# Natural Zeolites as Green Catalysts in Biomass Pyrolysis: Trends, Challenges and Perspectives from a Bibliometric Review

# Zeolitas naturales como catalizadores verdes en la pirólisis de biomasa: tendencias, desafíos y perspectivas desde una revisión bibliométrica

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SOUTH SUSTAINABILITY

**Cite as:** Venegas-Vásconez, D. *et al*. (2026). «Natural Zeolites as Green Catalysts in Biomass Pyrolysis: Trends, Challenges and Perspectives from a Bibliometric Review». *South Sustainability*, 7(2), e153. DOI: 10.21142/SS-0702-2026-e153

Received: 13/3/2026
Peer reviewed
Accepted: 17/6/2026



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Associate editor:
Alexander Perez Segovia

## ABSTRACT

Biomass pyrolysis has emerged as a key thermochemical pathway for generating renewable fuels and chemicals. However, the high oxygen content and inherent instability of bio-oil remain major challenges, hindering its commercial application. Zeolite catalysts are widely employed to improve product selectivity and enhance bio-oil stability, yet the high cost and energy-intensive synthesis of synthetic zeolites constrain their large-scale use. In this review, we provide a comprehensive analysis of natural zeolites as cost-effective and sustainable alternatives for catalytic biomass pyrolysis. A bibliometric assessment of 230 publications indexed in the Scopus database (2000-2025) was conducted to elucidate research trends, identify highly cited contributions, and highlight emerging knowledge gaps. The thematic analysis was guided by research questions focused on comparative catalytic performance, modification strategies and sustainability considerations. The results indicate that natural zeolites, such as clinoptilolite, can enhance liquid yields and improve fuel properties; however, their selectivity toward aromatics is generally lower than that of synthetic zeolites, such as HZSM-5. Modification approaches —including acid treatment, ion exchange and metal impregnation— significantly increase the acidity and porosity of natural zeolites, thereby promoting deoxygenation and hydrocarbon formation. Beyond catalytic performance, natural zeolites offer distinct sustainability benefits, including lower cost, reduced energy demand and a smaller carbon footprint. Nevertheless, challenges remain, related to structural heterogeneity, catalyst deactivation and limited pilot-scale validation. This review underscores the promising yet underexplored potential of natural zeolites for scalable, environmentally sustainable biomass valorization, while outlining key research directions to enhance catalytic efficiency and industrial applicability.

***Keywords:*** *Natural zeolites, pyrolysis, lignocellulosic biomass, catalyst*

## RESUMEN

La pirólisis de biomasa se ha consolidado como una ruta termoquímica clave para la generación de combustibles y productos químicos renovables. No obstante, el elevado contenido de oxígeno y la inestabilidad inherente del bioaceite continúan siendo desafíos críticos que limitan su aplicación comercial. Los catalizadores zeolíticos son ampliamente empleados para mejorar la selectividad de productos y aumentar la estabilidad del bioaceite; sin embargo, el alto costo y la síntesis energéticamente intensiva de las zeolitas sintéticas restringen su implementación a gran escala. Esta revisión presenta un análisis integral de las zeolitas naturales como alternativas rentables y sostenibles para la pirólisis catalítica de biomasa. Se llevó a cabo una evaluación bibliométrica de 213 publicaciones indexadas en la base de datos Scopus (2000-2025), con el objetivo de dilucidar las tendencias de investigación, identificar las contribuciones más citadas y destacar vacíos emergentes de conocimiento. El análisis temático fue orientado por preguntas de investigación centradas en el desempeño catalítico comparativo, las estrategias de modificación y las consideraciones de sostenibilidad. Los resultados indican que las zeolitas naturales, como la clinoptilolita, pueden incrementar los rendimientos líquidos y mejorar las propiedades del combustible; sin embargo, su selectividad hacia compuestos aromáticos es, en general, inferior a la de zeolitas sintéticas como HZSM-5. Las estrategias de modificación —incluyendo tratamiento ácido, intercambio iónico e impregnación metálica— incrementan significativamente la acidez y la porosidad de las zeolitas naturales, lo que promueve los procesos de desoxigenación y la formación de hidrocarburos. Más allá del desempeño catalítico, las zeolitas naturales ofrecen ventajas claras en términos de sostenibilidad, tales como menor costo, reducción en la demanda energética y una huella de carbono más baja. No obstante, persisten desafíos asociados con la heterogeneidad estructural, la desactivación catalítica y la limitada validación a escala piloto. Esta revisión destaca el potencial prometedor —aunque aún insuficientemente explorado— de las zeolitas naturales para la valorización escalable y ambientalmente sostenible de la biomasa, al tiempo que define líneas prioritarias de investigación orientadas a mejorar la eficiencia catalítica y la aplicabilidad industrial.

***Palabras clave:*** *zeolitas naturales, pirólisis, biomasa lignocelulósica, catalizador*

## 1. Introduction

Petroleum remains the primary global energy source; however, its medium-term availability is limited, rendering long-term supply uncertain (Mach *et al.*, 2016). This concern is compounded by climate change and environmental pollution from elevated levels of atmospheric $CO_2$ and particulate matter (Energy Institute, 2024). In this context, there is a substantial focus on research and policy efforts associated with renewable feedstocks as sustainable alternatives for producing fuels and high-value chemicals (Mignogna *et al.*, 2024; Chu and Majumdar, 2012). Technological advances, novel feedstocks, and evolving oil-pricing policies are projected to steadily increase global biofuel demand across multiple sectors (Bauen *et al.*, 2019; Balat and Balat, 2009). Current efforts emphasize large-scale bioproduction systems utilizing diverse sources, including industrial waste via biotechnology (Atabani *et al.*, 2012), vegetable oils from oilseed crops (Isikgor and Becer, 2015), lignocellulosic biomass and residues, municipal solid waste, forestry residues, dedicated energy crops and animal manure (Amulen *et al.*, 2022; Okafor *et al.*, 2022).

Widely recognized as a renewable energy resource, biomass includes feedstocks derived from organic matter of biological origin (Ramos Maldonado *et al.*, 2024). Its thermal decomposition involves bond cleavage that releases energy, enabling the generation of high-value products (Järvinen and Paulomäki, 2025). Although biomass can be converted into biofuels, several production challenges persist, including:

- The lower heating value (LHV) of biomass decreases markedly with moisture content; for instance, at 30% moisture, LHV may fall to approximately 11.6 MJ/kg, whereas dry coal can reach values of up to 27.9 MJ/kg (Orejuela-Escobar *et al.*, 2024).
- Structural composition strongly influences energy content: lignin exhibits higher calorific values (23-25 MJ/kg) compared to polysaccharides (~18 MJ/kg) (Esteves *et al.*, 2023).
- Bio-oil obtained from food waste pyrolysis often displays high viscosity (~136 cSt) and considerable water content (~4.6%), despite a relatively high HHV of 28.2 MJ/kg (Makkawi *et al.*, 2022).
- Torrefaction can enhance biomass LHV to approximately 20.4 MJ/kg, while the resulting char typically reaches 28-33 MJ/kg (Sugitha *et al.*, 2024).
- Recent reviews highlight ongoing challenges in bio-oil upgrading, particularly its high oxygen content and thermal instability, which require catalytic stabilization (Jerzak *et al.*, 2024).

Pyrolysis is a thermal decomposition process conducted in the absence of oxygen, typically at 250-900 °C (Osman *et al.*, 2023). It produces condensable liquids (bio-oil), solid char and non-condensable gases (Al-Rumaihi *et al.*, 2022; Venegas-Vásconez *et al.*, 2023). Its versatility with diverse feedstocks —agricultural residues, forestry waste, energy crops and municipal solid waste— makes pyrolysis highly adaptable and logistically advantageous (Jahirul *et al.*, 2012; Venegas-Vásconez, 2025).

Techno-economic assessments confirm that pyrolysis products can compete with conventional fuels without subsidies, affirming their economic viability (Badger *et al.*, 2010). Additionally, carbon-rich biochar contributes environmental benefits through carbon sequestration and climate mitigation (Enebe *et al.*, 2025). Catalysts enhance pyrolysis product properties by promoting selective transformations, altering kinetics and optimizing distributions (Lin *et al.*, 2021; Sun *et al.*, 2025). Catalytic pyrolysis accelerates reaction rates, lowers operating temperatures, upgrades bio-oil quality and increases overall efficiency (Zhang *et al.*, 2025; Rajak *et al.*, 2025). Key catalyst properties —surface area, acidity, pore size and volume— determine performance (Luna-Murillo *et al.*, 2021), while reducing energy demand by enabling lower reaction temperatures (Kabakcı and Hacıbektaşoğlu, 2017).

Natural zeolites, such as clinoptilolite, are far more affordable and abundant than their synthetic counterparts (e.g., ZSM-5 and Zeolite Y), with costs of ~USD 0.24/kg versus USD 120-180/kg for synthetics (Alejandro-Martín *et al.*, 2019). Their extraction and processing impose a lower environmental footprint and energy demand than the resource-intensive synthesis of artificial zeolites, yielding simpler protocols and reduced capital/operating expenses (Wahono *et al.*, 2020). Although native natural zeolites show modest activity, modifications via ion exchange, metal impregnation or acid activation significantly enhance Brønsted acidity and deoxygenation performance (Grifasi *et al.*, 2024; Velarde *et al.*, 2023). These characteristics support sustainable, cost-effective, scalable, and eco-friendly catalytic systems, particularly in decentralized or resource-constrained settings (Hrenović and Rajić, 2025; Pavlović and Rajić, 2024).

Given the pressing need for low-cost, sustainable catalysts in biomass valorization, a dedicated review of natural zeolites in pyrolysis is essential, in order to delineate the transition from laboratory-scale trials to industrial implementation (Valencia-Huamán *et al*., 2024; Muhammad *et al*., 2024). While synthetic zeolites like ZSM-5 dominate bio-oil upgrading research due to their defined pores and tunable acidity, their high cost and energy-intensive production constrain scalability (Nishu *et al*., 2020). The literature on natural zeolites' systematic application, modification strategies and techno-economic aspects remains fragmented (Chaihad *et al*., 2022). Consolidation of knowledge concerning their catalytic behavior, limitations and prospects will strengthen their viability as alternatives to synthetics and guide sustainable biofuel pathways (Abu El-Rub *et al*., 2023). Despite growing interest in natural zeolites as cost-effective, environmentally friendly catalysts for biomass pyrolysis, a clear consensus on the current research trajectory remains lacking. Specifically, it remains unclear whether or not the field is shifting progressively from fundamental characterization studies toward more advanced investigations focused on catalyst stability, deactivation mechanisms and product selectivity. While early studies predominantly emphasized the structural, textural and compositional characterization of natural zeolites, recent work has suggested a growing interest in the optimization of catalytic performance, particularly through enhanced selectivity toward high-value chemicals and improved catalyst reusability (Sun *et al*., 2025). However, this transition has not yet been systematically established across the literature, indicating a critical gap that this review aims to address.

Furthermore, although natural zeolites are derived inherently from abundant geological resources, their «green» character extends beyond their origin. In contrast to synthetic zeolites, which often require the use of toxic organic structure-directing agents (OSDAs) during synthesis, natural zeolites eliminate or significantly reduce this environmental burden. Additionally, their potential for regeneration and reuse under pyrolytic conditions enhances their sustainability profile, making them attractive candidates for circular and low-impact catalytic processes (Corma, 1997; Mishra *et al*., 2022). These aspects position natural zeolites as promising materials, not only from an economic standpoint but also within the broader context of green chemistry and sustainable catalysis.

This bibliometric review identifies the tipping points in natural zeolite modification by mapping publication trends, influential authors, key works and emerging research fronts in natural zeolite applications for biomass pyrolysis. It offers a rigorous framework for assessing major contributions and underscores sustainability advantages, circular economy alignment and responsible catalysis, thereby facilitating wider adoption in biomass valorization.

## 2. Materials and methods

In our study, we adopted a mixed-methods approach combining quantitative bibliometric analysis with qualitative thematic synthesis within the framework of a systematic literature review. The research design was structured in two main phases. First, a quantitative bibliometric analysis was conducted to map the intellectual structure, publication trends and research dynamics in the field, including co-occurrence of keywords, citation patterns and thematic evolution. Then, a qualitative thematic synthesis was performed to interpret and contextualize the identified trends, enabling a deeper understanding of research directions, knowledge gaps and emerging topics. This integrative approach enhanced the comprehensiveness and reliability of the findings, by combining the respective strengths of both quantitative and qualitative methodologies.

### *2.1. Comprehensive literature review*

The bibliometric review followed the methodology outlined by Pakdel and Erol (2025), which facilitates a systematic, quantitative assessment of publications within a specified research domain (Venegas-Vásconez *et al*., 2025). The process included: (i) a comprehensive search in the Scopus database using the following search string: TITLE-ABS-KEY (natural zeolites, catalyst, pyrolysis). The search was conducted on May 10, 2026; (ii) screening of retrieved documents against predefined inclusion and exclusion criteria to ensure relevance, quality and consistency; (iii) extraction of key bibliometric indicators, including publication year, journal, country of origin, citation counts and authorship patterns (Paul *et al*., 2024) (Venegas-Vásconez and Tirado-Lozada, 2026); (iv) network analyses of co-authorship, keyword co-occurrence, and citation linkages to reveal trends and influential contributors; and (v) synthesis of findings to highlight knowledge gaps and emerging research directions. Adherence to the Pakdel and Erol (2025) framework promotes transparency, reproducibility and a robust mapping of the scientific landscape (Venegas-Vásconez, 2026). Keyword co-occurrence analysis was conducted using VOSviewer (version 1.6.20), which is widely recognized for generating semantic relationship maps from bibliographic metadata. Graphs and visualizations were produced in OriginPro (version 2025) to ensure clarity, accuracy and reproducibility.

### *2.2. Thematic analysis guided by research questions*

Based on the selected studies, a thematic analysis was conducted to address the following research questions, with the aim of synthesizing key findings, identifying prevailing trends and highlighting research gaps and future opportunities in zeolite-catalyzed biomass pyrolysis:

- **RQ1:** How do the catalytic performances of natural and synthetic zeolites compare in biomass pyrolysis with respect to bio-oil yield, selectivity and product quality?
- **RQ2:** In what ways do modification strategies (e.g., acid treatment, ion exchange, metal impregnation) affect the physicochemical properties and catalytic efficiency of natural zeolites?
- **RQ3:** To what extent do natural zeolites provide sustainability advantages compared to synthetic zeolites, with respect to economic, environmental and energy-related factors?
- **RQ4:** What are the main research trends, thematic focuses and knowledge gaps identified in the ten most highly cited open-access articles published recently on natural zeolites in biomass pyrolysis? In this context, given the significant increase in scientific publications in recent years the PRISMA (Preferred Reporting Items for Systematic Reviews and Meta-Analyses) methodology (Pakdel and Erol, 2025) was applied, in order to ensure a systematic and rigorous selection process (Figure 1). Specifically, the analysis focused on identifying the most highly cited articles published from 2022 onwards, with the aim of capturing the most up-to-date and relevant contributions to the research topic. The search was exclusively limited to scientific articles, as these documents present research findings in a more detailed, structured and rigorous manner compared to other types of publications. Additionally, only open-access articles were considered, in order to ensure that the selected information was freely available to the global community, promoting accessibility, transparency and reproducibility, regardless of geographic location. The selection criteria further included publications written in English and indexed within the relevant subject area (Navas-Ulloa *et al*., 2026). This approach enabled the identification of the most influential and current contributions, strengthening the robustness and reliability of the literature review process. The following search string was used on May 10, 2026, to select the most relevant articles based on the specified selection criteria: TITLE-ABS-KEY (natural zeolites, catalyst, pyrolysis) AND PUBYEAR > 2021 AND PUBYEAR < 2027 AND (LIMIT-TO (DOCTYPE, "ar")) AND (LIMIT-TO (OA, "all")).

This approach enabled a structured synthesis of the literature, integrating both experimental findings and theoretical perspectives. It offers a comprehensive overview of the current state-of-the-research on zeolite-catalyzed biomass pyrolysis, underlining the potential of zeolites for sustainable biofuel production and constituting a guide to future investigative directions.

## 3. Results

### *3.1. Comprehensive literature review*

A bibliometric analysis was conducted using the Scopus database to examine the research landscape regarding the application of natural zeolites as catalysts in pyrolysis processes. Using the keywords «natural zeolite», «catalyst», and «pyrolysis», a total of 230 scientific documents were retrieved. The search was performed on May 10, 2026. This dataset provides a comprehensive overview of the evolution, research trends and scientific contributions in this field, forming the basis for identifying knowledge gaps and guiding future research directions.

#### *3.1.1. Publications per year*

The bibliometric analysis of Scopus-indexed publications between 2000 and 2025 (Figure 2) reveals a clear upward trend in research on the use of zeolites in biomass pyrolysis. Although studies in this field can be traced back to 1979 (Raskina, 1979), our analysis was limited to the period from 2000 onwards, as earlier publications were sporadic in nature and exhibited minimal continuity. From 2000 to 2009, annual scientific output remained low, with fewer than three publications per year, reflecting an early exploratory stage. Between 2010 and 2015, the number of studies increased moderately, ranging from three to nine publications annually, indicating growing academic interest aligned with emerging renewable energy strategies.

Notably, from 2016 onwards a marked and sustained acceleration was observed, with publication output increasing sharply and reaching a peak of over 25 papers per year in 2023 and 2024. While the absolute volume of publications (n = 230) has remained relatively specialized

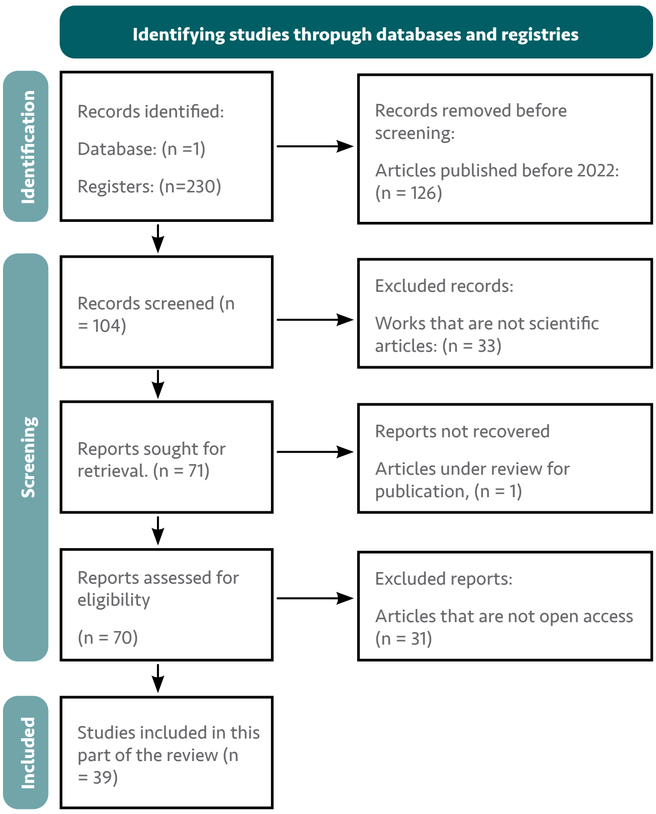


**Figure 1.** Flowchart of the PRISMA methodology illustrating the identification, screening, eligibility and inclusion stages of the selected studies

compared to broader catalytic research fields, the nearly 800% increase in annual output between 2015 and 2024 suggests a significant shift within the scientific community toward low-cost, naturally occurring mineral catalysts. This inflection point around 2016 can plausibly be associated with multiple converging drivers, including the global adoption of circular economy frameworks, increased policy emphasis on sustainable resource use, and alignment of research agendas with the United Nations 2030 Agenda for Sustainable Development (Cosentino *et al*., 2024). Additionally, economic factors, such as the relatively high cost and synthesis complexity of conventional zeolites (e.g., ZSM-5), may have further spurred interest in natural zeolites as viable, scalable catalytic alternatives. The lower number of publications observed for 2025 is likely due to incomplete indexing at the time of data collection. However, preliminary data for 2026, including records indexed up to May 10, indicate that the current year is following a trajectory comparable to the most productive recent years, reinforcing the sustained momentum of this research field. Overall, this strong upward trend not only reflects increasing scientific attention but also underscores the strategic relevance of natural zeolites in advancing sustainable catalytic processes within the circular economy and in accordance with global sustainability goals.

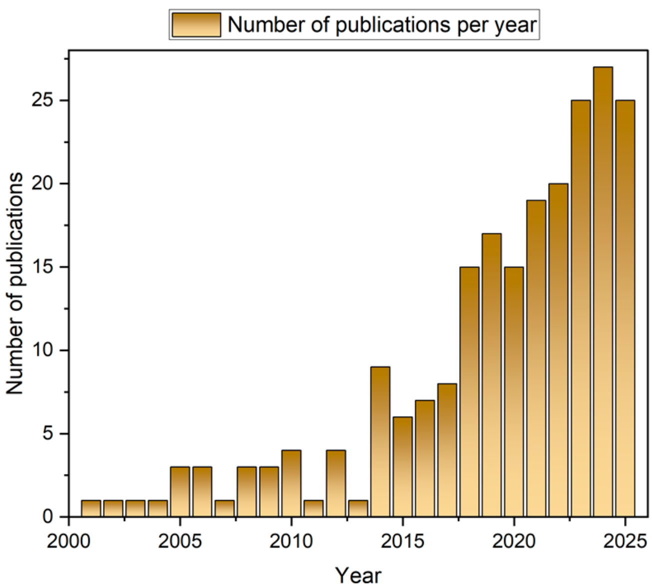


**Figure 2.** Publications per year in Scopus database

### 3.1.2. Correlation keywords

The keyword co-occurrence analysis generated using VOSviewer (Figure 3) revealed a well-structured intellectual landscape organized into five main thematic clusters, reflecting the multidimensional development of research on zeolites in biomass pyrolysis. The central green cluster represents the field's core, dominated by terms such as pyrolysis, zeolites, catalysts and biomass, highlighting the primary focus on catalytic pyrolysis processes and the production of value-added compounds, including bio-oil, aromatic hydrocarbons and biofuels. This cluster also emphasizes catalyst performance, product distribution and feedstock variability, indicating a mature and application-oriented research domain. The red cluster is associated with plastic waste and polymer-related studies, including terms such as polyethylene, polypropylene and plastic recycling, suggesting a parallel research stream focused on the catalytic conversion of synthetic waste materials, often integrated with biomass through co-pyrolysis approaches. The blue cluster reflects the fundamental and physicochemical characterization of catalysts, incorporating keywords such as reaction kinetics, activation energy and thermogravimetry, indicating strong efforts toward understanding reaction mechanisms and catalytic behavior at a fundamental level. Meanwhile, the yellow cluster is oriented toward process optimization and economic considerations, with terms such as low-costs, co-pyrolysis, mineral oils and coke, highlighting increasing attention to process efficiency, scalability and industrial feasibility. Finally, the purple cluster, although smaller, represents emerging or niche topics related to catalytic upgrading and advanced processes, suggesting ongoing diversification and specialization within the field. Overall, the network structure demonstrates a clear evolution from fundamental catalytic studies toward integrated, application-driven and sustainability-oriented research, with strong interconnections among clusters indicating a high degree of interdisciplinarity and consolidation in the field.

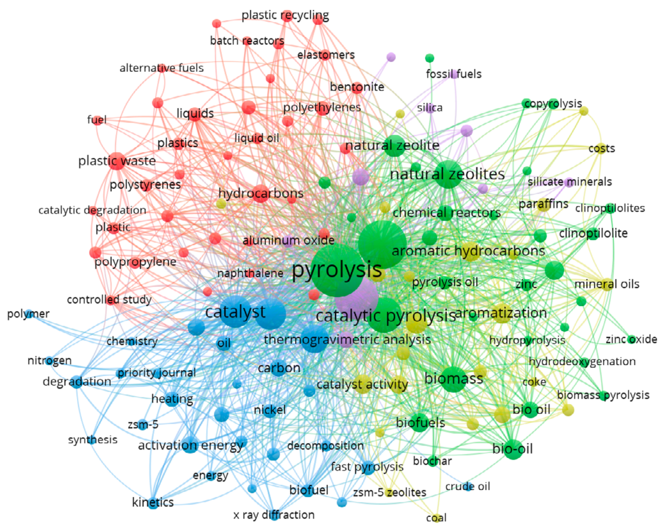


**Figure 3.** Correlation keywords in Scopus database

### 3.1.3. Publications per country

The global distribution of publications (Figure 4) reveals a geographically diverse but relatively specialized research landscape. Indonesia leads with 53 publications, representing approximately 20.4% of the total output, followed by China (22 publications, 8.5%), India (16 publications, 6.2%), and Saudi Arabia (15 publications, 5.8%). Other notable contributors include the United Kingdom and Malaysia (13 publications each, 5.0%), and South Korea (12 publications, 4.6%). Collectively, these countries account for a significant proportion of the global research activity, indicating a regional concentration, particularly in Asia. The predominance of Indonesia in terms of publication output (20.4%) can be partly attributed to the strong research focus on locally abundant natural resources, particularly zeolitic materials from regions such as Java. These natural zeolites have been widely investigated as low-cost catalysts in pyrolysis and related thermochemical processes, which has stimulated a relatively high number of experimental studies within the country. Therefore, Indonesia's leading position should not be interpreted as a clear global dominance in the field, but rather as a reflection of a geographically localized research niche driven by resource availability and applied research interests. This pattern highlights how regional material accessibility can shape publication volume, while not necessarily correlating with broader scientific impact or leadership, in terms of highly cited or foundational contributions. However, several countries with moderate or lower publication counts, such as the United States, Spain and Germany (each contributing between 1-2%), may still play a relevant role in terms of scientific impact, as publication volume does not necessarily reflect citation influence. The remaining countries contribute marginally, with individual shares below 3%, highlighting the fragmented and specialized nature of the field. Overall, this distribution suggests that research on this topic is characterized by localized clusters of activity and emerging global participation, rather than by a clearly dominant international leader.

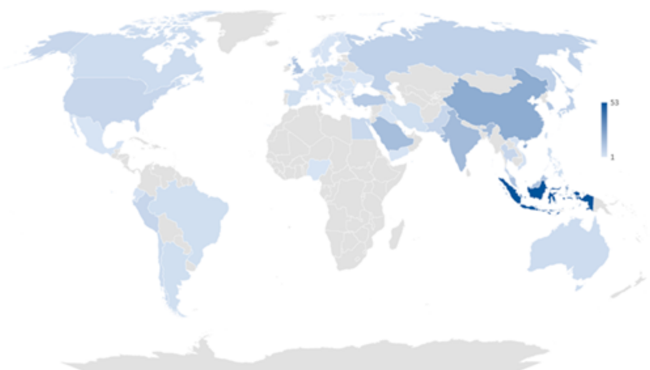

**Figure 4.** Publications per country in the Scopus database

#### *3.1.4. Publications per funding sponsor*

Table 1 summarizes the distribution of publications by funding sources, providing key insights into the global research landscape on natural zeolites in biomass pyrolysis.

The National Natural Science Foundation of China (NSFC) leads with 13 funded publications, followed by the National Research Foundation of Korea (6 publications), while other institutions contribute more modestly. Despite the global prominence of NSFC, this relatively low number of funded studies suggests that the topic is not yet a major national priority, but rather represents an emerging and specialized research niche.

A broader examination highlights the participation of institutions from Asia and Latin America, reflecting diverse and localized research dynamics. Latin American institutions, particularly Universidad del Bío-Bío (Chile) and Universidad Nacional de San Agustín de Arequipa (Peru), exhibit a notable level of participation. This pattern indicates the presence of consolidated research groups with sustained productivity and specialized expertise, which may play a significant role in advancing the field despite operating within smaller funding ecosystems. These cases illustrate that scientific relevance in this domain is not solely determined by the scale of funding, but also by the continuity and focus of research groups. In Indonesia, institutions such as Kementerian Pendidikan, Kebudayaan, Riset, dan Teknologi and Universitas Negeri Medan show sustained contributions, likely driven by the availability of natural zeolitic resources and their application in catalytic processes. This resource-based advantage has enabled Indonesia to develop a relatively strong and consistent research output, constituting an example of early and localized leadership within this niche field. Similarly, institutions from Saudi Arabia (King Abdulaziz University), Pakistan (National University of Sciences and Technology) and Malaysia (Ministry of Higher Education) contribute through targeted and application-oriented research initiatives. Their relatively modest but consistent output suggests focused scientific efforts aligned with specific technological or industrial interests, rather than broad national funding strategies.

Overall, the funding landscape suggests that the field is in an exploratory stage of development, characterized by dispersed institutional participation, regionally driven research priorities, and the absence of a clearly dominant global leader.

It is important to note that percentage contributions were not calculated, as many publications are supported by multiple institutions simultaneously. Consequently, the cumulative number of funding bodies exceeds the total number of documents, and reporting percentages could result in distorted interpretations of institutional participation.

 DOI: 10.21142/SS-0702-2026-e153

**Table 1.** Publications per funding sponsors in Scopus database

| Journal | Country | N. |
|---|---|---|
| National Natural Science Foundation of China | China | 13 |
| National Research Foundation of Korea | Korea | 6 |
| National University of San Agustín - Arequipa | Peru | 5 |
| University of Bío-Bío | Chile | 5 |
| Kementerian Pendidikan, Kebudayaan, Riset, dan Teknologi | Indonesia | 5 |
| Ministry of Trade, Industry and Energy | Korea | 4 |
| Kementerian Pendidikan, Kebudayaan, Riset, dan Teknologi | Indonesia | 4 |
| National University of Sciences and Technology | Pakistan | 3 |
| National Key Research and Development Program of China | China | 3 |
| Ministry of Science, ICT and Future Planning | Korea | 3 |
| Ministry of Higher Education, Malaysia | Malaysia | 3 |
| Universitas Negeri Medan | Indonesia | 3 |
| King Abdulaziz University | Saudi Arabia | 3 |
| Fundamental Research Funds for the Central Universities | China | 3 |
| Foundation of State Key Laboratory of Coal Combustion | China | 3 |

### *3.1.5. Publications per affiliation*

The institutional distribution of publications (Figure 5) indicates that research on natural zeolites applied to biomass pyrolysis is concentrated within a relatively small number of universities and research centers, reflecting the specialized and resource-driven nature of the field. Universitas Gadjah Mada (UGM, Indonesia) emerges as one of the most active institutions, supporting Indonesia's role as an early and localized contributor to this research area. Similarly, King Abdulaziz University (KAU, Saudi Arabia) and the Centre for Energy and Environmental Sustainability (CEES, Nigeria) demonstrate sustained participation, likely linked to growing regional interest in alternative fuels and catalytic technologies.

Contributions from Europe are primarily associated with institutions such as the Technical University in Zvolen (TvSTU, Slovakia) and Babeș-Bolyai University (UBB, Romania). This pattern suggests a strong correlation between research activity and the availability of natural zeolite deposits, particularly the clinoptilolite-rich formations found in Eastern European regions. The limited presence of major Western European institutions, traditional leaders in catalysis research, indicates that this topic has not yet been integrated fully into mainstream catalytic research agendas, reinforcing its characterization as a niche or emerging field.

In Latin America, institutions such as Universidad Nacional de Salta (UNSA, Argentina), Universidade Estadual de Londrina (UNILA, Brazil), and Universidad del Bío-Bío (UBB, Chile) have contributed through focused research groups with specialized expertise. Although their publication output remains moderate, their consistent activity suggests the presence of consolidated research lines within specific institutional contexts.

Overall, the observed distribution does not reflect a globally dominant institutional landscape, but rather a set of geographically distributed research clusters driven by local resource availability and applied research priorities. While a significant proportion of research activity is concentrated in Asia, this pattern is better interpreted as an expression of localized or initial leadership within a specialized field, rather than as a consolidated global dominance. This highlights the importance of considering the resource-institution relationship when interpreting publication patterns, as well as the need to foster greater international collaboration, in order to strengthen the contributions of institutions from Latin America, Africa and Europe, and to facilitate the integration of this topic into broader catalysis research frameworks (Zhang *et al.*, 2020; Li *et al.*, 2021; Chen *et al.*, 2022).

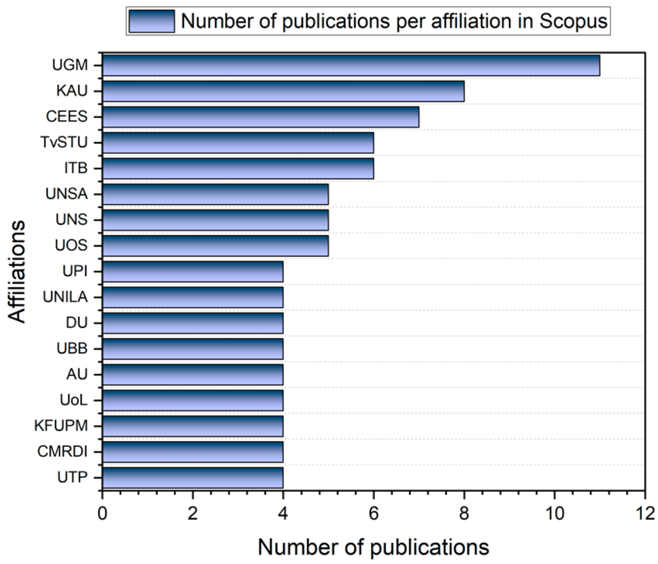


**Figure 5.** Publications per affiliation in Scopus database

### *3.1.6. Publications per subject area*

Figure 6 presents a treemap illustrating the disciplinary distribution of publications addressing natural zeolites as catalysts in biomass pyrolysis. The dominant fields — Energy, (17.65%), Engineering (15.25%), Environmental Science (14.60%) and Chemical Engineering (14.38%)—reflect the multidisciplinary character of the research and its strong alignment with global priorities in sustainable energy production and waste valorization. These disciplines constitute the core technological framework, focusing on catalytic performance, process optimization and product upgrading. A notable proportion of contributions arises from the fields of Chemistry (10.24%)

and Materials Science (8.71%), underscoring ongoing efforts to characterize the physicochemical properties of natural zeolites, modify their structures and improve catalytic efficiency. This emphasis highlights the critical role of material design and functionalization in enhancing pyrolysis yields and selectivity. The limited representation of Physics and Astronomy (5.01%), as well as Planetary Sciences (3.05%), appears marginal and likely results from the multidisciplinary scope of the journals in which these studies are published, rather than indicating a direct or substantial contribution from these fields. Smaller but meaningful shares from the perspectives of Computer Science (2.83%), Mathematics (2.18%), Social Sciences (1.74%), Business and Accounting (1.53%), Agricultural Sciences (1.53%) and Biological Sciences (1.31%) indicate the gradual incorporation of computational modeling, life-cycle assessments, techno-economic analyses and sustainability evaluations. At the same time, the relatively limited contribution from Agricultural Sciences is noteworthy, given the central role of biomass as the primary feedstock in pyrolysis processes. This suggests a disciplinary imbalance in the field, where research has been predominantly driven by chemical and engineering perspectives focused on catalyst development, with comparatively limited integration of agricultural knowledge related to biomass production, variability and supply systems. This disconnect highlights a critical research gap and underscores the need for stronger interdisciplinary collaboration, in order to fully align feedstock characteristics with catalytic and process optimization strategies. Smaller but meaningful shares from the perspectives of Computer Science (2.83%), Mathematics (2.18%), Social Sciences (1.74%), Business and Accounting (1.53%), Agricultural Sciences (1.53%) and Biological Sciences (1.31%) indicate the gradual incorporation of computational modeling, life-cycle assessments, techno-economic analyses and sustainability evaluations.

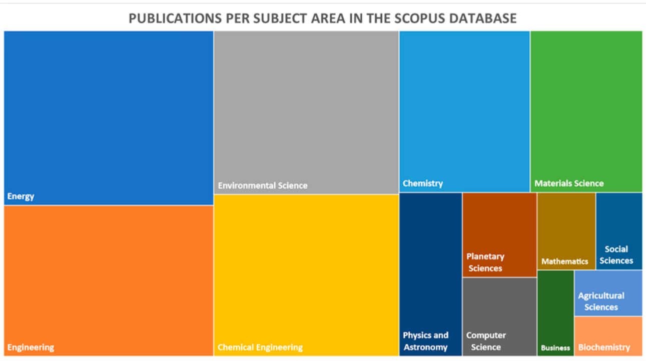


**Figure 6.** Publications per subject area in the Scopus database

### *3.1.7. Publications per journals*

Table 2 presents the distribution of publications across journals indexed in the Scopus database. For each journal, the publisher, quartile ranking and SJR2026 were retrieved from the official Scimago Journal & Country Rank (SJR) website (Scimago Journal and Country Rank, 2026), thereby ensuring the reliability and consistency of the bibliometric indicators. This information provides an overview of the quality, visibility and scientific influence of the journals through which the research outputs have been disseminated.

The results indicate a strategic preference for high-impact outlets. Prominent examples include the Journal of Analytical and Applied Pyrolysis, Energy, Polymer Degradation and Stability, Journal of the Energy Institute, Fuel, Energy and Fuels, and Bioresource Technology —all ranked in Q1— which together account for a substantial proportion of the total publications, with individual contributions ranging from four to nine articles. This suggests strong orientation toward journals with rigorous peer-review standards and recognized international visibility.

Meanwhile, journals with lower quartile rankings or impact factors, such as Key Engineering Materials (Q4), and Advanced Materials Research (Q4), and conference proceedings (e.g., IOP Conference Series: Earth and Environmental Science, and Multi Conference for Engineering Education and Technology, suggest that a significant portion of the research remains at an exploratory or regional level, yet to meet the rigorous characterization requirements of top-tier catalytic journals.

The concentration of articles in Q1 journals underscores researchers' emphasis on visibility and impact. However, the complementary presence of Q2-Q4 journals and conference proceedings highlights the use of alternative dissemination pathways that facilitate rapid knowledge exchange, collaboration and innovation at various research stages. Moving forward, the maintaining of a balanced publication strategy between established high-impact journals and emerging interdisciplinary platforms could enhance accessibility, foster cross-sector collaboration and accelerate the global development of the field.

**Table 2.** Publications per journals in the Scopus database

| Journal | Editorial | Quartile | SJR2026 | N. |
|---|---|---|---|---|
| Journal of Analytical and Applied Pyrolysis | Elsevier | Q1 | 1.28 | 9 |
| IOP Conference Series Earth and Environmental Science | IOP | | 0.21 | 9 |
| Key Engineering Materials | Trans Tech | Q4 | 0.16 | 6 |
| Sustainability Switzerland | MDPI | Q1 | 0.69 | 5 |
| Materials Today Proceedings | Elsevier | Q2 | 0.59 | 5 |
| Energy | Elsevier | Q1 | 2.21 | 5 |


DOI: 10.21142/SS-0702-2026-e153

| | | | | |
|---|---|---|---|---|
| Polymer Degradation and Stability | Elsevier | Q1 | 1.24 | 4 |
| Journal of the Energy Institute | Elsevier | Q1 | 1.21 | 4 |
| Fuel | Elsevier | Q1 | 1.61 | 4 |
| Energy and Fuels | ACS | | | |
| Q1 | 1.12 | 4 | | |
| Bioresource Technology | Elsevier | Q1 | 2.40 | 4 |
| Advanced Materials Research | Trans Tech | Q4 | | 4 |
| Multi Conference for Engineering Education and Technology | LACCEI | | 0.11 | 3 |
| E3S Web of Conferences | EDP Sciences | | 0.21 | 3 |
| Chemical Engineering Transactions | AIDIC | Q3 | 0.26 | 3 |
| AIP Conference Proceedings | AIP | | 0.15 | 3 |

### *3.1.8. Most cited papers*

The dataset summarized in Table 3 reveals substantial heterogeneity in the application of natural zeolites and related catalysts for biomass and waste pyrolysis, in terms of both feedstock diversity and operational parameters.

Highly cited studies in the field can be broadly divided into two categories: foundational works on pyrolysis technology and specialized research on catalytic upgrading using natural zeolites. Seminal contributions such as those by Bridgwater (1996) represent «general pyrolysis classics», providing a comprehensive overview of fast pyrolysis processes and their technological development, rather than focusing specifically on zeolite-based catalysis.

In contrast, more recent highly cited studies (Miandad *et al*., 2017; Miandad *et al*., 2019) specifically address the role of natural and modified zeolites in enhancing liquid oil yields, particularly from plastic feedstocks such as PS, PE and PP. These studies highlight acid activation and thermal treatment as the most common modification methods, with acid-treated zeolites consistently exhibiting superior catalytic performance by promoting aromatic hydrocarbon formation.

For lignocellulosic biomass, low-cost natural oxides (e.g., MgO and clinoptilolite) have demonstrated competitive performance compared to synthetic zeolites such as ZSM-5. Rather than maximizing deoxygenation alone, natural zeolites offer a more balanced deoxygenation-to-coke ratio, which can reduce excessive coke formation while maintaining acceptable levels of oxygen removal. This behavior, combined with their low cost and availability, makes them attractive alternatives for practical and scalable catalytic pyrolysis applications (Stefanidis *et al*., 2016). These findings indicate that natural catalysts offer not only economic benefits, but also deliver effective functional properties for improving bio-oil quality. Moreover, composite catalysts (e.g., Mg-Al LDO/ZSM-5) display promising synergistic effects by balancing higher heating values, reduced nitrogen content and selective conversion of pyrolysis vapors (Gao *et al*., 2017).

The variety of feedstocks —ranging from lignin, rice husk, cottonseed cake and cassava rhizome, to plastic and tire wastes— demonstrates that catalytic pyrolysis using natural zeolites constitutes a robust thermo-chemical platform. However, product yields vary widely (20-80%), reflecting a strong dependence on feedstock composition, catalyst structure and operating parameters. Interestingly, while natural zeolites tend to favor the production of aliphatic compounds and liquid fractions, synthetic zeolites such as ZSM-5 enhance aromatic hydrocarbon selectivity, highlighting complementary functionalities that could be exploited in hybrid catalyst systems.

A critical observation is related to the temporal evolution of research. Earlier studies (1990s-2000s) primarily investigated feasibility and basic product characterization, whereas more recent contributions (post-2010) emphasize pilot-scale testing, advanced analytical techniques (GC-MS, FTIR, TGA-MS) and functional catalyst modifications. Despite these advances, scalability remains a major challenge, as only a limited number of studies have examined catalyst regeneration, long-term stability or integration into continuous processes. However, the low cost and widespread availability of natural minerals may enable a shift in perspective, moving from conventional regeneration-intensive strategies toward a «disposable catalyst» model. In this context, catalysts could be used for shorter operational cycles and replaced rather than regenerated, potentially reducing operational expenditures (OPEX) and simplifying process design. This approach may be particularly attractive to decentralized biorefineries, where economic and logistical constraints favor robust, low-cost and easy-to-handle catalytic systems.

From a bibliometric standpoint, citation frequency correlates strongly with studies reporting novel catalytic mechanisms or high liquid oil yields, underscoring the research community's focus on practical energy recovery. Nevertheless, techno-economic assessments, environmental trade-offs and the integration of natural zeolite-based pyrolysis within broader circular bioeconomy frameworks have received comparatively little attention. The addressing of these gaps constitutes a clear opportunity for future research.

**Table 3.** Publications per journal in Scopus database

| Biomass used | Catalysts used | Catalyst modifications | Main results | Journal | Year | Cited | Reference |
|---|---|---|---|---|---|---|---|
| Biomass and solid wastes (general) | Natural catalysts in biomass | Reinforcement or removal of natural catalysts | - Catalytic processes can upgrade pyrolysis products to higher value fuels. - Natural catalysts influence product yield and composition. | Catalysis Today | 1996 | 340 | (Bridgwater, 1996) |
| Lignocellulosic biomass | Low-cost natural MgO | Produced from magnesite, calcined, crushed, sieved | - Reduced oxygen content in bio-oil, similar/better than ZSM-5. - Promoted deoxygenation via $CO_2$ formation. - Slightly higher coke, easily regenerated. | Applied Catalysis B Environmental | 2016 | 236 | (Stefanidis *et al.*, 2016) |
| Spent FCC catalyst | Aspergillus niger | Batch bioleaching at various pulp densities | - Higher metal extraction than chemical leaching (Ni, Fe, Al, V, Sb). - Extraction decreased with higher pulp density. | Journal of Biotechnology | 2005 | 200 | (Aung and Ting, 2005) |
| Cotton-seed cake | Clinoptilolite | Varying content (1-20 wt%), pyrolysis at 550°C, 100 $cm^3$/min | - Max liquid yield 30.84% at 20% catalyst. - Chemical composition analyzed. | Bioresource Technology | 2006 | 137 | (Pütün *et al.*, 2006) |
| Cassava rhizome | ZSM-5, Al-MCM-41, Al-MSU-F, metal oxides, commercial catalysts, natural catalysts | Analytical pyrolysis, Py-GC/MS | - Zeolites, commercial catalysts, Cu chromite reduced oxygenated lignin derivatives. - ZSM-5, Criterion-534, Al-MSU-F enhanced aromatics and phenols. | Fuel | 2010 | 120 | (Pattiya *et al.*, 2010) |
| Rice husk | Industrial waste coal bottom ash, nickel, natural zeolite | Catalyst to rice husk ratio 0.1, TGA-MS 323-1173 K | - Coal bottom ash: lowest coke (3.65 wt%), highest syngas (68.3 vol%), $H_2$ +8.4 vol%. | Energy Conversion and Management | 2018 | 116 | (Loy *et al.*, 2018) |
| Natural zeolite (KSA) | Natural zeolite | Characterization for energy applications | - High thermal stability, adsorption, ion exchange; suitable for WTE and solar energy. | Energy | 2016 | 116 | (Nizami *et al.*, 2016) |
| Eucalyptus oil | Fe/Co on high silica zeolite | Spray pyrolysis, 850°C, 5 wt.% | - Synthesized single-walled carbon nanotubes (SWNTs), 0.79-1.71 nm, well graphitized. | Materials Letters | 2007 | 105 | (Ghosh *et al.*, 2007) |
| Lignin | Natural zeolite (NZ, in situ), HZSM-5 (ex situ) | Two-stage catalytic pyrolysis | - Larger aromatics yield, smaller coke amount; effect maximized at higher NZ amount and HZSM-5 temperature (600°C). | Polymer Degradation and Stability | 2001 | 100 | (Lee *et al.*, 2001) |
| Biomass pyrolysis vapors | Clinoptilolite (NZ), ZSM-5, H-Y | Catalyst/feed ratio 0.05-0.25; 350-500°C | - Synthetic zeolites enhanced gas yield; NZ favored bio-oil production. - Long-chain hydrocarbons converted to lower-weight; NZ better for aliphatics, ZSM-5 for aromatics. | Journal of Cleaner Production | 2018 | 98 | (Loy *et al.*, 2018) |
| Euphorbia rigida | Criterion-534, activated alumina, natural zeolite | 550°C, heating rate 7°C/min, particle size 0.55 mm | - Oil yields: 27.5% NZ, 31% Criterion-534, 28.1% alumina, 21.6% no catalyst. - Catalysts influenced chemical composition. | Environmental Science and Pollution Research | 2017 | 88 | (Ma *et al.*, 2017) |
| Peanut shells | Clinoptilolite (native and ion-exchanged) | Ion exchange to develop Brønsted acid sites | - Reduced oxygen content; improved HHV; slight decrease in bio-oil yield, increase in gases. - Ion-exchanged catalyst better performance. | Process Safety and Environmental Protection | 2018 | 86 | (Miandad *et al.*, 2018) |
| Solid biomass pyrolysis oils | FCC catalyst | Co-feeding with model hydrocarbon (C8) | - Upgrading of oxygenated compounds; potential for large-scale refinery fuel production. | Energy and Fuels | 2009 | 83 | (Pütün *et al.*, 2009) |

### *3.2. Thematic analysis guided by research questions*

Guided by the selected literature, a structured thematic analysis was conducted to address the research questions, synthesizing evidence on zeolite-catalyzed biomass pyrolysis with an emphasis on bio-oil yield, selectivity and product quality. It highlights key differences and convergences between natural and synthetic zeolites, while identifying gaps and opportunities. It is this framework which organizes the discussion below.

#### *3.2.1. What are the comparative catalytic performances of natural versus synthetic zeolites in biomass pyrolysis in terms of bio-oil yield, selectivity and product quality?*

Across biomass pyrolysis studies, synthetic zeolites —particularly HZSM-5— generally sacrifice liquid yield in order to achieve greater selectivity and product quality, whereas natural zeolites (e.g., clinoptilolite) tend to preserve higher liquid fractions with broader product distributions. Under comparable conditions, mixed plastic/biomass feedstocks processed over natural zeolites yield more liquid oil than those catalyzed by synthetic zeolites, although with lower aromatic selectivity. In contrast, HZSM-5 promotes deeper cracking and aromatization, reducing total bio-oil yield while enriching BTX-range hydrocarbons (Nishu *et al*., 2020).

HZSM-5's shape-selective 10-membered ring channels and strong Brønsted acidity drive dehydration, decarbonylation, oligomerization and cracking, converting oxygenates to mono-aromatics and light olefins. Metal promotion (Ga, Zn, Ni) further enhances aromatization. Natural zeolites, with lower Si/Al ratios and wider pores, favor aliphatics and wide-boiling distillates, unless modified by acid activation or metal doping to increase acidity and constrain diffusion (Lok *et al*., 2019) (Khan *et al*., 2021).

Reports using mixed plastic/biomass feeds generally show higher liquid yields over natural zeolite (e.g., PS giving ~54% vs. ~50% with synthetic), while HZSM-5 typically shifts carbon toward gases and water via deeper deoxygenation and cracking, thereby reducing organic liquid yield. This yield-selectivity trade-off persists from bench to pilot scale (Miandad *et al*., 2017).

Despite lower yields, synthetic zeolites produce higher-quality oils characterized by lower O/C ratios, higher heating values (HHV), and greater aromatic carbon content —which correlates with enhanced post-condensation stability. Model-compound studies (e.g., guaiacol over ZSM-5) mechanistically confirm extensive deoxygenation and aromatization but also reveal a strong coking tendency that can shorten catalyst lifetime. Hierarchical or metal-promoted ZSM-5 mitigates these diffusional limitations and deactivation effects. Natural zeolites, sometimes metal-modified, can meaningfully reduce oxygenates and improve HHV relative to non-catalytic runs, yet seldom match ZSM-5's aromatic selectivity, without additional modification or conversion to the H-form (Zhang *et al*., 2016).

When the objective is to maximize liquid yield with moderate upgrading at lower catalyst cost, natural zeolites —particularly those acid-activated or metal-modified— are attractive (Yan *et al*., 2023). Conversely, if the goal is to obtain fuel-range, aromatic-rich, low-oxygen oils, HZSM-5 (optionally metal-promoted or hierarchical) is superior, despite the lower organics yield, provided that coke formation is managed through regeneration cycles or morphology control (Nishu *et al*., 2020). Recent reviews emphasize integrating zeolite catalysis with advanced heating methods (e.g., microwave catalytic pyrolysis) and *in situ/ex situ* vapor upgrading, in order to balance yield and quality while addressing coking and scale-up challenges (Ke *et al*., 2024).

#### *3.2.2. How do modification strategies (e.g., acid treatment, ion exchange, metal impregnation) influence the physicochemical properties and catalytic efficiency of natural zeolites?*

A comprehensive thematic analysis of modification strategies reveals that acid activation, ion exchange and metal impregnation profoundly refine the physicochemical properties of natural zeolites —modulating acidity, porosity, Si/Al ratio and metal-dispersion— and thereby dictating catalytic performance during biomass pyrolysis.

##### *a. Acid treatment*

Acid treatment selectively removes framework aluminum, increases the Si/Al ratio, and introduces mesoporosity, improving mass transfer and access to active sites. For instance, acid-activated natural zeolite yielded bio-oil enriched in phenolics and aromatics during pine cone pyrolysis, enhancing fuel quality relative to non-catalytic runs (Zulkania *et al*., 2019). In hydrotreating contexts (e.g., α-cellulose bio-oil), sequential HCl and NaOH treatments on synthetic Zeolite Y generate hierarchical, mesoporous forms with reduced acidity but increase liquid yield (Mandela *et al*., 2020). A recent review concluded that acid/base treatments enhance ion-exchange capacity, surface area and Si/Al ratios, although challenges remain in terms of sustainability and cost-effectiveness (Suyitno *et al*., 2024).

##### *b. Ion exchange*

Ion exchange (e.g., $K^+$ for $H^+$) moderates strong Brønsted acidity and balances acid-base functionality. In a wheat-straw pyrolysis, a partially $K^+$-exchanged ZSM-5 (KH-ZSM-5) achieved more efficient deoxygenation with reduced coke formation and a favorable bio-oil yield-oxygen content ratio, compared to the parent H-ZSM-5 (Alejandro-Martín *et al*., 2018). This demonstrates how tailored acidity via ion exchange can attenuate excessive cracking while preserving liquid stability.

#### *c. Metal impregnation*

Metal-doped natural zeolites (e.g., $Cu^5H_2NZ$, $Ni^5H_2NZ$) significantly modify product selectivity. Py-GC/MS studies using Chilean natural zeolites modified via dual ion exchange and metal impregnation showed enhanced BTX (benzene, toluene, xylene) yields from lignocellulosic biomass (Venegas-Vásconez *et al.*, 2024). Similarly, Ni-impregnated Aceh natural zeolite (Ni/ZAA) exhibited uniform Ni dispersion, confirmed via TEM and XRD, indicating improved active-site accessibility for gasoline-range hydrocarbon (Husin *et al.*, 2023). In hydropyrolysis experiments (Ni-Fe on clinoptilolite and BEA), BEA incorporation improved $H_2$ adsorption, acidity balance and aromatic monomer selectivity, while mitigating coking and over-cracking (Yan *et al.*, 2023).

### *3.2.3. What are the main sustainability advantages of using natural zeolites over synthetic ones, including economic, environmental and energy considerations?*

Natural zeolites offer clear sustainability advantages over synthetic analogues when evaluated across economic, environmental and energy dimensions (Lang *et al.*, 2024; Tao *et al.*, 2024). In economic terms, their production entails straightforward mining followed by minimal processing (crushing and sieving), yielding unit costs up to almost three orders of magnitude lower than those of hydrothermally synthesized zeolites (e.g., USD 0.24/kg vs. USD 120/kg), which require expensive precursors, specialized equipment and complex infrastructure (Khaleque *et al.*, 2020). This cost differential becomes critical in large-scale catalytic operations, where frequent regeneration or replacement directly influences both capital and operating expenditures (Pérez-Ramírez *et al.*, 2008).

In environmental terms, natural zeolites possess substantially lower life-cycle footprints (Zheng *et al.*, 2022). The de-novo synthesis of aluminosilicates entails high-enthalpy processes, including gel preparation, hydrothermal crystallization and calcination, which collectively generate notable greenhouse gas emissions. Conversely, natural zeolites are geologically formed and require only comminution and classification, resulting in a markedly reduced global warming potential (Grimaldi *et al.*, 2021). Life-cycle assessments indicate up to an 8% reduction in carbon footprint for construction and catalytic systems when natural zeolites replace synthetic additives (Anekwe *et al.*, 2025).

From an energetic standpoint, natural zeolites are preferable due to their low cradle-to-gate energy demand, dominated by low-density mining operations. In contrast, synthetic zeolites require substantial thermal and chemical energy inputs for crystallization and template removal. Despite advances in greener synthetic methods, synthetic zeolites still exhibit higher baseline energy intensities than natural clinoptilolite (Pérez-Botella *et al.*, 2022) (Shi *et al.*, 2025). In biomass pyrolysis, where catalytic upgrading already entails significant thermal input, employing low-embodied-energy catalysts, such as natural zeolites, can reduce the total energy consumption per liter of upgraded bio-oil (Mijailović *et al.*, 2022).

Collectively, their economic affordability, lower environmental burden and reduced energy intensity position natural zeolites as sustainable and multifunctional materials for future large-scale applications in energy, environmental remediation and biomass valorization.

To address this limitation, a more explicit quantitative comparison has been incorporated. Recent life-cycle assessment (LCA) studies have reported that the cradle-to-gate energy demand for synthetic zeolites ranges between 60-120 $MJ \cdot kg^{-1}$, whereas natural zeolites such as clinoptilolite typically require less than 5-10 $MJ \cdot kg^{-1}$, due to the absence of hydrothermal synthesis and template removal steps (Król, 2020). Similarly, global warming potential (GWP) values for synthetic zeolites can exceed 5-8 kg $CO_2$-eq$\cdot kg^{-1}$, compared to <1 kg $CO_2$-eq$\cdot kg^{-1}$ for natural counterparts (Chen *et al.*, 2024). From an economic perspective, reported costs range from ~USD 100-150 $kg^{-1}$ for high-silica synthetic zeolites versus <USD 1 $kg^{-1}$ for natural zeolites (Shi *et al.*, 2025). These quantitative indicators confirm that natural zeolites offer reductions of one to two orders of magnitude in energy use, carbon footprint and cost, thereby substantiating their sustainability advantages in catalytic and large-scale industrial applications.

### *3.2.4. What are the main research trends, thematic focuses and knowledge gaps identified in the ten most highly cited open-access articles published in the last five years on natural zeolites in biomass pyrolysis?*

Table 4 summarizes these ten studies along with their most significant contributions. Analysis of these high-impact publications reveals several key research trends, including the growing emphasis on the use of natural zeolites as low-cost and environmentally friendly catalysts, the optimization of catalytic performance for enhanced bio-oil quality and aromatic hydrocarbon production, and the increasing integration of co-pyrolysis strategies involving biomass and plastic waste. Thematically, the literature exhibits a strong focus on catalyst modification, acidity control and structural properties to improve selectivity and stability. However, important knowledge gaps remain, particularly regarding long-term catalyst deactivation mechanisms, regeneration efficiency, and the lack of standardized methodologies for comparing natural and synthetic zeolites under similar operating conditions. Furthermore, limited attention has been given to large-scale implementation and techno-economic assessments, highlighting the need for future research to bridge the gap between laboratory findings and industrial applications.


DOI: 10.21142/SS-0702-2026-e153

**Table 4.** Most cited publications after applying the PRISMA methodology

| Title | Journal | Year | Cites | Cited | Reference |
|---|---|---|---|---|---|
| Green hydrocarbons and fuels from municipal polymer waste co-fed with natural gas using a batch catalytic slurry process | *Green Technologies and Sustainability* | 2024 | 66 | Municipal polymer waste was converted into high-octane green fuels via catalytic depolymerization using methane and zeolite catalysts in a slurry reactor. Temperature and polymer-to-catalyst ratio were key variables. Methane enhanced aromatic production, achieving up to 89% aromatics. The process demonstrates sustainable waste valorization through zeolite-catalyzed conversion of plastics into valuable hydrocarbons. | (Fegade, 2024) |
| Synergistic effects of catalytic co-pyrolysis *Chlorella vulgaris* and polyethylene mixtures using artificial neuron network: Thermodynamic and empirical kinetic analyses | *Journal of Environmental Chemical Engineering* | 2022 | 55 | Catalytic pyrolysis of *Chlorella vulgaris* and HDPE mixtures was evaluated using HZSM-5, natural limestone, and bifunctional HZSM-5/LS catalysts. The bifunctional system significantly reduced activation energy. Kinetic modeling showed Flynn-Wall-Ozawa as most accurate. ANN-GA modeling supported predictions, highlighting synergistic effects in co-pyrolysis and improved catalytic performance. | (Yap *et al.*, 2022) |
| Catalytic hydropyrolysis of biomass using natural zeolite-based catalysts | *Chemical Engineering Journal* | 2023 | 36 | Catalytic hydropyrolysis of *Eucalyptus globulus* using natural clinoptilolite (Escott), BEA, and mixed zeolites supported Ni-Fe catalysts improved liquid and gas yields. The Escott-BEA system enhanced aromatic selectivity and reduced coke formation. Catalyst acidity and metal-support interactions governed product distribution, highlighting the effectiveness of natural clinoptilolite-based catalytic systems. | (Yan *et al.*, 2023) |
| Effective hydrodeoxygenation bio-oil via natural zeolite supported transition metal oxide catalyst | *Arabian Journal of Chemistry* | 2023 | 23 | Bio-oil upgrading was performed via hydrodeoxygenation using natural zeolite impregnated with transition metal oxides. ZnO/HZ (5 wt%) showed the highest performance, achieving ~50% organic phase conversion. Improved physicochemical properties were obtained, demonstrating that natural zeolite-based catalysts provide an efficient and low-cost approach for enhancing biofuel quality. | (Sihombing *et al.*, 2023) |
| Microwave catalytic pyrolysis of solid digestate for high quality bio-oil and biochar | *Journal of Analytical and Applied Pyrolysis* | 2024 | 16 | Microwave-assisted catalytic pyrolysis of solid digestate using natural zeolite and $K_3PO_4$ was evaluated at 300-500 °C. Optimal conditions yielded high aromatic hydrocarbons (>90%) and reduced bio-oil acidity. Catalyst type and temperature influenced product distribution, demonstrating the effectiveness of natural zeolite for upgrading waste into fuels and value-added materials. | (An *et al.*, 2024) |
| Thermo-catalytic pyrolysis of sewage sludge and techno-economic analysis: The effect of synthetic zeolites and natural sourced catalysts | *Bioresource Technology* | 2024 | 16 | Pyrolysis of animal sewage sludge was evaluated using synthetic (ZSM-5, Y-zeolite) and natural catalysts (diatomite, kaolin, perlite). Catalysts enhanced volatile yields and improved bio-oil properties. Kaolin showed increased activity at 500-600 °C, while higher temperatures promoted hydrogen-rich gas production, demonstrating the effectiveness of natural materials in catalytic waste valorization. | (Csutorás and Miskolczi, 2024) |
| Biomass Valorization through Catalytic Pyrolysis Using Metal-Impregnated Natural Zeolites: From Waste to Resources | *Polymers* | 2024 | 12 | Catalytic pyrolysis of lignocellulosic biomass using Chilean natural zeolite and metal-modified variants (Cu, Ni) enhanced BTX production. Ni-modified zeolite showed highest aromatic yields and reduced oxygenates. Improved performance was attributed to increased acidity and metal sites, promoting deoxygenation and aromatization reactions, thus upgrading bio-oil quality. | (Venegas-Vásconez *et al.*, 2024) |
| Addition of Hydrocarbon Components to Products in the Catalytic Pyrolysis of Sawdust by Natural Catalysts | *Engineered Science* | 2024 | 9 | This study evaluates natural zeolite and dolomite catalysts in sawdust fast pyrolysis. Catalysts reduce bio-oil yield (~2 wt%) but increase gas production and hydrocarbon content. Zeolite improves bio-oil heating value (up to 38.1 MJ/kg), while dolomite enhances gas efficiency, supporting optimized energy recovery strategies. | (Pannucharoenwong *et al.*, 2024) |
| Processing of *Leucaena leucocephala* for renewable energy with catalytic fast pyrolysis | *Energy Reports* | 2022 | 9 | *Leucaena leucocephala* fast pyrolysis (500 °C) with zeolite, kaolin, and dolomite shows catalyst-dependent trade-offs. Kaolin maximizes bio-oil yield (59.6 wt%), zeolite enhances thermal quality (up to 37.3 MJ/kg), and dolomite reduces acidity and improves viscosity. ESP condensation increases heating value and shifts hydrocarbon range toward heavier fractions. | (Duanguppama *et al.*, 2022) |
| Valorization of municipal solid wastes via pyrolysis and hydropyrolysis: Unveiling the role of natural zeolites as catalysts and supports for Ni and Cu | *Journal of Environmental Chemical Engineering* | 2024 | 6 | Pyrolysis and hydropyrolysis of MSW using natural and metal-doped zeolites improve fuel quality. Hydropyrolysis (550 °C, 14 bar $H_2$) reduces oxygenates (~36%) and boosts hydrocarbons. Ni-doped zeolite achieves highest selectivity (88.2%), favoring aromatic compounds (BTEX), highlighting catalyst type and atmosphere as key factors. | (Márquez Negro *et al.*, 2024) |

## 4. Pending issues and research perspectives in biomass catalytic pyrolysis using natural zeolites

Despite the promising potential of natural zeolites as low-cost and environmentally benign catalysts in biomass pyrolysis, several unresolved challenges continue to hinder their full exploitation and large-scale implementation. A critical examination of the literature reveals at least four key research gaps that require urgent attention:

**Understanding lignin conversion intermediates for enhanced selectivity:** Most current studies report product distributions without providing a mechanistic understanding of lignin-derived intermediates. Gaining deeper molecular-level insight into these transient species is essential for improving zeolite selectivity and enabling the production of higher-quality bio-oils with greater yields.

**Deciphering fundamental reaction pathways within zeolitic pores:** The confined pore environment of natural zeolites imposes distinct steric and electronic constraints on biomass molecules during pyrolysis. However, the fundamental thermochemical behavior of cellulose-, hemicellulose- and lignin-derived fragments within these catalytic sites remains poorly understood. Future research should focus on elucidating these mechanisms, in order to better predict the formation of high-value selective products from diverse feedstocks.

**Advancing kinetic modeling to predict biomass thermal behavior:** In order to accurately forecast product selectivity and thermal decomposition patterns, it is necessary to transcend purely empirical correlations towards mechanistic, first-principles modeling. The use of advanced kinetic approaches, such as the Distributed Activation Energy Model (DAEM), offers a promising framework to describe the complex multicomponent reactions that occur in catalytic pyrolysis. However, the main challenge extends beyond intrinsic reaction kinetics to include mass transfer limitations, which are particularly significant in natural zeolites, where intraparticle diffusion often becomes the rate-limiting step. Therefore, future modeling efforts should aim to integrate DAEM with diffusion-related parameters, enabling a more realistic representation of the coupled reaction-diffusion phenomena. Such an approach will be essential for accurately predicting catalyst performance and facilitating the scale-up of catalytic pyrolysis processes.

**Achieving structural and compositional homogeneity in natural zeolites:** Unlike synthetic zeolites, natural zeolites typically display heterogeneity in composition, crystallinity and pore structure, which impedes inter-study reproducibility and statistical validation across studies. Developing strategies for structural modification, purification and standardized pretreatment is therefore critical to ensure systematic comparison of catalytic performance and reliable scale-up.

Collectively, these challenges underscore the need for integrative studies that couple experimental investigation, kinetic modeling and catalyst engineering. The addressing of these knowledge gaps will enhance the fundamental understanding of catalytic pyrolysis and accelerate the rational design of cost-effective, sustainable zeolite-based catalysts capable of delivering consistent and scalable bio-oil production.

## 5. Conclusions

This review demonstrates that natural zeolites represent a viable and sustainable alternative to synthetic catalysts for biomass pyrolysis, providing substantial economic, environmental and energy benefits. Quantitative analyses show cost reductions of up to two orders of magnitude (approximately USD <1 $kg^{-1}$ compared to >USD 100 $kg^{-1}$), as well as lower energy requirements (<10 $MJ \cdot kg^{-1}$ compared to 60-120 $MJ \cdot kg^{-1}$) and a reduced carbon footprint (<1 compared to 5-8 kg $CO_2$-eq·$kg^{-1}$). Bibliometric analysis indicates significant growth in research output over the past decade, with Asia contributing most prominently and clinoptilolite emerging as the most extensively studied natural zeolite framework.

Although synthetic zeolites continue to exhibit superior aromatic selectivity and stability, modified natural zeolites can achieve comparable catalytic performance, particularly when acidity and porosity are optimized through acid activation or metal incorporation. These results underscore the practical potential of natural zeolites as cost-effective catalysts for decentralized or large-scale biofuel production, particularly in resource-limited environments.

However, key limitations persist, including insufficient mechanistic understanding of reaction pathways, variability in natural zeolite composition, and limited data on catalyst regeneration and long-term stability. Based on these gaps, this review proposes a focused research agenda: (i) development of standardized characterization protocols to address material variability; (ii) integration of kinetic modeling and advanced spectroscopy to elucidate reaction mechanisms; (iii) systematic evaluation of catalyst deactivation and regeneration cycles; and (iv) validation under pilot- and industrial-scale conditions.

To summarize, this review offers a quantitative and critical framework to support the transition toward more sustainable catalytic systems, positioning natural zeolites as strategic materials within circular economy models for biomass valorization.


DOI: 10.21142/SS-0702-2026-e153

## Authors' contributions

V.V.D. contributed to the conceptualization, methodology, software, validation, formal analysis, investigation, resources, data curation, writing (original draft), writing (revision and editing), visualization, supervision, project administration and funding acquisition.

A.M.S. contributed to the conceptualization, validation, formal analysis, writing (revision and editing), and supervision.

V.F. contributed to the conceptualization, validation, formal analysis, writing (revision and editing), and supervision.

T.S.L. contributed to the conceptualization, validation, formal analysis, writing (revision and editing), and supervision.

O.E. L.M. contributed to the conceptualization, methodology, validation, formal analysis, investigation, writing (original draft), and writing (revision and editing).

## Funding

The research received no external funding.

## Conflicts of interest

The authors declare no conflicts of interest.

## Artificial intelligence use

In this study, artificial intelligence was utilized exclusively for writing refinement.

## Acknowledgments

We gratefully acknowledge the School of Habitat, Infrastructure and Creativity and the Research Directorate of the Pontifical Catholic University of Ecuador, Ambato Campus for their support in conducting this research.

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