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# Green and Sustainable Zeolite Synthesis: A Review of Waste-Derived Feedstocks and Energy-Efficient Methods

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Abstract

Conventional zeolite manufacturing incurs escalating environmental and economic costs driven by costly reagent-grade precursors, energy-intensive autoclaved heating, and organic structure-directing agents requiring high-temperature calcination, which collectively generate significant waste and emissions. To address this, zeolites—defined as crystalline microporous aluminosilicates with well-defined pore architectures that underpin their widespread industrial utility as catalysts, adsorbents, and ion exchangers—are the focus of recent advancements. This review critically examines green and sustainable synthesis by focusing on two interconnected strategies: waste-derived aluminosilicate feedstocks such as coal fly ash, volcanic ash, kaolin, and waste glass as sustainable silica and alumina sources, and energy-efficient synthesis routes including microwave-assisted, solvent-free, and seed-assisted methods that dramatically lower crystallization time and energy consumption. Furthermore, the review analyzes the vital roles of alkaline fusion pre-treatment, structure-directing agents, Si/Al ratios, and gel compositions in precisely controlling framework type and crystallinity under sustainable constraints. Ultimately, a comprehensive comparative assessment of all major synthesis routes is provided alongside future research directions aimed at achieving scalable, template-free, and waste-valorizing zeolite production.

**Keywords:** Zeolite synthesis, green synthesis, waste-derived feedstocks, sustainable materials, energy-efficient methods

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## Introduction

Zeolites are crystalline microporous aluminosilicate minerals built from corner sharing $TO_4$ tetrahedra (T = Si, Al, or other tetravalent/trivalent heteroatoms) that form regular networks of channels and cavities of molecular dimensions. This unique framework architecture, combined with the presence of exchangeable extra framework cations, gives zeolites their characteristic molecular sieving, ion-exchange, and catalytic properties (Kordala & Wyszkowski, 2024; Vasconcelos et al., 2023) . Since the discovery of synthetic zeolite A and zeolite X by Milton and Breck in the 1940s and 1950s, the field of zeolite synthesis has expanded into a mature and highly active discipline encompassing hundreds of distinct framework topologies recognized by the International Zeolite Association (Nykiel et al., 2026; Clayson et al., 2020). Industrially, zeolites are indispensable in petroleum refining, petrochemical catalysis, gas separation, water softening, and detergent formulation, while their environmental applications in heavy-metal adsorption, wastewater treatment, and soil amendment have grown substantially in recent years (Koshlak, 2023; Shofia et al., 2025). Despite this remarkable utility, the dominant synthesis route conventional hydrothermal crystallization using reagent grade silica and alumina sources carries a significant environmental and economic burden. It relies on high purity precursors, sustained high temperature and high pressure processing in sealed autoclaves, and organic structure directing agents that must be removed by energy intensive calcination, generating volatile organic emissions and wastewater (Nemeș et al., 2026; Dey et al., 2024; Hang et al., 2021).

These challenges have catalyzed a major research shift toward greener and more sustainable zeolite synthesis strategies. Two broad and complementary approaches have emerged as the most impactful. The first is the substitution of conventional precursors with waste derived aluminosilicate feedstocks, including coal fly ash, volcanic ash, kaolin, and waste glass, which are abundant, inexpensive, and would otherwise require costly disposal (Yang et al., 2024; Luhar & Luhar, 2022). The second is the adoption of energy efficient synthesis routes principally microwave-assisted, solvent-free, and seed-assisted methods that sharply reduce crystallization time, energy consumption, and solvent use compared with conventional hydrothermal processing The intersection of these two strategies is particularly promising (Sulistyaningsih et al., 2024; Perera et al., 2025). When waste derived feedstocks are combined with microwave-assisted or alkaline fusion assisted hydrothermal synthesis, the result is a process that simultaneously valorizes industrial by products, reduces synthesis time and energy input, and produces zeolite products of comparable or superior quality to those obtained by conventional means (Tenza & Aphane, 2026; Luukkonen et al., 2018). This synergy represents the frontier of sustainable zeolite manufacturing and is the central focus of this review.

From a structural standpoint, zeolite framework type, Si/Al ratio, crystallinity, and morphology are all governed by synthesis conditions, making the choice of synthesis route not merely a processing detail but a fundamental determinant of product performance (Serrano et al., 2024). Understanding how green synthesis conditions lower temperatures, shorter times, alternative solvents, waste-derived precursors influence these structural outcomes is therefore essential for designing sustainable processes without sacrificing product quality. Historically, the development of new synthesis routes has been driven by the search for novel framework topologies, improved process economics, and more environmentally benign pathways (Zafar et al., 2024; Manyepedza & Kabomo, 2026). Today, the sustainability imperative has moved to the center of this agenda: international pressure to reduce industrial waste, lower carbon footprints, and move toward circular economy principles has made green zeolite synthesis a priority across both academic and industrial research (Stephen et al., 2025).

## Methodology

This review critically synthesizes the recent literature on green and sustainable zeolite synthesis, with emphasis on waste-derived feedstocks and energy-efficient methods. The discussion is organized by synthesis route from alkaline fusion-assisted and microwave-assisted hydrothermal methods to sol-gel, solvothermal, ionothermal, seed-assisted, and solvent-free approaches and culminates in a comparative assessment that highlights the most promising combinations for future sustainable zeolite production. The goal is to provide researchers and engineers with a coherent, evidence-based framework for selecting and developing greener synthesis strategies suited to their specific framework, feedstock, and application requirements.

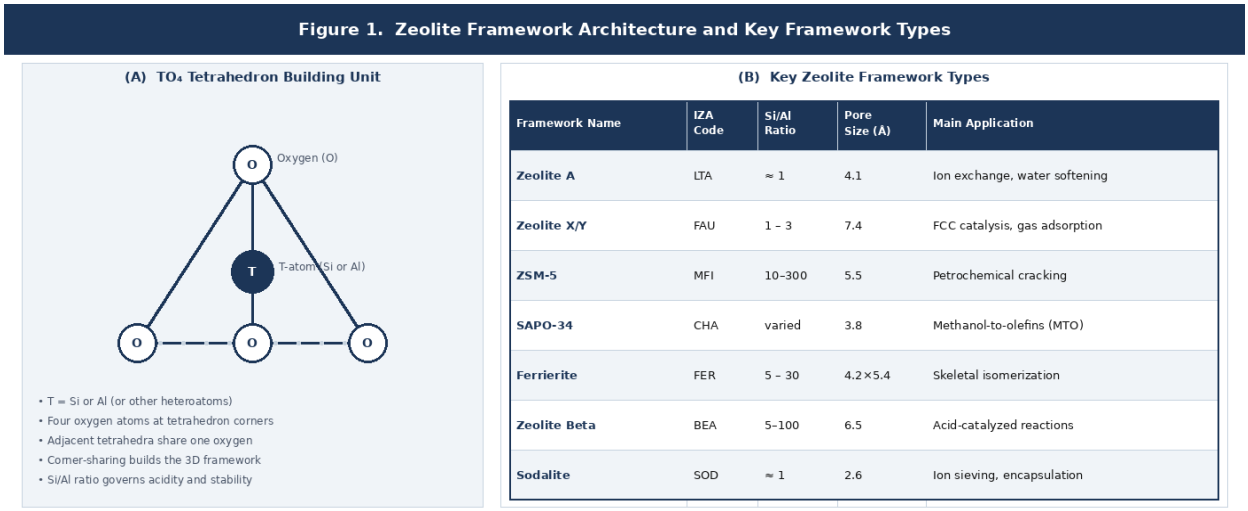


Figure 1. Zeolite framework architecture showing the $TO_4$ tetrahedron building unit and key zeolite framework types with their IZA codes, Si/Al ratios, pore sizes, and main applications

*Zeolite Synthesis Methods*

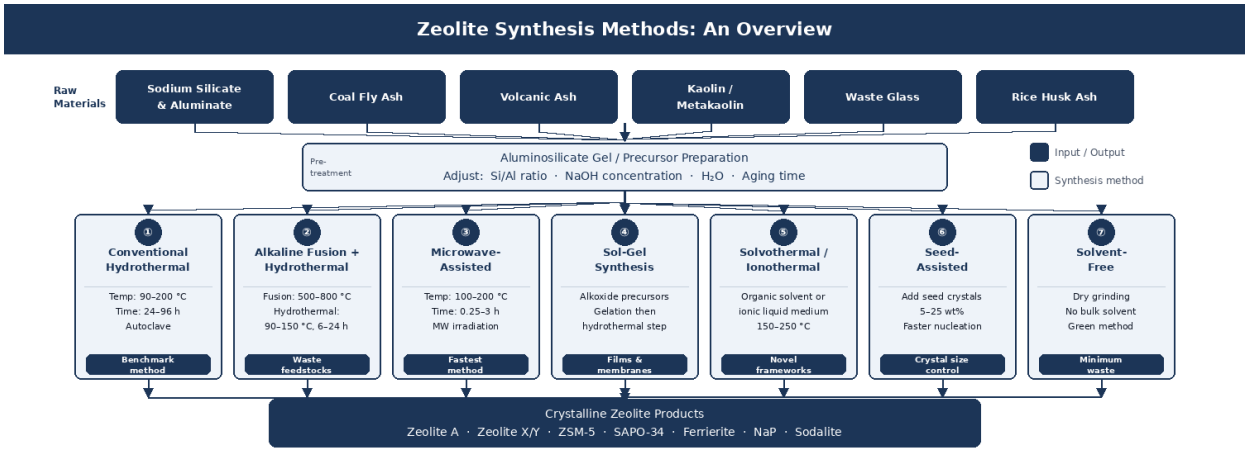


Figure 2. Overview of zeolite synthesis methods showing the progression from raw materials through gel preparation to seven principal synthesis routes and the resulting crystalline zeolite products. Key synthesis parameters are listed at the base of the figure.

## Results and Discussion

Conventional hydrothermal synthesis among all reported routes for producing synthetic zeolites, the hydrothermal method occupies a central position as the industrial and academic benchmark. In this approach, a reactive aluminosilicate gel assembled from silica and alumina precursors, an alkali hydroxide mineralizer, water, and optionally a structure-directing agent is sealed within an autoclave and subjected to elevated temperature and the resulting autogenous pressure. Crystallization temperatures generally span 90–200 °C and reaction durations range from a few hours to several days, depending on the target framework. Within this confined aqueous environment, water performs a triple function: it dissolves the amorphous gel constituents, transmits the hydraulic pressure that stabilizes the reaction vessel, and mediates the transport and condensation of silicate and aluminate species into ordered crystalline nuclei. The outcome of hydrothermal crystallization is exquisitely sensitive to the interplay of several compositional and process variables. Chief among these are the molar ratios of the gel components particularly $SiO_2/Al_2O_3$, $Na_2O/SiO_2$, and $H_2O/Na_2O$ together with the alkalinity of the reaction medium, the gel aging period prior to heating, and the chosen temperature duration profile.

As a general rule, aluminum rich gels with Si/Al ratios at or below five tend to crystallize into low silica frameworks such as zeolite A, zeolite X, and SAPO-based materials. Conversely, gels enriched in silica at ratios of five or above predominantly yield silica-rich topologies including ZSM-5, zeolite beta, and zeolite Y. When the NaOH concentration exceeds approximately 3 mol $L^{-1}$, the excess alkalinity disrupts the delicate nucleation balance, depressing crystallinity and promoting the formation of unwanted dense phases such as hydroxysodalite. Despite the reliability and wealth of optimization knowledge that characterize conventional hydrothermal synthesis, the method carries inherent limitations that restrict its appeal in a sustainability context. Crystallization duration of 24–96 hours are routine, and the pressurized autoclaves required for the process demand capital investment and impose safety constraints on scale-up. These drawbacks have directly motivated the investigation of the alternative routes discussed in the sections that follow.

*Alkaline fusion-assisted hydrothermal synthesis and alternative raw materials*

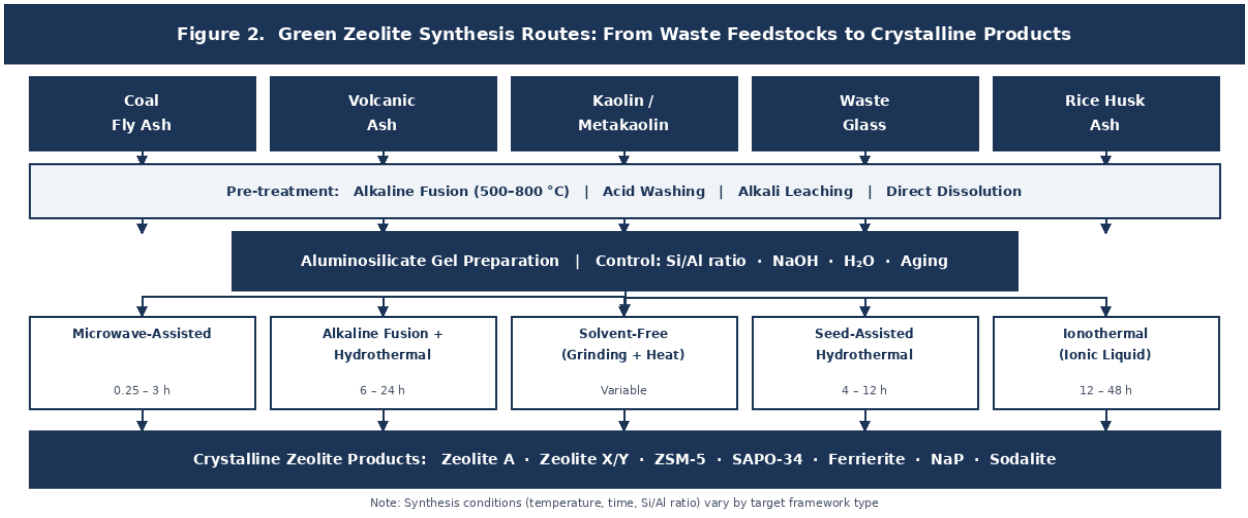


Figure 3. Green zeolite synthesis routes illustrating the progression from waste-derived feedstocks through pre-treatment and gel preparation to crystalline zeolite products via multiple energy-efficient pathways.

A major direction in zeolite research has been the substitution of conventional reagent grade silica and alumina sources with low-cost, often waste-derived, aluminosilicate materials. Coal fly ash (CFA), a by product of coal combustion in thermal power plants, is one of the most extensively studied alternative feedstocks owing to its high silica and alumina content and its large global production volume. Other reported sources include volcanic ash, kaolin and metakaolin, and waste glass from liquid crystal display panels or fluorescent tubes. Because these raw materials often contain low proportions of reactive silica and alumina, or are mineralogically inert, direct hydrothermal treatment alone is frequently insufficient to achieve high zeolite yields. The alkaline fusion-assisted hydrothermal method addresses this limitation through a two stages process: the raw material is first fused with solid NaOH at high temperature (typically 500–800 °C) to convert it into a reactive sodium aluminosilicate or sodium silicate/aluminate phase, which is then subjected to hydrothermal crystallization under conditions similar to conventional synthesis. This approach was first applied to the synthesis of zeolite X from coal fly ash and has since been extended to a range of framework types including zeolite A, zeolite P, and faujasite.

Compared with direct hydrothermal treatment, alkaline fusion typically produces zeolites with higher crystallinity, larger specific surface area, and greater cation exchange capacity, because fusion increases the proportion of amorphous, reactive aluminosilicate available for dissolution and crystal growth. However, the fusion step itself is energy intensive, and the overall process involves multiple stages of washing, aging, and crystallization, which increases process complexity relative to a single-step hydrothermal route. A related two step hydrothermal approach has also been described, in which Si- and Al-rich species extracted from the raw material during an initial hydrothermal step are recovered from the resulting filtrate and used to synthesize a second, higherpurity zeolite product, thereby improving overall raw material utilization. The use of volcanic ash as a silica source illustrates a further variant: high purity silica is first extracted by acid washing to remove calcite impurities, after which the purified silica is combined with a commercial aluminate source and subjected to fusion and hydrothermal crystallization, yielding zeolite Na-X, Na-P, or hydroxysodalite depending on the $SiO_2/Al_2O_3/Na_2O/H_2O$ molar ratios employed.

Sol-Gel Synthesis The sol-gel method involves the formation of a colloidal sol from molecular precursors (such as tetraethyl orthosilicate and aluminum alkoxides), followed by gelation and subsequent hydrothermal crystallization. This approach is particularly important for the preparation of zeolite coatings, thin films, and membranes on metallic or ceramic substrates, where it enables fine control over film thickness, adhesion, and crystal orientation. Zeolite synthesis methods overall can be classified into categories based on the use of templates, the type of silicon and aluminum precursors employed, the solvent system, and the strategies used to accelerate crystallization. Sol-gel routes fall within this classification primarily as a precursor based and solvent-mediated approach, and they are frequently combined with hydrothermal or microwave treatment to complete the crystallization step, as in the preparation of nanosized ZSM-5 from a template-free sol-gel precursor under microwave-assisted hydrothermal conditions. The principal advantage of the sol-gel route lies in the high homogeneity and purity of the resulting gel, which can translate into more uniform crystal size distributions; its main disadvantage is the relatively high cost of alkoxide precursors compared with mineral or waste-derived silica and alumina sources.

Microwave-Assisted Synthesis Microwave-assisted synthesis has emerged as one of the most effective process intensification strategies for zeolite preparation. Microwave irradiation couples directly with polar molecules and ions in the synthesis gel, generating volumetric and rapid heating that disrupts hydrogen bonding in water and produces highly active water species, thereby accelerating the dissolution of the amorphous gel and the nucleation of zeolite crystals. This results in crystallization times that can be reduced by an order of magnitude relative to

conventional hydrothermal heating; for example, the preparation time for a zeolite T membrane has been shortened from over 100 hours to approximately 9 hours under microwave assistance. Microwave heating has been successfully applied to the template-free synthesis of several zeolite frameworks. Rapid synthesis of zeolite NaP was achieved by crystallizing NaY gel under microwave irradiation without any organic template, with pure-phase NaP obtained after only one hour at 150 °C. Similarly, ferrierite zeolite has been synthesized within two to three hours through a microwave-assisted, organic template-free route using zeolite seeds, with the crystallization temperature and seed dosage identified as the key controlling variables; the resulting ferrierite exhibited catalytic performance comparable to material obtained by conventional hydrothermal synthesis.

Microwave-assisted methods have also been applied to waste-derived feedstocks. Microwave irradiation of South African coal fly ash in the presence of alkaline reagents was shown to provide a greener alternative to conventional thermal and fusion techniques, reducing both energy consumption and reaction time while still producing crystalline zeolite phases as confirmed by XRD and SEM analysis. More recently, a deep eutectic solventassisted seeding strategy combined with microwave hydrothermal crystallization was reported for the production of hierarchical micro-mesoporous Na-A zeolite, with total synthesis times of less than 60 minutes and the incorporation of recycled aluminosilicate feedstocks. Beyond direct synthesis, alternative heating technologies such as magnetic induction have also been explored as energy-efficient substitutes for conventional oil-bath heating in hydrothermal zeolite synthesis, offering higher heat-transfer rates relevant to potential scale-up of microwave- or induction-assisted processes.

Solvothermal Synthesis Solvothermal synthesis is a generalization of the hydrothermal method in which water is replaced, partially or entirely, by an organic solvent such as an alcohol, glycol, or amine; all hydrothermal methods can in principle be regarded as special cases of solvothermal synthesis, while not all solvothermal methods are hydrothermal. The replacement of water can alter the solubility of the gel components, the polarity of the reaction medium, and the interactions between the structure directing agent and the developing framework, enabling access to framework topologies or morphologies that are difficult to obtain under purely aqueous conditions. A notable example is the solvothermal synthesis of lithosite (−LIT)-type zeolite, in which low silica zeolite powders (such as zeolite Y, NH4-exchanged chabazite, and NH4 exchanged phillipsite) were dispersed in an alcohol containing dissolved KOH and treated solvothermally at 200–250 °C for 15–120 hours without stirring, yielding a single-phase monoclinic −LIT framework. Solvothermal conditions have likewise been applied to the synthesis of silicoaluminophosphate (SAPO) molecular sieves, where organic amines serve simultaneously as solvent and template; this aminothermal approach has been reported for SAPO-34 and related CHA-type materials used in methanol-to-olefins catalysis.

Ionothermal synthesis, in which an ionic liquid or a deep eutectic mixture serves as both solvent and structure-directing agent, was first reported for aluminophosphate and silicoaluminophosphate molecular sieves and offers the advantage of operating near atmospheric pressure, since the negligible vapor pressure of ionic liquids eliminates the autogenous pressure build up characteristic of aqueous hydrothermal systems. Microwave enhanced ionothermal synthesis has also been described for aluminophosphate molecular sieves, combining the rate acceleration benefits of microwave heating with the low-pressure operation afforded by ionic liquid media. Structure-Directing Agents and Crystallization Mechanisms Structure-directing agents (SDAs), commonly referred to as templates, are organic or inorganic species incorporated into the synthesis gel that direct the assembly of TO4 units into a specific framework topology. Common SDAs include alkali and alkaline-earth hydroxides, quaternary ammonium cations such as tetrapropylammonium hydroxide, organic amines, and more complex bulky cations derived from adamantamine or quinuclidinol. The role of an SDA extends beyond simple pore-filling:

detailed structural studies of ferrierite synthesized with pyrrolidine as the SDA, combined with X-ray Rietveld refinement and computational modeling, demonstrated that the template occupies welldefined positions within the framework cavities and channels, exerting a genuine structure-directing influence rather than acting merely as a space filler. The effectiveness of a given SDA in producing a target framework has been linked to the thermodynamics of its local solvation environment; calorimetric measurements of SDA dissolution enthalpy in water versus organic solvents have been used to rationalize why particular quaternary ammonium cations preferentially yield CHA-type SSZ-13 zeolite.

Beyond organic templates, inorganic anions present in the synthesis gel can also significantly influence crystallization kinetics. A systematic study of anion effects in the SiO2–TPAOH–H2O system for silicalite-1 synthesis found that the crystallization rate followed a Hofmeister-type ordering, with sulfate accelerating crystallization more strongly than chloride, bromide, iodide, or thiocyanate. Such findings illustrate that, in addition to the principal SDA, the broader ionic environment of the gel offers an additional, often underexploited, degree of freedom for controlling zeolite crystallization. A further phenomenon of mechanistic interest is the so-called “one structure/multiple templates” effect, in which a single zeolite framework, such as AlPO4-5, can be obtained using any of dozens of structurally distinct organic templates ranging from small amines to large diquaternary ammonium cations. Molecular dynamics simulations suggest that the template-framework binding free energy and the degree of charge transfer between template and framework are the primary determinants of which framework forms, rather than the precise molecular shape of the template alone.

Seed-assisted and solvent-free synthesis seeding, the addition of small quantities of pre formed zeolite crystals to the synthesis gel, is widely used to accelerate nucleation, shorten crystallization time, and control the final particle size of the product. In the microwave-assisted synthesis of ferrierite described above, seed crystals were found to dissolve only partially during crystallization, with new zeolite material growing epitaxially on the surface of the residual seed in a core shell mechanism; varying the seed particle size between approximately 0.4 and 3.0 μm allowed corresponding control over the final crystal size. Solvent-free synthesis represents a further development in green zeolite chemistry, in which the aluminosilicate gel, template, and any required metal precursors are combined and crystallized through mechanical mixing, grinding, and heating, without the addition of bulk water or organic solvent. This approach is especially advantageous for the synthesis of heteroatom-substituted zeolites, since conventional post synthetic ion-exchange routes for introducing metal cations such as copper require multiple aqueous exchange steps and generate substantial volumes of wastewater, whereas a one-pot solvent-free route can incorporate metal-amine complexes directly during crystallization. Solvent-free methods have also been used to prepare hierarchically porous SAPO-34 with enhanced selectivity toward light olefins in methanol-to-olefins catalysis.

Comparative Assessment of Synthesis Methods Across the methods reviewed, several consistent trends emerge. Conventional hydrothermal synthesis remains the benchmark against which alternatives are evaluated, offering well-characterized control over framework type through gel composition but requiring long crystallization times and significant energy input for sustained heating under autogenous pressure. Alkaline fusion-assisted hydrothermal synthesis substantially improves the utilization of low-cost, waste-derived silica and alumina sources and generally yields products of higher crystallinity than direct hydrothermal treatment of the same raw material, at the cost of an additional high-temperature fusion step.

Table 1. Comparison of major zeolite synthesis methods

| Method | Typical conditions | Common raw materials | Advantages | Limitations | Key references |
|---|---|---|---|---|---|
| Conventional hydrothermal | 90–200 °C, autogenous pressure, 24–96 h | Sodium silicate/aluminate, colloidal silica, alumina | Well understood; broad framework control via gel composition | Long crystallization time; high energy use; sealed autoclaves | (Zhang et al., 2020) |
| Alkaline fusion-assisted hydrothermal | Fusion 500–800 °C, then hydrothermal 90–150 °C, 6–24 h | Coal fly ash, volcanic ash, waste glass, kaolin | Higher crystallinity and CEC from low-cost/waste feedstock | Energy-intensive fusion step; multi-stage washing/aging | (Otieno et al., 2023) |
| Sol-gel | Gelation of alkoxide precursors, then hydrothermal or microwave crystallization | Tetraethyl orthosilicate, aluminum alkoxides | High gel homogeneity/purity; good for films and membranes | Costly precursors; limited to small-scale/coating applications | (Rebrov, 2009) |
| Microwave-assisted | 100–200 °C, 0.25–3 h under MW irradiation | Standard gels, NaY gel, fly ash, deep-eutectic-solvent systems | Drastically shorter crystallization time; energy efficient; template-free options | Equipment cost; scale-up challenges for uniform irradiation | (Patel & Pereira, 2022); (Ungureanu et al., 2025) |
| Solvothermal | 150–250 °C, organic solvent or ionic liquid medium, 15–120 h | Low-silica zeolite powders, SAPO/AlPO precursors, ionic liquids | Access to novel frameworks/morphologies; near-atmospheric pressure (ionothermal) | Solvent cost/recovery; mostly lab-scale; long reaction times | (Mao et al., 2017); (Tchessalov et al., 2022) |
| Seed-assisted | Conventional or MW hydrothermal with 5–25 wt% seed crystals added | Pre-formed zeolite seed crystals plus standard gel | Faster nucleation; controllable particle/crystal size | Requires pre-synthesized seed material; added | (Zhao et al., 2022); (Harada et al., 2014) |

| Method | Typical conditions | Common raw materials | Advantages | Limitations | Key references |
|---|---|---|---|---|---|
| | | | | process step | |
| Solvent-free / solid-state | Mechanical mixing/grinding then heating, no bulk water or solvent | Aluminosilicate gel/powder, templates, metal-amine complexes | Minimal wastewater; suitable for one pot heteroatom incorporation | Limited framework scope; potential heterogeneity at larger scale | (Sousa et al., 2025); (Frick et al., 2024) |

As summarized in Table 1, the methods differ primarily along the dimensions of energy and time input, raw material flexibility, and scale-up readiness. This comparison highlights why hybrid combinations, such as fusion-microwave or seeded solvent-free routes, are increasingly favored, since they allow the strengths of one method (e.g., low-cost feedstock utilization) to offset the limitations of another (e.g., long crystallization time). Microwave-assisted synthesis offers the most consistent reductions in processing time, in several cases lowering crystallization times from tens of hours to under one hour while maintaining or improving product crystallinity, and is increasingly applied in template-free and waste-feedstock systems (Da Silva Moreira et al., 2024); (Li et al., 2023). Solvothermal and ionothermal methods extend the accessible chemical space for framework types and operating pressures but generally remain at a more exploratory, laboratory scale compared with hydrothermal and fusion-based routes (Takako et al., 2012); (Eren et al., 2024). Seed-assisted and solvent-free approaches contribute complementary benefits in terms of crystal size control and waste minimization, respectively, and are readily combined with the other methods discussed (Ren et al., 2020); (Mei et al., 2021). Taken together, the literature suggests that no single method is universally optimal; rather, the choice of synthesis route should be guided by the target framework type, the available raw materials, and the relative priorities placed on product quality, processing time, energy consumption, and environmental impact (Mohammadi-Jam et al., 2026); (Parvulescu & Maurer, 2022). Hybrid approaches that combine waste-derived feedstocks with microwave or induction heating and template-free formulations appear particularly well positioned to meet the dual goals of cost reduction and sustainability in future zeolite production (Ferdov, 2026); (Tontisirin et al., 2022).

## Conclusion

This review demonstrates that green and sustainable zeolite synthesis is no longer an aspirational concept but an increasingly practical reality, supported by a substantial and rapidly growing body of experimental evidence. Two principal strategies, the use of waste-derived aluminosilicate feedstocks and the adoption of energy-efficient synthesis routes have individually shown considerable promise, and their combination represents the most compelling direction for future zeolite manufacturing. Coal fly ash, volcanic ash, kaolin, and waste glass have each been demonstrated as viable and cost-effective substitutes for reagent grade silica and alumina sources, particularly when combined with alkaline fusion pre-treatment to enhance the reactivity of otherwise inert raw materials. Microwave-assisted synthesis consistently delivers the most dramatic reductions in crystallization time and energy input, with total synthesis times in some cases reduced from days to under one hour without sacrificing product crystallinity or purity.

Solvent-free and seed-assisted methods contribute further gains in waste minimization and process control, and are readily combined with both waste feedstock utilization and microwave heating. Taken together, the evidence reviewed here supports the conclusion that hybrid approaches particularly fusion-microwave and microwave-seeded routes using industrial waste precursors offer the best available balance between sustainability, product quality, and scalability. Key priorities for future research include the scale-up of microwave-assisted processes beyond laboratory reactors, the development of template-free routes for a wider range of commercially important framework types, and the systematic life-cycle assessment of green synthesis routes to quantify their environmental and economic advantages over conventional methods.

## Conflicts of interest

There were no conflicts of interest

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## Author contributions

P. W: Conceptualization, Methodology, Software, Data curation, Writing-original draft preparation.: P. M: Visualization, Investigation, Supervision, Software, Validation, Writing-reviewing, and editing. All authors have read and agreed to the published version of the manuscript.

## Declaration of generative AI in scientific writing

The authors acknowledge the use of generative artificial intelligence (AI) tools exclusively for language editing, grammar refinement, and enhancing the academic phrasing of this manuscript. The AI assistance was utilized solely to polish the text and improve readability. The authors reviewed, edited, and verified all final content to ensure accuracy and take full responsibility for the integrity of the research.

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