
COMPARATIVE STUDY OF THERMAL, ZEOLITE-Y CATALYTIC AND NI-DOPED ZEOLITE-Y CATALYTIC CRACKING OF GAS CONDENSATE

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DOI: <https://doi.org/10.67203/sp.ICESID.2026>

ABSTRACT

This study investigated the modification of zeolite-Y and its application in the cracking of gas condensate using thermal, catalytic, and nickel-doped catalytic processes. Raw zeolite-Y (ZY-1) was chemically pretreated (ZY-2) and subsequently modified with nickel to produce Ni-doped zeolite-Y (ZY-3). The catalysts were characterized using Brunauer–Emmett–Teller (BET) analysis, X-ray diffraction (XRD), and Fourier-transform infrared spectroscopy (FTIR), while the hydrocarbon products were evaluated by GC-MS/PIONA analysis. BET analysis showed progressive increases in the surface area from 181.549 m²/g for ZY-1 to 245.368 and 278.956 m²/g for ZY-2 and ZY-3, respectively, accompanied by increases in total and micropore volumes. XRD confirmed the preservation of the characteristic zeolite-Y framework after modification, with additional reflections in ZY-3 consistent with crystalline NiO. FTIR further confirmed the retention of the principal aluminosilicate framework vibrations following pretreatment and nickel incorporation. GC–MS/PIONA analysis revealed substantial changes in the hydrocarbon-class distribution following cracking. The raw gas condensate (G-1) contained 47.47% paraffins and 30.49% aromatics, while thermal cracking (G-2) produced 46.10% paraffins and 32.10% aromatics. Catalytic cracking (G-3) increased the aromatic fraction to 40.90% while reducing the paraffin fraction to 38.40%. Ni-doped catalytic cracking (G-4) resulted in 39.48% aromatics, 39.98% paraffins, and a slight increase in olefins from 0.40% to 0.91% relative to G-3. Overall, the results demonstrate that pretreatment and nickel incorporation significantly modified the textural and structural properties of zeolite-Y, while catalytic and Ni-doped catalytic cracking produced distinct changes in the gas-condensate hydrocarbon composition.

Keywords: Zeolite-Y; Nickel-doped Zeolite-Y; Gas condensate; Catalytic cracking; Thermal cracking

INTRODUCTION

The increasing demand for transportation fuels, petrochemical intermediates, and high-value hydrocarbon products has intensified interest in technologies that can convert and improve the efficiency of underutilized petroleum fractions into more valuable products. Among these resources, natural gas condensate is an attractive feedstock because it consists predominantly of relatively light hydrocarbons and can serve as an intermediate source of gasoline-range and petrochemical hydrocarbons. Gas condensates have attracted attention not only as blending components but also as feedstocks for catalytic upgrading, particularly where their hydrocarbon distribution can be selectively shifted toward products with improved octane quality and greater commercial value (Salim & Saed, 2023; Ganiyu et al., 2026). Recent work on Nigerian gas condensate has further demonstrated the potential of catalytic conversion over modified zeolitic materials to alter the distribution of paraffins, naphthenes, aromatics, and isoparaffins to obtain more desirable gasoline-range products (Ganiyu et al., 2026).

The conversion of petroleum-derived hydrocarbon streams is driven by competing pathways, such as thermal and catalytic cracking. While thermal cracking operates at high temperatures and often produces undesirable secondary reactions, such as excessive coke and gas, catalytic cracking offers better product control at lower temperatures. Ultimately, the specific pore architecture, acidity, and stability of zeolite catalysts determine the exact cracking pathways and product selectivity for complex feeds (Wei et al., 2025; Magaji et al., 2024).

Zeolites are highly attractive catalytic materials for hydrocarbon upgrading because their crystalline frameworks combine tunable acidity with precise microporous structures that ensure shape selectivity while promoting key chemical reactions. Despite these benefits, narrow micropores often impose severe diffusion limitations when handling large or complex petroleum fractions, thereby restricting access to internal acid sites and increasing both secondary reactions and coke deposition (Dong et al., 2024; Wei et al., 2025). Zeolite-Y is widely used in petroleum refining due to its large-pore architecture and established role in fluid catalytic cracking, yet even conventional zeolite Y topologies face mass-transport constraints when processing bulky hydrocarbon feeds. To mitigate these transport limitations, current research focuses on designing hierarchical Y zeolites that combine intrinsic micropores with larger mesoporous pathways to balance selectivity, molecular transport, and structural stability. Recent studies have demonstrated that hydrothermal and post-synthetic modifications of Y zeolites successfully engineer these hierarchical networks, thereby significantly improving acid-site accessibility and hydrocracking performance (Zhou et al., 2024; Wei et al., 2023).

The incorporation of nickel into zeolitic materials introduces additional metal-associated functionalities that can complement the intrinsic acidity of the zeolite, thereby influencing hydrocarbon conversion and product distribution through synergistic metal–acid interactions.. The interaction between nickel and the framework directly influences acidity, active site dispersion, and accessibility. Recent work has demonstrated that nickel-modified hierarchical zeolites enhance hydrocracking performance and mass transfer by combining metal functionality with interconnected larger pores rather than relying on metal loading alone (Zhang et al., 2024; Hu et al., 2024).

Despite these advances, an important gap remains in understanding how controlled Zeolite Y pretreatment, followed by nickel incorporation, affects the structural characteristics required for the catalytic cracking of actual gas condensates, particularly under continuous-flow conditions. Most existing studies focus on modeling hydrocarbons or heavier feeds, overlooking the integrated relationship between modified Zeolite Y, nickel active sites, and complex condensate streams. This study addresses these limitations by systematically examining the structure-property relationship of raw, pretreated, and Ni-modified Zeolite Y and its effect on pore accessibility and hydrocarbon transformation.

2. MATERIALS AND METHODS

2.1 Materials

The materials used in this study included commercial zeolite-Y, nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), sodium hydroxide (NaOH), ammonium chloride (NH_4Cl), deionized water, nitrogen (N_2), glass beads, and gas condensate. Commercial zeolite-Y (ZY-1) was used as the parent material for the preparation of the pretreated zeolite-Y (ZY-2) and Ni-doped zeolite-Y (ZY-3) catalysts. Nickel nitrate hexahydrate was used as the nickel precursor, while NaOH and NH_4Cl were employed during the alkaline treatment and conversion of the zeolite to the acidic H-form, respectively. Deionized water was used to prepare the treatment and precursor solutions, while glass beads served as an inert packing material in the fixed-bed reactor. Nitrogen was used as the carrier/diluent gas during the cracking experiments.

The gas-condensate feedstock was obtained from the Ogunu field in the Niger Delta region of Nigeria. The raw gas condensate was designated **G-1**, while the products obtained after thermal cracking, catalytic cracking over ZY-2, and catalytic cracking over ZY-3 were designated G-2, G-3, and G-4, respectively. All materials and reagents were used as received, unless otherwise stated.

2.2 Catalyst Preparation

2.2.1 Preparation of Pretreated Zeolite-Y (ZY-2)

Commercial zeolite-Y (20 g) was initially calcined in a muffle furnace at 550 °C for 5 h. The calcined zeolite was subsequently treated with 100 mL of 0.5 M NaOH solution at 70 °C for 2 h under continuous stirring. After alkaline treatment, the solid was repeatedly washed and centrifuged with deionized water while monitoring the pH until neutrality (pH 7) was achieved. The washed material was dried at 90 °C for 1 h and subsequently converted to the acidic H-form by treatment with 100 mL of 0.5 M NH₄Cl solution at 70 °C for 3 h under continuous stirring. The resulting material was dried at 100 °C for 2 h and calcined at 550 °C for 5 h to obtain the pretreated zeolite-Y, designated ZY-2.

2.2.2 Preparation of Ni-Doped Zeolite-Y (ZY-3)

Nickel-doped zeolite-Y was prepared using the incipient-wetness impregnation (IWI) method using Ni(NO₃)₂·6H₂O as the nickel precursor. The required quantity of Ni(NO₃)₂·6H₂O was calculated to obtain a target nickel loading of 5 wt.% and dissolved in 100 mL of deionized water. The resulting solution was added dropwise to the pretreated zeolite-Y (ZY-2) under continuous stirring and subsequently aged for 4 h at ambient conditions. The impregnated material was dried at 120 °C for 12 h and then calcined at 550 °C for 4 h to obtain the Ni-doped zeolite-Y catalyst, designated as ZY-3.

2.2.3 Catalyst Characterization

The structural and textural properties of the prepared catalysts were evaluated using powder X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FT-IR), and nitrogen (N₂) adsorption–desorption analysis. Analyses were performed on raw Zeolite-Y (ZY-1), pretreated Zeolite-Y (ZY-2), and Ni-doped Zeolite-Y (ZY-3) to determine the effects of pretreatment and nickel incorporation on the zeolite structure and textural properties.

Powder X-ray diffraction (XRD) patterns were recorded using a PANalytical X'Pert PRO MPD diffractometer equipped with Cu K α radiation (λ = 0.1541 nm), operated at 40 kV and 30 mA, over a 2θ range of 5–60° at room temperature. The diffraction patterns were used to identify the characteristic crystalline phases and assess changes in the crystallinity and structural integrity of the Zeolite-Y framework following pretreatment and Ni incorporation.

The Fourier transform infrared (FT-IR) spectra of ZY-1, ZY-2, and ZY-3 were obtained using an Agilent MicroLab instrument to identify characteristic zeolite framework vibrations and surface functional groups. The spectra were compared to identify changes in the zeolite's structural features resulting from alkaline treatment, conversion to the acidic form, and subsequent nickel incorporation.

N₂ adsorption–desorption measurements were conducted using an ASAP 2020 V4.02 analyser at 77 K to determine the textural properties of the catalysts. Before analysis, each sample was evacuated under vacuum at 573 K for 8 h. The BET method was used to determine the specific surface area. The total pore volume was determined from the amount of nitrogen adsorbed at P/P_0 = 0.9915, while the pore-size characteristics were obtained from the adsorption–desorption data.

2.3 Catalytic Cracking of Gas Condensate

The catalytic cracking of gas condensate was conducted at atmospheric pressure in a continuous-flow fixed-bed reactor using nitrogen (N₂) as the carrier/diluent gas. Prior to each experiment, the reactor bed was purged under a continuous N₂ flow of 400 mL/min at 550 °C for 1 h. The N₂ supply pressure was maintained at 3 bar at the regulator, whereas the reactor itself remained at atmospheric pressure.

Thermal cracking was first conducted under the same reaction conditions to provide a baseline for comparison with catalytic cracking. Both thermal and catalytic experiments were performed at 550 °C, atmospheric pressure, and reaction duration of 1 h. For the catalytic experiments, 0.5 g of either ZY-2 or ZY-3 was mixed with 0.5 g of glass beads and packed into the reactor. The gas-condensate feed was introduced continuously using a positive-displacement peristaltic pump at 4 mL/min, while the N₂ flow rate was maintained at 400 mL/min. The heating lines were maintained at 100 °C.

The reactor effluent was passed through a condenser maintained at 2.5 °C to recover the condensable liquid products, while the uncondensed gaseous products were collected in a gas collection bag. The condensed liquid products were subsequently analyzed using an Agilent 7890 gas chromatograph equipped with a DB-624 capillary column. The identified compounds were subsequently classified according to the PIONA groups (paraffins, isoparaffins, olefins, naphthenes, and aromatics).

The apparent liquid recovery was calculated as the ratio of the recovered condensable liquid to the total feed introduced, as expressed in Equation (1):

$$\text{Liquid recovery(\%)} = \frac{V_{\text{Liquid recovered}}}{V_{\text{feed}}}$$

3.0 RESULTS AND DISCUSSION

3.1 Surface Functional Group Analysis

The FTIR spectra for raw Zeolite-Y (ZY-1), pretreated (ZY-2), and Ni-doped (ZY-3) are shown in Figure 1. The FTIR spectrum of raw zeolite-Y (ZY-1), recorded over approximately 400–4000 cm^{-1} , exhibited the characteristic vibrational features of an aluminosilicate zeolite, with prominent bands at approximately 459, 529, 613, 837, 1080, 1209, 1408, and 1645 cm^{-1} . The strong absorption near 1080 cm^{-1} is attributed mainly to the asymmetric stretching of the Si–O–Si/Si–O–Al framework, while the bands in the lower wavenumber region arise from framework bending and deformation vibrations. Such bands are characteristic of zeolitic aluminosilicate structures and are widely used to evaluate changes in their framework chemistry (Eren et al., 2024). The absorption near 1645 cm^{-1} , together with the broad O–H stretching region around 3400 cm^{-1} , is associated with H–O–H bending and O–H stretching of adsorbed water and surface hydroxyl species. Water-related absorption around 1650 cm^{-1} is commonly observed in zeolitic materials because of their ability to retain physically adsorbed water within their porous structure (Uygun et al., 2023). The ZY-1 spectrum represents the characteristic aluminosilicate framework and hydrated surface environment of the starting zeolite-Y.

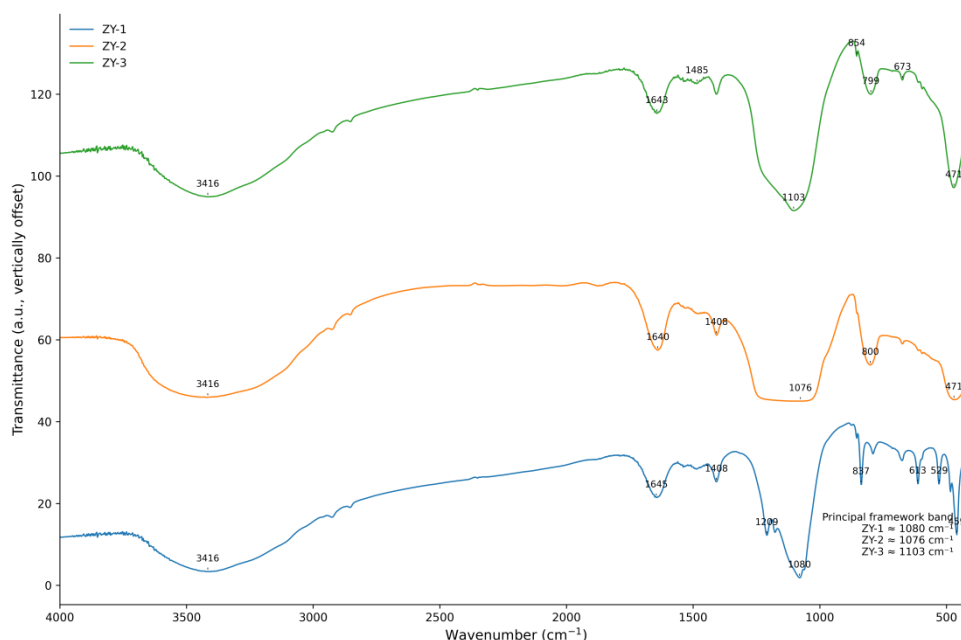


Figure 1: The FTIR spectra for raw Zeolite-Y (ZY-1), pretreated (ZY-2) and Ni-doped (ZY-3)

Following pretreatment, ZY-2 retained the principal framework vibrations observed in ZY-1, indicating preservation of the characteristic aluminosilicate structure. The most notable change was a small displacement of the principal framework-stretching band from approximately 1080.17 cm^{-1} in

ZY-1 to 1076.32 cm^{-1} in ZY-2, corresponding to a shift of approximately 4 cm^{-1} toward lower wavenumbers. This displacement indicates a change in the local chemical environment of the aluminosilicate framework following pretreatment and may reflect the redistribution or modification of the framework-associated and extraframework species. The water-associated band also shifted from approximately 1645.34 cm^{-1} in ZY-1 to 1639.55 cm^{-1} in ZY-2, suggesting a change in the amount or environment of adsorbed water. Importantly, the persistence of the major framework bands indicates that pretreatment modified the chemical environment of the zeolite without eliminating its characteristic aluminosilicate structure. This interpretation is consistent with recent studies showing that chemical treatment of zeolitic materials can alter framework associated species and surface chemistry while retaining identifiable zeolitic vibrational features (Eren et al., 2024).

The incorporation of nickel produced a more pronounced change in the FTIR spectrum, with the principal framework stretching band shifting from 1076.32 cm^{-1} in ZY-2 to 1103.32 cm^{-1} in ZY-3, indicating a change in the local aluminosilicate environment. Nevertheless, the characteristic framework vibrations remained evident, suggesting preservation of the zeolitic structure. The H–O–H bending band was also retained near 1643.41 cm^{-1} , indicating the continued presence of adsorbed water. Similar Ni zeolites have shown that Ni incorporation can modify the local metal support environment without necessarily disrupting the zeolite framework (Zhang et al., 2024).

3.2 Crystalline Phase and Structural Analysis

The XRD pattern of ZY-1 (Figure 2) displayed the characteristic reflections of the faujasite (FAU) type zeolite-Y framework. Prominent reflections were observed at approximately 6.5° , 10.5° , 12.3° , 16.1° , 19.2° , 20.9° , 24.3° , 27.7° and 32.1° , confirming the crystalline nature of the starting material. The characteristic low-angle and high-angle reflections are consistent with the established diffraction pattern of FAU type zeolites and provide a suitable structural reference for assessing the effects of subsequent modification. Manrique et al. (2024) similarly used the persistence and intensity of characteristic zeolite Y reflections to evaluate the structural integrity following chemical modification.

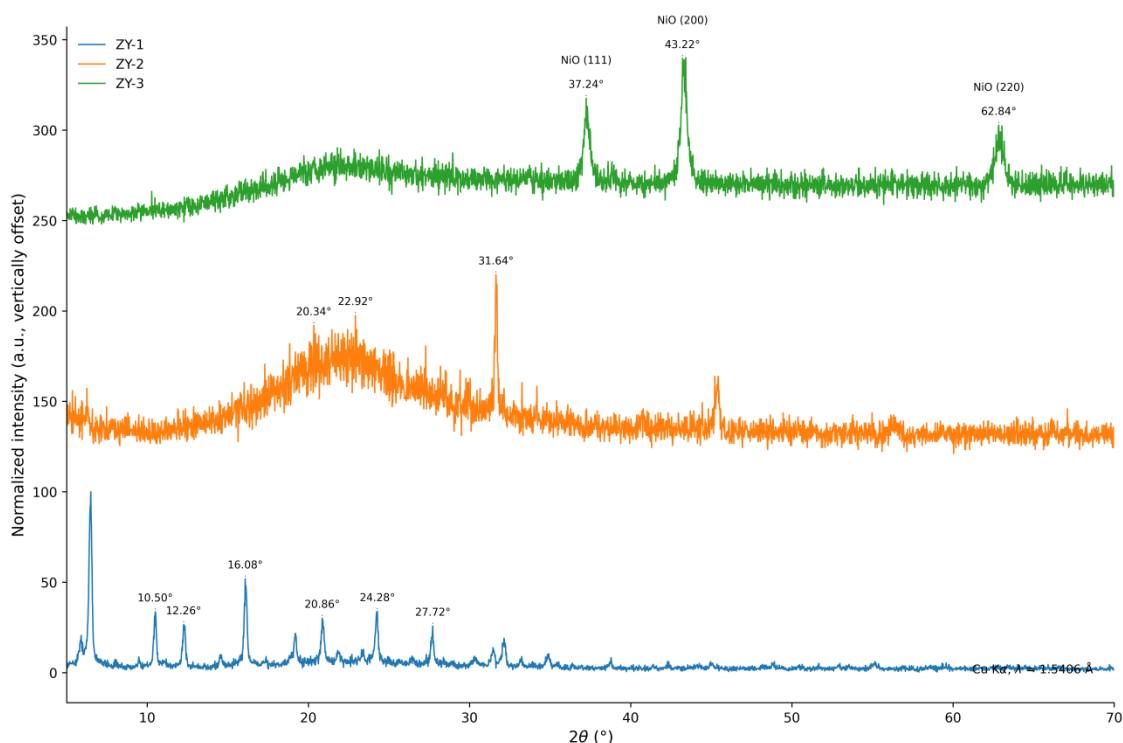


Figure 2: The XRD profiles for raw Zeolite-Y (ZY-1), pretreated (ZY-2) and Ni-doped (ZY-3)

After pretreatment, the sharp, multi-peak reflections observed in ZY-1 were largely replaced in ZY-2 by a broad amorphous hump, with only weak shoulder features near 20.3° and 22.9°. This indicates a substantial loss of long-range crystalline order following alkaline treatment. The changes in diffraction intensity may reflect alterations in framework ordering, extraframework species, or crystallite characteristics resulting from pretreatment. Similar observations have been reported for chemically modified zeolite Y, where changes in diffraction intensity can accompany structural modification, while the principal zeolitic framework remains detectable (Manrique et al., 2024). The observed changes can be more appropriately described as structural modifications with partial loss of the crystalline zeolite-Y framework.

In Ni-doped zeolite-Y, the diffractogram likewise shows a broad amorphous background.. In addition, new reflections appeared at approximately 37.2°, 43.4°, and 62.8°, corresponding to the (111), (200), and (220) planes of crystalline NiO, respectively, which is in agreement with the reported NiO diffraction patterns (Miskan et al., 2024; Zhang et al., 2024). The coexistence of partial zeolite-Y framework and NiO reflections indicates that nickel modification introduced a detectable crystalline NiO phase in the catalyst. However, the presence of NiO peaks does not necessarily mean that all nickel occurs as crystalline NiO, since highly dispersed or poorly crystalline Ni species may remain undetectable by conventional XRD.

3.3 Surface Area and Pore Structure

The textural properties of ZY-1, ZY-2, and ZY-3 (Table 1) showed clear evolution in accessible porosity following pretreatment and Ni incorporation. The BET surface area increased from 181.549 m²/g for ZY-1 to 245.368 m²/g for ZY-2, representing an increase of approximately 35.2%, while the total pore volume increased from 0.096 to 0.133 cm³/g. This indicates that the pretreatment substantially increased the accessible surface area and pore volume of the zeolite. This behaviour may be associated with the generation or opening of secondary pore environments through alkaline modification, thereby improving access to the intrinsic microporous network. However, the accompanying loss of sharp XRD reflections indicates that the textural enhancement may not arise solely from beneficial hierarchical pore development. Partial dissolution of the zeolite framework and associated loss of crystallinity may also have contributed to the increased accessible surface area and pore volume. Thus, the combined BET and XRD results suggest that alkaline pretreatment produced enhanced textural accessibility, but with some degree of structural degradation. This balance is consistent with the need to control alkaline treatment severity, since excessive framework extraction can compromise microporosity and crystallinity (Han et al., 2024).

Table 1: Textural properties of ZY-1, ZY-2, and ZY-3.

Parameter	ZY-1	ZY-2	ZY-3
BET area (m ² /g)	181.549	245.368	278.956
Total pore volume (cm ³ /g)	0.096	0.133	0.163
Micropore volume (cm ³ /g)	0.063	0.092	0.110

The effect of nickel incorporation is also evident from the textural data. Following Ni modification, the BET surface area increased further to 278.956 m²/g, while the total pore volume increased to 0.163 cm³/g. The micropore volume also increased from 0.092 cm³/g in ZY-2 to 0.110 cm³/g in ZY-3, indicating that Ni incorporation did not cause substantial blockage of the zeolite micropore network. The simultaneous increase in the surface area and pore volume suggests that the modified catalyst retained appreciable internal accessibility after metal incorporation. Comparable studies have shown that maintaining accessible micro- and larger-pore networks in metal-modified zeolites is beneficial because the additional transport pathways can improve access to active sites and facilitate the diffusion of relatively bulky hydrocarbon molecules (Manrique et al., 2024; Hu et al., 2024).

The pore size distributions in Figure 3 further showed that the modified zeolites retained predominantly small pores. The DFT profiles showed pore width between 1.8 to 2.8 nm while the BJH adsorption profiles (insert) concentrated within approximately 1.4–2.4 nm. The DFT distributions reflected a progressive change in the pore width profile from ZY 1 through ZY 2 to ZY 3, consistent with modification of the accessible pore environment following pretreatment and Ni incorporation. However, DFT-derived distributions exhibit a pronounced minimum near 2.1 nm, whereas the BJH distributions show a continuous pore-volume contribution through this region. This difference highlights the sensitivity of calculated pore-size distributions to the underlying pore-size analysis methodology and suggests that the DFT minimum should not be interpreted directly as evidence for the absence or blockage of pores at approximately 2.1 nm. Such differences are expected because DFT based pore-size distributions depend on the selected theoretical model and kernel, which can influence the calculated position and intensity of features within the narrow micropore to mesopore range (Gor et al., 2017).

Overall, the BET results indicate a progressive improvement in textural accessibility from ZY-1 to ZY-2 to ZY-3, with the highest values observed for the Ni-modified material. When considered alongside the retained Zeolite-Y reflections observed by XRD, the increased surface area and pore volume are consistent with enhanced pore accessibility and partial development of a hierarchical pore structure, rather than complete framework destruction. This balance between additional porosity and preservation of the zeolitic framework is particularly desirable for hydrocarbon conversion because hierarchical Y zeolites can reduce diffusion limitations while retaining the catalytic functionality of the parent microporous structure (Han et al., 2024; Manrique et al., 2024).

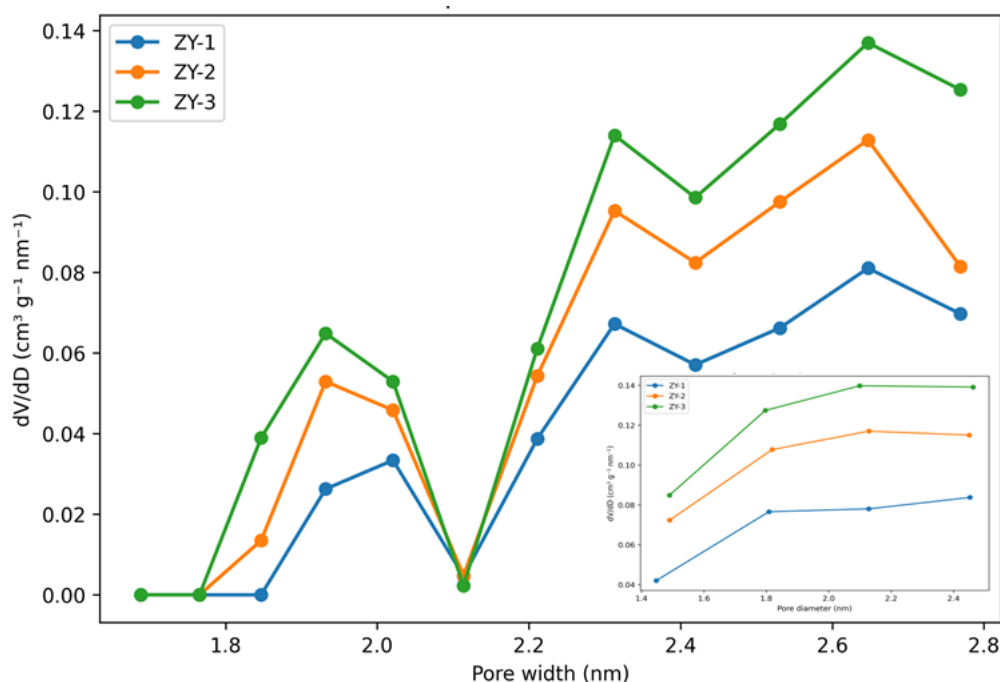


Figure 3: Catalyst pore size distributions using DFT and BJH (insert) methods

3.4 Hydrocarbon Composition and Product Distribution

Thermal treatment of the gas condensate (S1 → G-2) in Figure 4 resulted in only fractional compositional redistribution, with saturates decreasing from 67.47 to 60.80 area% and aromatics increasing from 30.49 to 32.10 area%. The persistence of the C10–C28 n-alkane series and the negligible olefin formation suggest that the thermal step resulted in limited cracking, with redistribution of existing hydrocarbons rather than extensive C–C bond scission. In contrast, the introduction of ZY-2 produced a pronounced change. The n-paraffins decreased from 46.10 to 38.40

area%, while aromatics and naphthenes increased from 32.10 to 40.90 and 3.80 to 6.70 area%, respectively. The high acidity and accessible pore structure of zeolite-Y are known to favour hydrocarbon conversion through acid-catalysed cracking, isomerisation and related secondary reactions (Ding et al., 2021).

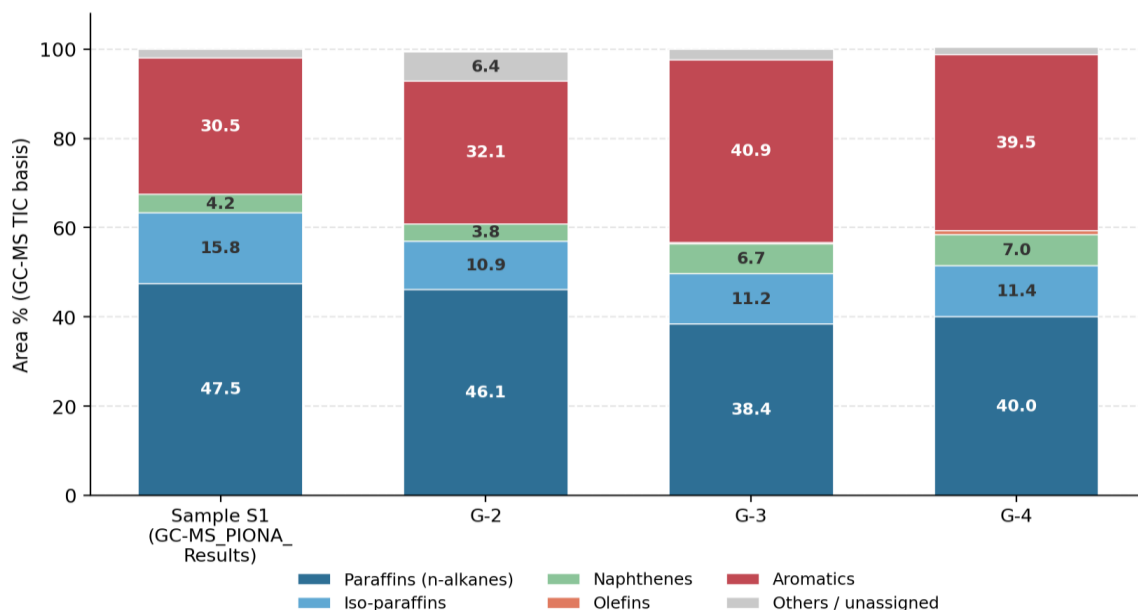


Figure 4: Composition distributions across gas condensate samples

The increase in aromatics following ZY-2 treatment is particularly significant, indicating that paraffin transformation was accompanied by cyclisation and aromatisation rather than being limited to C–C bond cleavage. Metal-modified zeolites have been shown to facilitate dehydrogenation pathways that contribute to aromatic formation (Uslamin et al., 2020), the present ZY-2 results primarily demonstrate the contribution of the acidic zeolite framework to hydrocarbon transformation.

3.5 Liquid Recovery

The apparent liquid recovery (Figure 5) decreased progressively from 25.00% for thermal cracking (G-2) to 18.33% over ZY-2 (G-3) and 14.17% over Ni-ZY-3 (G-4), corresponding to reductions of 26.7 and 43.3%, respectively, relative to thermal cracking. This decline indicates that the catalysts promoted greater transformation of the feed beyond the condensable-liquid fraction, potentially through secondary cracking to lighter non-condensable hydrocarbons. For the Ni-containing catalyst, additional reactions such as dehydrogenation and aromatization may also have contributed to the observed reduction in liquid recovery.

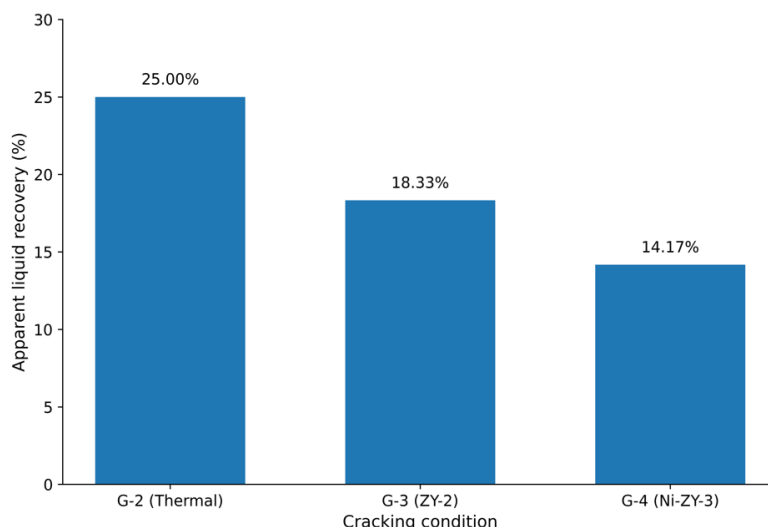


Figure 5: Apparent liquid recovery of cracking products

The lower liquid recovery is consistent with the PIONA and carbon-number distribution results, which show increased hydrocarbon transformation over the catalysts, evidenced by the reduction in n-paraffins and enrichment of aromatics and naphthenes, particularly after ZY-2 treatment. These compositional changes indicate that catalytic cracking promoted secondary reactions beyond simple C–C bond scission.

4.0 CONCLUSION

Characterization of the catalysts showed that alkaline pretreatment and nickel incorporation altered both the textural and structural properties of zeolite-Y. BET analysis indicated a progressive increase in surface area and pore volume from ZY-1 (181.5 m²/g) to ZY-2 (245.4 m²/g) and ZY-3 (279.0 m²/g); however, XRD showed that this gain was accompanied by substantial loss of long-range crystalline order in ZY-2 and ZY-3, indicating that the increased porosity partly reflects framework degradation rather than purely beneficial hierarchical pore development. XRD further confirmed the formation of crystalline NiO in ZY-3, while FTIR confirmed retention of the principal aluminosilicate framework vibrations, with modest band shifts following pretreatment and nickel incorporation.

The study showed that zeolite-Y pretreatment significantly influenced gas condensate conversion, with ZY-2 promoting greater hydrocarbon redistribution toward aromatic and naphthenic products. The apparent liquid recovery also decreased from 25.00% for thermal cracking to 18.33% with ZY-2 and 14.17% with Ni-ZY-3, indicating greater transformation of the feed beyond the recovered condensable-liquid fraction. Ni incorporation produced a comparatively subtle change, including a slight increase in the olefin and naphthene fractions. Overall, the results confirmed that both zeolite modification and Ni incorporation influenced the hydrocarbon distribution during gas condensate cracking.

Acknowledgement

The authors gratefully acknowledge the support provided by the Petroleum Technology Development Fund (PTDF) Chair in Chemical Engineering at Ahmadu Bello University, Zaria.

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