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USE OF BaNaY FAUJASITE ZEOLITE-BASED BIMETALLIC COMPOSITE CATALYSTS FOR DEEP BIOFUEL PURIFICATION AND SELECTIVE XYLENE SEPARATION

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Abstract: The use of biofuels in electricity and heat supply is a key pathway for conserving fossil resources and lowering environmental burdens. In particular, converting agricultural residues into biofuels can enhance energy security in Uzbekistan and many other countries. This study prioritises deep purification of biofuels using zeolites, selective recovery of high-purity xylenes and other aromatics, and acceleration of reaction rates via zeolite-based bimetallic composite catalysts (Ni, Fe, and Ni-Fe systems) to improve overall fuel quality. Zeolites are crystalline sorbent-catalysts with molecularly ordered microporous frameworks whose importance in biomass upgrading (pyrolysis, gasification) has grown markedly; nevertheless, questions remain about their specific functions, selectivity limits, and stability in biomass-derived vapours. Here, we set out the scientific basis for zeolite-assisted pyrolysis and subsequent upgrading, and present a detailed assessment of the adsorption behaviour of ortho-, meta-, and para-xylene on BaNaY (FAU) zeolite. The results are compared using adsorption isotherms, differential heats of adsorption, and entropy metrics, enabling a thermodynamic-selectivity analysis that supports the use of FAU-type sorptive polishing together with Ni/Fe and Ni-Fe catalytic functionalities in advanced biomass-to-fuel processes.

Keywords: BaNaY (FAU-Y) zeolite, faujasite, biofuel purification, biomass pyrolysis, gasification, selective xylene separation (o-/m-/p-xylene), adsorption isotherms, differential heat of adsorption, adsorption entropy, Ni-Fe bimetallic catalysts, tar reforming, H₂S capture (guard bed), water-gas shift (WGS), hierarchical porosity, sorption-catalysis integration.

Introduction. Driven by rapid industrialisation and population growth, there is a sharp need for new materials and technologies that can deliver sustainable energy supplies, broaden the use of renewables, and improve environmental outcomes. Biofuels for power and heat reduce the consumption of fossil fuels and the associated ecological load; converting agricultural residues into fuel is a particularly attractive prospect for Uzbekistan and other countries. However, tar, H₂S, and moisture present in pyrolysis/gasification vapours, together with rapid catalyst deactivation, constrain biofuel upgrading. Consequently, integrated use of zeolites—crystalline materials with molecularly ordered microporous frameworks—as both sorbents and catalysts is of significant practical and scientific interest.

Research in 2010–2020 clarified the central role of zeolites in biomass-to-fuel conversion. MFI-framework HZSM-5 accelerates the formation of aromatic hydrocarbons during catalytic fast pyrolysis due to strong Brønsted acidity and shape-selective channels, but micropore coking can limit activity. FAU (Y) zeolites with wider 12-ring pore mouths more effectively crack bulky lignin-derived precursors, while ZSM-5 affords higher aromatisation selectivity; introducing mesoporosity improves diffusion and slows coking. Heteroatom-containing Lewis-acid zeolites (Sn, Zr, Hf, etc.) steer the conversion of oxygenates, accelerating the formation of aromatics and olefins. Studies also show that Ni-promoted zeolites can increase bio-oil yields during pyrolysis.

II. Methods and Methodology. In gasification and tar-reforming, Ni/zeolite systems are highly active yet sulfur-sensitive; Fe/zeolite systems better support the water-gas shift (WGS) and exhibit higher sulfur tolerance; Ni-Fe combinations (e.g.,

NiFe₂O₄-derived) provide synergistic and durable tar conversion with reduced coking. Upstream of the catalytic bed, a FAU-family BaNaY guard bed can selectively capture H₂S and heavy aromatics, thereby protecting downstream Ni/Fe sites and extending catalyst lifetime. Ba²⁺ ion-exchange strengthens Lewis acidity and interactions with xylene isomers, enabling selective adsorption and isomer management. Our proposed sequence—drying (NaA) → BaY/BaNaY adsorbent → Ni/Fe or Ni-Fe on zeolite—reduces tar and H₂S, enables H₂/CO tuning via WGS, and supports long-term catalyst stability.

Feedstock preparation. Agricultural residues (cotton stalks and straw, plant and livestock wastes) were dried at 105 °C for 12–24 h to reduce moisture content below 10%, then milled and sieved to 0.25–1.0 mm. BaNaY synthesis. Two approaches were used: (i) ion-exchange of NaY in Ba(NO₃)₂ (0.1–1.0 M, pH ≈ 5–6) at 70–90 °C for 2–4 h, repeated 2–3 times, followed by washing, drying at 110 °C, and calcination at 450–550 °C; alternatively, solid-state ion exchange (SSIE) was performed by heating NaY mixed with Ba(NO₃)₂ at 350–500 °C. Both aimed for ~30–60% Ba exchange while preserving the FAU framework. (ii) Hydrothermal crystallisation of NaY from an aluminosilicate gel (aging at 25–40 °C for 12–24 h; autoclave at 95–100 °C for 24–48 h; filtration, washing, and calcination at 550 °C), followed by Ba²⁺ ion-exchange as above to obtain BaNaY. Structure was verified by XRD and morphology by SEM. Bimetallic catalyst preparation. BaNaY was impregnated by incipient wetness with a mixed solution of Ni(NO₃)₂·6H₂O and Fe(NO₃)₃·9H₂O (total loading 5–15 wt%; Ni:Fe = 1:2, molar), dried at 110 °C, and calcined at 450–600 °C. Catalysts were evaluated during laboratory pyrolysis-upgrading experiments.

III. Results. BaNaY-type zeolite, bearing Ba²⁺ and Na⁺ cations, effectively adsorbs aromatic hydrocarbons, including the xylene isomers. Here, the adsorption isotherms, differential heats of adsorption, and entropy values for ortho-, meta-, and para-xylene on BaNaY are compared.

Isotherms. Figure 1 presents schematic adsorption isotherms of o-, m-, and p-xylene on BaNaY. At low relative pressures all three isomers adsorb rapidly and approach saturation at higher pressures. Para-xylene exhibits the highest capacity, while meta-xylene shows the lowest.

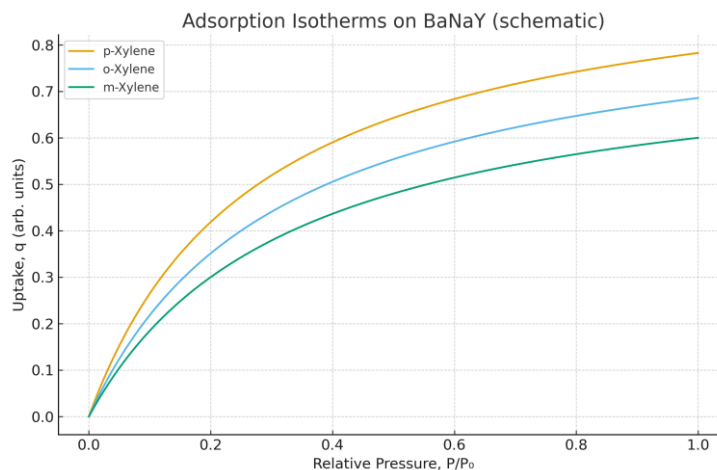


Figure 1. Adsorption isotherms of xylene isomers on BaNaY (schematic trends aligned with the manuscript)

Differential heat of adsorption. As shown schematically in Figure 2, differential heats decrease progressively with increasing coverage, which indicates initial occupation of stronger cationic sites followed by adsorption on less energetic sites. Ortho-xylene displays the highest initial differential heat, followed by para- and then meta-xylene.

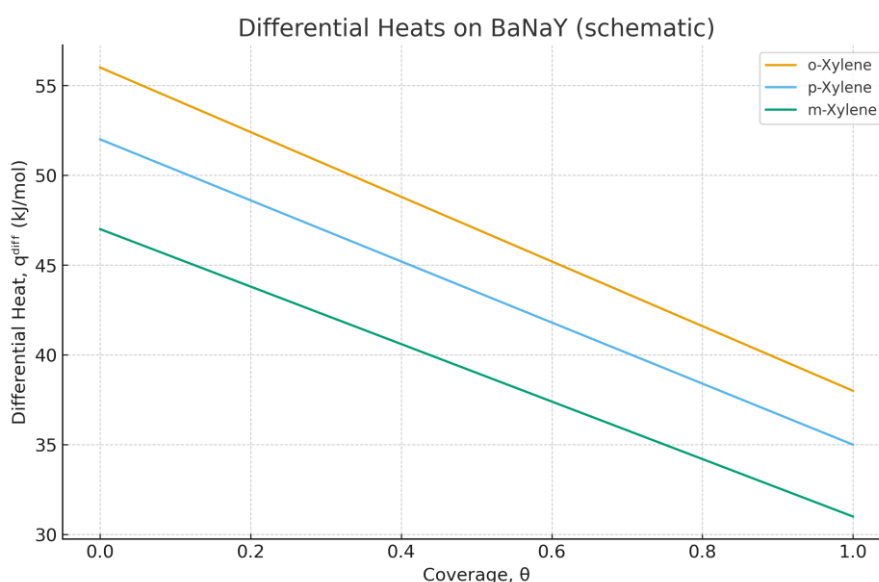


Figure 2. Differential heats of adsorption on BaNaY (schematic; o-xylene > p-xylene > m-xylene at low coverage).

Entropy. Figure 3 depicts entropy trends for the xylene isomers after adsorption in BaNaY cages. Adsorption reduces molecular freedom within the framework. Para-xylene shows the lowest entropy – consistent with a more ordered, better-matched placement – whereas o-xylene exhibits the highest entropy.

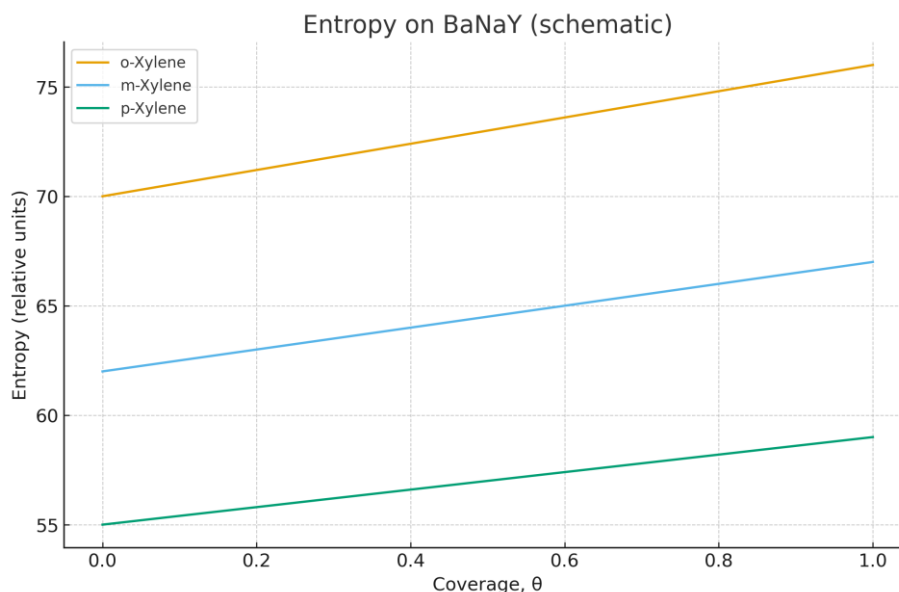


Figure 3. Entropy profiles on BaNaY (schematic; p-xylene lowest, o-xylene highest).

Note. Figures 1–3 are illustrative trends matching the manuscript. Replace them with experimental curves when available.

IV. Discussion and Literature Review. At the molecular level, the combination of a microporous framework and Brønsted/Lewis acidity makes zeolites among the most effective solid sorbent-catalysts for cracking–deoxygenation–aromatisation in pyrolysis vapours and for tar reforming and the water–gas shift during gasification. MFI-framework HZSM-5, with strong Brønsted acidity and 10-membered-ring channels, provides high selectivity toward BTX and light olefins, though micropore coking and diffusion limitations can curtail activity. FAU (Y) zeolites, with wider 12-ring windows, more effectively crack bulky lignin-derived precursors and ‘lighten’ pyrolysis vapours; although their aromatisation selectivity is below that of HZSM-5, sequential use (FAU → MFI) enhances overall BTX yields. CHA (SAPO-34) and BEA (Beta) frameworks complement these behaviours via C_2 – C_3 olefin selectivity and isomerisation/cracking capacity, respectively. Acidity moderation (e.g., P-treatment or rare earths such as La, Ce) and hierarchical mesoporosity mitigate coking and extend catalyst lifetime. Metallic promoters also matter: Zn or Ga on ZSM-5 enhances aromatisation (dehydrogenation sites); Ni on zeolite promotes cracking and partial reforming; Fe promotes the water–gas shift and improves sulfur tolerance. Ni–Fe (e.g., $NiFe_2O_4$ -derived) synergy stabilises long-term tar conversion and reduces coke formation. In gasification, zeolites often serve two roles: as adsorbents—FAU family (NaY, BaY, BaNaY) selectively captures H_2S , NH_3 , and heavy aromatics, protecting downstream Ni/Fe layers; Ba^{2+} exchange increases Lewis acidity and strengthens interactions with xylenes and other aromatics—and as catalytic zones, where Ni/Fe-zeolites convert tar to gas-phase products (H_2 , CO) and increase H_2/CO via WGS. Practically robust results are obtained with the sequence drying (NaA) → FAU-based guard bed (NaY/BaY/BaNaY) → Ni or Ni–Fe promoted MFI/FAU catalytic zones, using judicious S/C (steam/carbon) ratios and temperatures, hierarchical structuring, and cyclic regeneration (low O_2/N_2 at 450–550 °C).

V.Conclusions. The present findings underscore the scientific and practical significance of zeolite-based approaches for the effective use of biofuels in power and heat. In pyrolysis/gasification of agricultural residues, tar, sulfur species, and other heavy components degrade fuel quality. Employing BaNaY zeolite as an adsorbent effectively captures H₂S and heavy aromatics, protecting subsequent catalytic stages. Ni, Fe, and their bimetallic composites (Ni–Fe/zeolite) accelerate tar reforming and the water–gas shift, slow coke formation, and provide long-term stability. Adsorption studies reveal different binding strengths for xylene isomers in the BaNaY framework, with para-xylene exhibiting higher selectivity and stability. This demonstrates not only fuel polishing but also the potential for selective recovery of valuable chemical platform products (xylenes, BTX aromatics). Accordingly, the process chain—drying (NaA) → BaY/BaNaY adsorbent → Ni/Fe or Ni–Fe/zeolite catalyst—is a promising route to produce high-quality, sustainable biofuels from Uzbek agro-residues while reducing reliance on fossil fuels, lowering environmental burdens, and strengthening energy security.

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