (the cracking stage of heavy components at 240–300°C), indicating the strong catalytic activity of ZnFe₂O₄ nanoparticles.

5. Catalytic Co-pyrolysis of oil sludge with additives

5.1 Effect of catalysts on product yield

The co-pyrolysis of OS with polyethylene (PE) waste in the presence of an acidic catalyst (K-10® clay) was studied in a stainless steel tubular reactor.^[107] The addition of PE waste to oil sludge reduced oil production, while the addition of the catalyst (5 or 10 wt%) at 450°C led to a 50% increase in gas yield. High-density polyethylene (HDPE) was also used in

catalytic co-pyrolysis of OS with modified Y zeolites (HUSY) (at 450°C and atmospheric pressure for 15 minutes) to produce oil products rich in paraffins.^[108] Without catalyst additives, the oil, gas, and solid residue yields were 56%, 5%, and 39%, respectively. Among the tested catalysts, the highest oil yields were observed with zeolites Mod-Si-10 (59%) and Mod-Al (63%), while other zeolites reduced oil yield compared to the non-catalytic process. The highest gas yields were obtained with zeolites CBV-720 (13%) and Mod-Si-5 (11%). The lowest oil (41%) and gas (3%) yields and the highest solid residue (56%) were recorded with zeolite Mod-Si-20.

The co-pyrolysis of peat and oil sludge in the presence of bentonite clay (a metal-containing aluminosilicate) was conducted in a fixed-bed reactor under various conditions (350–650°C, catalyst loading: 1–30 wt%).^[109] The co-pyrolysis of peat and oil sludge in the presence of bentonite clay resulted in an increased gas product yield (from 18 to 26 wt%) and liquid fraction yield (from 45 to 55 wt%). Additionally, the use of bentonite clay led to a higher concentration of C₁–C₃ gases (up to 10–20%). However, in the co-pyrolysis of petroleum residue with lignite under similar technological conditions, no synergistic effect was observed.^[110]

5.2 Effect of catalysts on product distribution and quality

Microwave catalytic co-pyrolysis of *Chlorella vulgaris* (CV) and OS was investigated using a microwave reactor with NiO, activated carbon (AC), and their 1:1 combination (N₁A₁) with various additive loadings (5%, 10%, 15%, and 20%).^[111] The optimal mixing ratio for co-pyrolysis was determined to be CV/OS = 1:1 (C₁O₁) with 10% AC. The strongest synergistic effect from the interaction between NiO and AC was observed in the 10% N₁A₁ group. Hydrocarbon content in bio-oil from the C₁O₁ group increased by 31.84%, while nitrogenous and oxygenated compounds decreased by 74.18% and 19.01%, respectively. The addition of 10% N₁A₁ to the C₁O₁ group further increased the content of aliphatic hydrocarbons by 22.44% and reduced nitrogenous and oxygenated compounds by 41.79% and 36.58%, respectively, contributing to the overall production of high-quality biofuel in the C₁O₁ group.

A similar study on the microwave catalytic co-pyrolysis of *Chlorella vulgaris* and OS was conducted using catalysts (Ni/AC, Ni-Cu/AC, and Ni-Fe/AC), where activated carbon (AC) served as a support material.^[112] The results showed improved pyrolysis characteristics and enhanced oil quality. The addition of 10% catalysts exhibited a stimulating effect on bio-oil production, with the highest yield (22.1%) obtained in the 10% Ni-Cu/AC group. The addition of 10% Ni-Fe/AC increased the content of aliphatic hydrocarbons in bio-oil to

44.52%, while 10% Ni/AC resulted in the highest hydrocarbon content at 76.62%.

In the catalytic co-pyrolysis of two different refinery oil sludge (ROS) samples with sawdust (SD) (at 500–600°C), it was found that catalytic pyrolysis of the first sample had no significant effect on oil yield; however, high hydrocarbon, phenol, and aromatic compound contents were obtained in the presence of molecular sieve (MS), metallic slag, and ZSM-5, respectively.^[113] The catalytic co-pyrolysis of the second sample with SD in the presence of MS increased oil yield, with the final product containing a high proportion of hydrocarbons (~54%) and aromatic compounds (~44%).

In the co-pyrolysis of OS and HDPE, the addition of modified Y zeolites (Mod-Si-10 and Mod-Al) enabled a high conversion into paraffinic products.^[108] For these zeolites, the proportions of light and heavy hydrocarbons were 72/28(%) and 79/21(%), respectively, while the distribution by hydrocarbon type (aromatic, paraffinic, and cyclic) was 5/87/8(%) and 3/91/8(%), respectively. Similar results regarding the selective conversion of light hydrocarbons with reduced aromatic compound content were obtained in a previously described study using mesoporous zeolite catalysts (CBV 720, 760, and 780).^[62]

In the co-pyrolysis of oil sludge and cellulose, catalysts such as Ni/HZSM-5, HZSM-5, HBeta, HY, and Al-MCM-41 were used to enhance the production of aromatic compounds.^[114] TGA results indicated that Ni/HZSM-5 exhibited the lowest apparent activation energy (97.6 kJ/mol) due to the reduction in process temperature, followed by HY, HBeta, HZSM-5, and Al-MCM-41. The presence of Ni in the Ni/HZSM-5 catalyst provided additional cracking and led to the highest production of benzene, toluene, ethylbenzene, and xylenes (BTEXs) due to the optimal pore size of HZSM-5 and the synergistic effect of Ni. The synergistic effect was confirmed as the experimental BTEX yields exceeded the theoretical values for all catalysts. The strongest synergistic effect was achieved at a cellulose-to-oil sludge ratio of 1:3.

The primary types of plastic found in municipal solid waste include high-density polyethylene (HDPE), low-density polyethylene (LDPE), and polypropylene (PP). These polyolefins were used in the pyrolysis of dry oil sludge (DS) in the presence of various mesoporous Y-zeolites with different acidity levels (CBV 720 > CBV 760 > CBV 780).^[115] The experiments were conducted in a fixed-bed reactor at 450°C for 15 minutes. It was found that the linear structure of polyolefins facilitated the formation of paraffinic products, whereas tertiary chains hindered the formation of linear and paraffinic compounds due to possible steric hindrance. The CBV 720 catalyst enabled the production of paraffinic

products with reduced aromatic and cyclic compounds when polyolefins with a linear structural chain were used. CBV 760 and CBV 780 catalysts yielded inconsistent (discrete) results and were ineffective in producing paraffinic products in the light hydrocarbon fraction.

In the co-pyrolysis of oil sludge and two types of polyethylene waste high-density polyethylene (HDPE) and low-density polyethylene (LDPE) with the addition of an acidic catalyst (K-10® clay), the resulting oil primarily consisted of paraffins (over 80 wt%) and small amounts of olefins and aromatics (each below 10%).^[24] As the polyethylene waste content increased, the proportion of heavy hydrocarbons in the oil also increased. Pyrolysis oil from the thermal pyrolysis of OS with HDPE (5 wt%) mainly contained light hydrocarbons, while the oil obtained with LDPE consisted of 64% heavy hydrocarbons. Catalytic co-pyrolysis produced oil with a higher content of light hydrocarbons compared to oil from thermal pyrolysis. The oil from catalytic co-pyrolysis was predominantly composed of paraffins with low levels of olefins and aromatic compounds. Additionally, the average calorific value of the oil (44.1 MJ/kg) was similar to the default calorific value for diesel fuel (43.7 MJ/kg), suggesting its potential for energy applications.

The co-pyrolysis of scrap tires (ST) with oily wastes (OW) was conducted in the absence and presence of catalysts (red mud and a commercial ReUS-Y catalyst used in fluid catalytic cracking units at refineries) at 400 and 500 °C.^[116] Both catalysts demonstrated similar catalytic activity in waste degradation. Although catalytic use resulted in a lower yield of pyrolysis oil, the oil contained more light hydrocarbons than that obtained from thermal degradation. Pyrolysis oil from ST contained a significant amount of aromatic compounds, whereas the co-pyrolysis of both wastes (with and without a catalyst) produced oil with a high paraffin content and a low concentration of aromatic compounds. The results suggested that co-pyrolysis of ST with OW yields oil suitable for fuel applications.

A reduction in aromatic compounds was also observed in another similar study on the co-pyrolysis of scrap tires with oily wastes in a fixed-bed reactor, both in the absence and presence of catalysts (zeolite-based catalyst and red mud) at 500 °C.^[117] The catalysts had no significant effect on product yield or composition. The amount of aromatic and olefinic compounds (undesirable for fuel applications) in the oil from co-pyrolysis was lower than in the oil derived from scrap tires alone. The fuel characteristics of the co-pyrolysis oil except for flash point and sulfur content such as density, viscosity, and calorific value, were comparable to commercial diesel fuel. The higher calorific value of the pyrolysis gases ranged from

20.4 to 26.4 MJ/Nm³. It was also found that the concentration of metallic impurities in the resulting oil was less than 0.3 ppm, which was significantly lower than in the original oily wastes.

5.3 Effect of catalysts on other factors

To study the thermodynamics and synergistic effects, catalytic co-pyrolysis of OS and walnut shell (WS) was conducted using thermogravimetric analysis in the temperature range of 50–850 °C.^[49] It was found that the heating rate (10–40 °C/min) did not significantly affect the pyrolysis process. The addition of walnut shell could coat OS, which favorably influenced the completeness of the pyrolysis process. Catalytic co-pyrolysis predominantly produced aromatic compounds, which could be attributed to the unique pore structure of ZSM-5 zeolites, as also confirmed in previous studies.^[97,98,114] The strongest synergistic effect of co-pyrolysis was observed at an OS content of 25 wt% and was further enhanced by ZSM-5 zeolites, which accelerated the pyrolysis process. The activation energy after the addition of the catalyst was significantly lower than without it, indicating that Zn-ZSM-5 facilitates the co-pyrolysis process.

ZSM-5 zeolite was also used for producing aromatic platform chemicals in the catalytic co-pyrolysis of OS and rice husk (RH).^[63] As a result of non-catalytic co-pyrolysis of RH and OS, the initial pyrolysis temperature of RH was significantly reduced (by approximately 55 °C) at lower temperatures, and the weight loss coefficient of OS decreased (by about 10–18 wt%) in the higher temperature range. Oxygenated and aliphatic compounds from non-catalytic co-pyrolysis were effectively converted into aromatic compounds with a maximum yield of 46%, while the selectivity toward benzene, toluene, and xylene increased up to 60%. The ZSM-5 catalyst facilitated the degradation of OS and RH mixtures at temperatures below 150 °C but had a negligible effect on weight loss at higher temperatures.

A significant reduction in heavy oil viscosity was achieved through co-pyrolysis of OS with sawdust using calcium naphthenate as a catalyst.^[118] Under optimal reaction conditions (reaction temperature of 350 °C, reaction time of 30 minutes, sawdust content of 5 wt%, and catalyst content of 0.05 wt%), the viscosity reduction ratio reached 98%. Additionally, the degree of heavy oil cracking significantly increased during catalytic co-pyrolysis, leading to a sharp decrease in heavy oil viscosity after the reaction.

6. Conclusion

This review shows that catalytic pyrolysis of oil sludge can (i) raise liquid and gas yields and fuel quality, (ii) suppress toxic N/S-containing compounds, and (iii) reduce energy demand

by lowering temperature and residence time. The outcome is governed jointly by catalyst chemistry/texture and the operating window.

Zeolite-centered systems (including metal-modified HZSM-5) as well as KOH, CaO, and β -zeolite tend to promote aromatics, whereas mesoporous zeolites, dolomite, nano-MgO/CuO/ γ -Al₂O₃, modified Y-zeolites, and K-10 clay favor light paraffins. Dolomite, CoCl₂, Ni/HZSM-5, and Al₂O₃ can increase calorific value while reducing S and N in the liquids; CaO efficiently binds sulfur, and FeMg-LDH helps suppress N/S-bearing pollutants. Because identical catalysts (*e.g.*, KOH, CaO, β -zeolite) can perform inconsistently across reactor configurations, careful co-optimization of reactor type, temperature, and residence time is essential.

Unresolved gaps include: (1) robust links between OS feed composition and catalyst acidity/redox/pore architecture under common temperature–time windows; (2) standardized reporting of oil quality (LHV/HHV, viscosity at temperature, S/N speciation) and gas composition (H₂/CO/CH₄) enabling meta-aggregation; (3) heavy-metal immobilization and leachability protocols for char aligned with fuel and environmental end-uses; (4) coke propensity and regeneration cycles across catalyst classes (zeolites, metal oxides, LDHs, industrial residues); (5) techno-economic assessments with recycle/heat-integration; and (6) scale-up studies comparing fixed-bed vs microwave-assisted systems with common mass-balance and emission controls.

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Conflict of Interest

There is no conflict of interest.

Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Supporting Information

Not applicable.

CRedit Statement

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