

Perspective

# A Short Review on the Utilization of Incense Sticks Ash as an Emerging and Overlooked Material for the Synthesis of Zeolites

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**Abstract:** The traditional hydrothermal synthesis methods are mainly performed under batch operation, which generally takes few days to weeks to yield a zeolite with the desired properties and structure. The zeolites are the backbone of the petrochemical and wastewater industries due to their importance. The commercial methods for zeolite synthesis are expensive, laborious and energy intensive. Among waste products, incense sticks ash is a compound of aluminosilicates and could act as a potential candidate for the synthesis of zeolites for daily needs in these industries. Incense sticks ash is the byproduct of religious places and houses and is rich in Ca, Mg, Al and Si. As a result, incense sticks ash can be proven to be a potential candidate for the formation of calcium-rich zeolites. The formation of zeolites from incense sticks ash is an economical, reliable and eco-friendly method. The application of incense sticks ash for zeolite synthesis can also minimize the problem related to its disposal in the water bodies, which will also minimize the solid waste in countries where it is considered sacred and generated in tons every day.

**Keywords:** incense sticks ash; zeolites; wastewater treatment; aluminosilicates; tetrahedron

## 1. Introduction

In the last decades the demand for zeolites has increased drastically due to their emerging applications in the petroleum industries, catalyst industries and water-softeners-based industries. Commercially, zeolites are produced from various chemical precursor which makes them quite expensive. Therefore, there is need for some reliable and economical source material such as coal fly ash, red much, kaolin, etc., which could act as a precursor

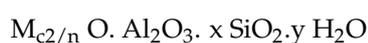
material for zeolites. One such overlooked waste material is incense sticks ash (ISA), which are produced during the combustion of incense sticks in houses or at religious places. ISA is rich in alkali metals like Ca, Mg, carbon, Si and Al. In addition, it has numerous elements in intermediate to trace levels. The basic requirement for a precursor material to be suitable for zeolite synthesis is high Si and Al content along with certain alkali metals [1]. There are various industrial, biological and domestic wastes such as coal fly ash, bauxite, red mud and incense sticks ash (ISA), which have an optimum amount of Si and Al [2,3]. The literature is flooded with research and review work on zeolites synthesis from all the above-mentioned waste products except ISA. ISA is produced at religious places, after the complete burning of incense sticks. These are mainly used in South Asian countries, the Middle east, etc., but in India, it is mainly disposed of into rivers and other water bodies due to their sacred value. Therefore, ISA could act as a potential candidate for the synthesis of zeolites as few works have reported its chemical composition unlike CFA and kaolin or red mud. However, it has been most underestimated in the scientific community, limiting its application to our daily life [1–16].

Zeolite is a porous material with a large surface area and cavities arranged in a basket-like frame [14,17], which are hydrates of aluminosilicates (interlinked tetrahedra of alumina and silica) and bear negative charges on them. They have a three-dimensional structure made up of Al, O, Si and other alkaline earth metals (Na, K and Mg) and H<sub>2</sub>O molecules. The water molecules in these zeolites are confined in the gaps of the elements [18,19]. Zeolites have a tendency to exchange other cations for the metal ions originally trapped inside them (better known as cation exchange) [20]. Zeolites have fixed-size regular openings or pores, which enable larger molecules to pass through but trap the smaller molecules, hence they are often also called molecular sieves [21]. The percentage of Si and Al varies from one kind of zeolite to another. Some have more numbers of Si atoms compared to Al present in their structure, while others have more Al atoms present relative to the number of Si atoms present ensuring their interim ratios is always greater than one. However, all of them are predominantly made up of alumino-silicates complexes [22]. The Si/Al ratio is the most important parameter in a zeolite structure as it determines the cation exchanged properties, acid properties, hydrophilic/hydrophobic properties, catalytic properties and many others. According to Löwenstein's empirical rule, when two tetrahedra are connected in a structure, there cannot be two aluminum tetrahedra next to each other. In zeolite structures, only Si-O-Si or Al-O-Si bridges can exist, but no Al-O-Al. In this way, the range of the Si/Al ratio can be from one (where the amount of Si and Al in the structure is equal) to infinity (pure silica samples) [23]. Zeolites rich in Al are attracted towards polar molecules (H<sub>2</sub>O) such as water, while zeolites rich in Si are attracted towards nonpolar molecules [24]. Due to all these above-mentioned features, zeolites have been widely used in wastewater treatment as an adsorbent [25], in agriculture as fertilizers [26] and as ion exchangers [27–29].

The aim of this review work is to suggest emerging applications of ISA, towards the metallurgical and recovery of value-added minerals. This work will draw the attention of the global scientific community towards the most overlooked material. Here we have reported recent work carried out in the field of ISA and zeolite synthesis from it. This could prove to be valuable information towards the reduction of solid waste and ultimately water pollution arising from the disposal of ISA into water sources.

## 2. Structural Properties of Zeolites

In zeolite structure, few of the quadrivalent Si cations are substituted by triply charged Al cations, which causes a deficiency of positive charges [30]. These developed charges are balanced either by the single or double-charged cations, such as Na, K, Ca and Mg<sup>2+</sup>, anywhere in the structure, leading to spacious pores or rings [31]. The most common formula of a zeolite is:



where  $M_c$  is any alkali and alkaline earth atom,  $n$  is a charge on the atom,  $x$  is the number of Si tetrahedra in the structure, which varies from 2 to 10, and  $y$  is the number of water molecules that varies from two to seven. Si and Al atoms combine in such a way that they form a structural framework in the zeolites, with centrally placed Si and Al atoms, while O is present in the corners [32,33]. The O atoms are common between the  $[\text{SiO}_4]_{4-}$  and  $[\text{AlO}_4]_{5-}$  tetrahedra, which remain oriented in such a way that the frameworks develop voids or pores in the form of cages and channels between the tetrahedra [34,35].

### 3. Types of Zeolites

Broadly, zeolites fall into two types based on their origin, i.e., natural or synthetic [36]. Zeolites are present in nature and are mined in various parts of the world, but they are impure and less organized, so for smother work they have been synthesized commercially. Natural zeolites are formed in volcanic ashes, sedimentary rocks [37], clay-kaolins [38], red mud [39], coal fly ash (CFA) [6], bauxite minerals [1,40] and other earthy materials [41]. On the other hand, synthetic zeolites are synthesized in laboratory conditions by optimizing the conditions of various elements [42]. All of these have unique structures, properties and catalytic functions.

#### 3.1. Natural Zeolites

Zeolites are basically of two types natural and synthetic, where in nature they are present in minerals, fly ash, rocks and volcanic ash. Currently there are about 50 naturally occurring zeolites out of which chabazite, clinoptilolites and mordenite are the three most mined forms of natural zeolites and they vary in physical and chemical properties [43]. Particle density, cation selectivity, molecular pore size and strength are only some of the properties that can differ depending on the type of zeolites [44,45]. These natural zeolites occur in random forms and mixed sizes. In nature, they are present in the form of crystals in small cavities of basaltic rocks, and several geological environments such as alkaline deserts, lake sediments, marine sediments and ash ponds at lower temperatures [46]. Such zeolites, due to their unique structure, are filled up with water molecules and show honeycomb-like structures when losing water upon heating [47]. The best example is chabazite, which are the most commonly used natural zeolites [48]. The clinoptilolites zeolites have been used in agriculture, fertilizer, soil amendments and feed additives due to their higher acid-resistant Si content. However, their impurity, i.e., the contamination by other minerals, limits their wider applications in industries where accuracy and uniformity are required [49]. The most common general formula for natural zeolites is:



$p$  = number of monovalent metal ions;

$q$  = number of divalent metal ions;

$n$  = half of the number of oxygen atoms;

$m_0$  = number of water molecules.

#### 3.2. Synthetic Zeolites

Synthetic zeolites are synthesized in laboratories according to the need of industries and are considered a backbone of these industries. They are used as a catalyst, adsorbents for wastewater treatment, removal of pollutants, etc. Currently, around 150 different synthetic zeolites are synthesized throughout the globe, which in total accounts for around 3 million tons of zeolites produced annually [50]. These have been synthesized and designed according to their needs for specific purposes. Synthesized zeolites are more uniform and purer than natural zeolites in terms of their lattice structures, pore sizes and the cages in their framework [51,52]. In comparison to natural zeolites, synthetic zeolites are synthesized in very precise and uniform sizes to suit a particular application especially to trap certain smaller molecules inside them [53,54]. Among all the synthesized zeolites, zeolite A is the best known and is widely used as a laundry detergent. Synthetic

zeolites like X and Y (two different types of faujasites, used for catalytic cracking) [55], and the petroleum catalyst ZSM-5 [56] are also widely synthesized. Synthesized zeolites are unreactive and made up of minerals that are mainly present in nature and are therefore not considered to have any harmful effects on the environmental [57]. Synthetic zeolites have more accurate features, which increase their wider applications in industries compared to natural zeolites. Such types of CFA-based synthetic zeolites are A, X, Y, P and Na-P1. The Si and Al composition of CFA determines their types, which can be classified into four groups. However, until now, among waste products, all the zeolites have been synthesized from CFA [6,58], kaolin [40,59] and red mud [39].

Recently, Yadav et al. (2021) reported the synthesis of zeolites (sodalite) from CFA waste. Their synthesis parameters were almost the same, i.e., initially, ferrous particles were removed from the slurry by using an external magnet, as the presence of iron could interfere with the properties of the final zeolite product. The obtained ferrous free particles were treated with 8 M NaOH for 16 h, along with continuous stirring at a temperature of 95 °C under reflux conditions. The analysis by the instruments revealed that the synthesized particles were rod-spherical-shaped of sizes 60–200 nm, while few were also in microns [6].

#### 4. Incense Sticks Ash (ISA) as an Emerging Source of Zeolites

ISA are one of the major household wastes in many countries like China, Thailand, Japan and India [60]. In all these countries, every day, tons of incense sticks are used in both houses and religious places. In a country like India, incense sticks ash is considered sacred and disposed of in rivers. Indian incense sticks ash has high Ca, Mg, Fe Si and Al, out of which Ca and Mg alone constitute 50–55% of the total weight of ISA [61]. These two elements are also responsible for the hardening of the river water, therefore, instead of disposing of these ashes into the river, it could be used for the recovery of valuable minerals. However, to date, no attempt has been made to synthesize zeolites from waste such as incense sticks ash.

The chemical composition of ISA makes it a potential material for the synthesis of zeolites using the hydrothermal method, which involves the treatment of ISA with NaOH. Table 1 shows the major elemental composition of ISA collected from temples in India, though the composition of ISA may further vary based on trade secret components, ingredients and burning conditions.

**Table 1.** Major elemental oxides of ISA.

Major Elements	wt %
Calcium oxides	49.61
SiO <sub>2</sub>	20.36
Alumina	4.77
Ferrous	4.28
K <sub>2</sub> O	8.24
MgO	3.91
Si/Al	4.26

#### *Hurdles in Using ISA as a Precursor Material for Zeolite*

Yadav et al. (2021) reported that the ISA typically collected from Indian temples and also generated after calcination of incense sticks powder in a muffle furnace, contains several toxic heavy metals such as Cu, Cr, Ni, Co, As, Hg, Zn (shown in Table 2) and Mo, which could restrict their application in zeolites where even traces of heavy metals are not required [62]. If ISA is used without prior treatment for the synthesis of zeolites, then there is a possibility that the toxic heavy metals may leach out when it is used in agriculture for biofertilizers and it may lead to heavy metal toxicity in the plants and soil. However, it could be applied without prior treatment for environmental purposes, for instance for the removal of dye, effluents, pesticides, etc. Recently, Jain et al. have used ISA directly for the removal of Victoria blue dye from aqueous solutions using a batch adsorption study and

obtained up to 80–90% efficiency. The whole process was reported to be economical and eco-friendly [63]. In addition, this ISA also had a high amount of unburned carbon (31% in ISA burned on an electric heater under controlled conditions) in the form of soot and char, which has to be removed prior to its use for the synthesis of zeolites [62]. As a result, the carbon particles could be eliminated by using the froth flotation method that is widely used for the recovery of carbon particles from coal fly ash.

**Table 2.** Heavy metals in ISA.

Heavy Metals	mg/L
Cd	0.002
Co	0.305
Cr	1.771
Cu	3.602
Ni	1.284
Pb	0.156

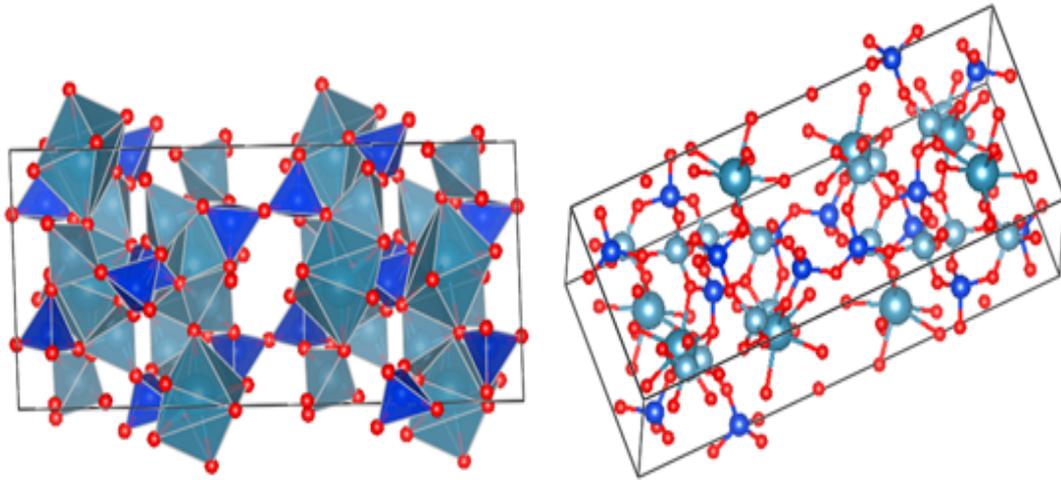
However, in comparison to CFA, ISA has many times less amount of toxic heavy metals. Therefore, the presence of such heavy metals should not be a significant hindering factor for the synthesis of zeolites [62].

### 5. Synthesis of Zeolites from ISA

To date, hardly any information is available on the formation of zeolites from ISA. From the above sections, we know that the chemical composition of ISA is very similar to that of CFA; they only differ in their percentage of elements, i.e., ISA has more Ca and Mg and less Si and Al, whereas CFA has more Si and Al and less Ca. The amounts of heavy metals are also similar, differing only in concentration. CFA has a higher concentration of heavy metals as this concentration depends on the coal. On the other hand, as the incense sticks use a small amount of coal powder as a facilitating agent for the burning of the sticks, heavy metals are in small amounts. As a result, the chemical approach for the synthesis of zeolites from ISA would be more or less similar to CFA. To date, zeolites have been synthesized from CFA by using microwave-assisted techniques, hydrothermal method and many others. Therefore, the most widely used and easiest approach is a hydrothermal method where Si/Al-rich material is treated with a strong alkali such as 4–16 M NaOH/KOH at 80–110 °C for 6–24 h along with continuous stirring. These operating parameters will produce morphologically different classes of zeolites. Since ISA is rich in Ca and Mg, there is the possibility that the synthesized zeolites would be rich in Ca and Mg, which are required by some of the zeolites. As alkali-metals-based cations favor the formation of a certain class of zeolites, there is a possibility of forming chabazite [64] and gismondine class of zeolites [65]. The peculiarity of such zeolites is that they are rich in Ca and Mg. Only one attempt has been made for the synthesis of zeolites from ISA, by Yadav et al. in 2020. In this patent, they have reported the synthesis of calcium-rich zeolites by using ISA.

ISA can be washed several times to remove the impurities, such as unburnt carbon, soot and other particles. Furthermore, by using a magnet, ferrous particles could be extracted, which generally interfere with the properties of zeolites. The removal of ferrous particles from the ash enhances the zeolitic property, as they interfere with the transformation of zeolites. The nonferrous fractions can be dried in an oven to further use as a precursor. Furthermore, the dried incense sticks ash can be treated with 1–2 N H<sub>2</sub>SO<sub>4</sub> by maintaining a solid-to-liquid ratio of 1:10, at room temperature, along with a continuous stirring on a magnetic stirrer with a reflux condenser. This removes the excess of Ca in the form of chlorides from the ash. The residue can be obtained by centrifugation at above 5000 × g. The residue can be collected and dried, which can be further treated with 4–8 M NaOH by keeping an appropriate solid-to-liquid ratio in a round bottom flask at a temperature of 90–95 °C, along with continuous stirring for 60–90 min in a reflux condenser. The residue can be collected by centrifuging the mixture at 5000 × g for 10 min. The residue

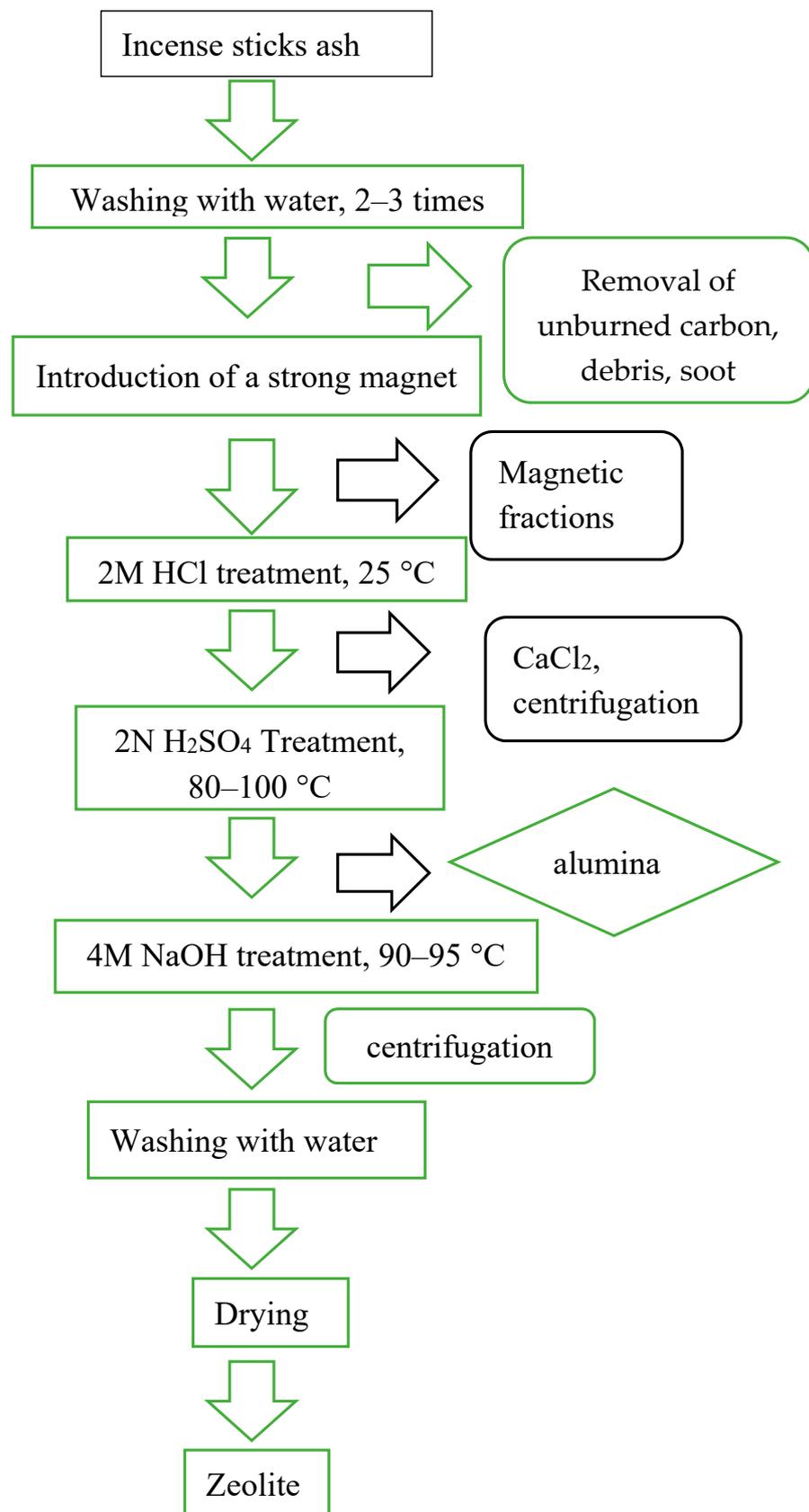
can be retained while the supernatant can be discarded. Furthermore, the final residue can be washed with DDW 2–3 times to eliminate any NaOH or NaCl particles from the surface. The final residue can be dried in an oven at 40–60 °C till complete dryness [66]. After analysis, it was found that the synthesized zeolites belonged to the gismondine class, whose formula is  $(\text{Ca}_2\text{Al}_4\text{Si}_4\text{O}_{16}\cdot 9\text{H}_2\text{O})$  and whose structure is shown in Figure 1.



**Figure 1.** The crystal structure of gismondine ( $\text{Ca}_2\text{Al}_4\text{Si}_4\text{O}_{16}\cdot 9\text{H}_2\text{O}$ ).

In one more approach, Yadav et al. (2021, unpublished) reported the synthesis of zeolites from ISA by a similar approach. However, in the case of ISA, the ferrous composition is very low 2–5%, so the removal of ferrous particles step is an optional one as it would not affect the purity of the zeolite to a great extent. The zeolite was synthesized using the hydrothermal method by treating the ISA with 8 M NaOH at 95 °C for 90 min, along with continuous stirring. After completion of the reaction, the mixture was cooled at room temperature and centrifuged at  $7000\times g$  for 5 min. The Si-rich supernatant was discarded while the zeolitic residue was retained and washed with distilled water several times. Washing ensured the removal of any alkali moieties from the surface of zeolites. After characterization of the zeolites, it was found that the particles were calcium-rich, cuboidal in shape and whose size varied from 0.2 microns to several microns. EDS also confirmed the high amount of calcium in the samples, because the ISA is rich in calcium and magnesium, which tend to remain associated with the aluminosilicates very firmly. Based on the above two approaches, we can derive the steps involved in the synthesis of zeolites from ISA, which is shown below in Figure 2.

In addition, there are also several other methods for the synthesis of zeolites from waste materials like red mud, fly ash and bauxite, etc. The major similarity among all these methods is the parameters, which should be maintained throughout the synthesis in order to obtain a zeolite with the desired properties. The variations in pH, temperature, stirring time, etc., will further govern the shape, size and class of zeolites that will determine their future applications and that can be confirmed by analytical instruments.



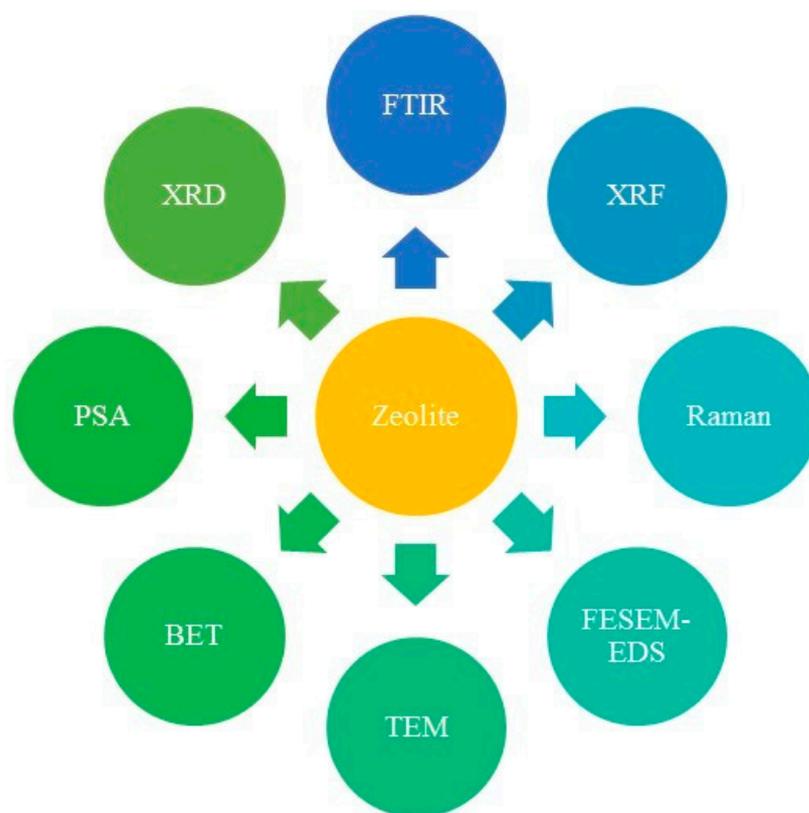
**Figure 2.** Schematic steps engaged in the formation of zeolites from incense sticks ash (ISA) [66] [Patent no 202011036844 A].

## 6. Major Characterization Techniques for Identification of Zeolites

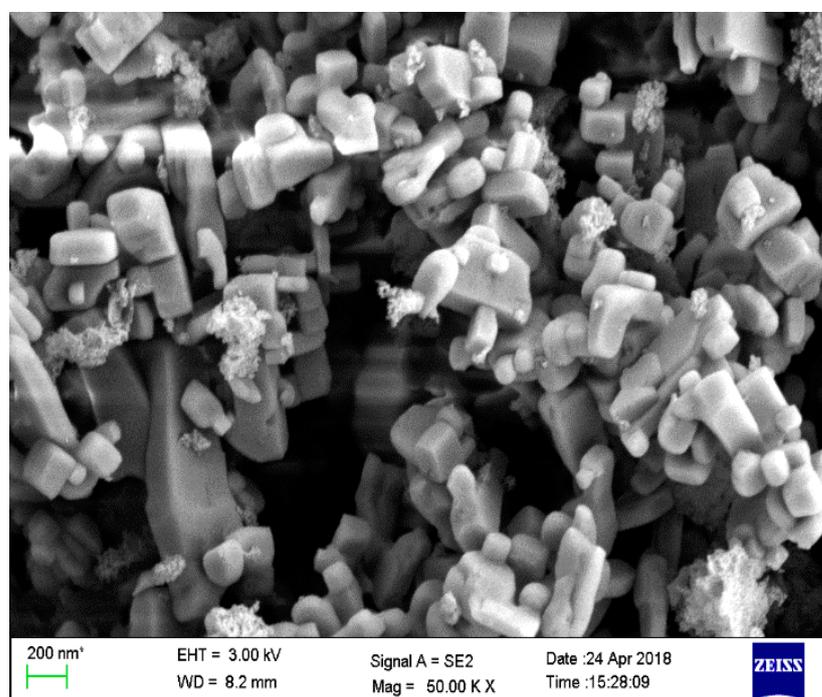
When it comes to zeolites, characterization becomes an important step, as the morphological properties and class of zeolites could be determined based only on the characterization. Microscopic analysis helps reveal the shape and size of the zeolites, as there are certain reactions in industries where both parameters become very important. As a result, scanning electron microscopy, transmission electron microscopy and atomic force microscopy could prove to be very reliable. Some of the zeolites are shape-specific, which helps in their categorization. Infrared spectroscopy and Raman spectroscopy help reveal the various functional groups present on the surface of synthesized zeolites. Zeolites have OH groups in their structure, which determine their hydration nature. Therefore, this analytical technique becomes a basic tool for the identification and confirmation of zeolites. The OH group could be detected by the presence of broadband peaks near 3200 to 3600  $\text{cm}^{-1}$  and, due to water molecules, near 1400–1700  $\text{cm}^{-1}$ , which must be present in any class of zeolites, until they get dehydrated. The formation of new bonds in the range of 400–2000  $\text{cm}^{-1}$  indicates the synthesis of zeolites. Further, FTIR could help in the identification of single-bond or double-bond carbon present in the sample. Both the instruments are useful for the characterization of zeolites from incense sticks ash. The chemical composition of synthesized zeolites become of utmost importance as it helps in the classification of zeolites into different classes. For instance, the percentage of Si, Al and cations all will determine the class of zeolites. Major oxides of zeolites could be detected by X-ray fluorescence (XRF), which helps in classifying zeolites either in low silica-containing, intermediate or high silica-containing zeolites. Since the Si/Al ratio is an important factor in deciding the class of zeolites, the contribution of XRF cannot be underestimated in the confirmation and identification of zeolites. Energy dispersive spectroscopy (EDS) also helps in the identification of the chemical composition but is restricted to only intermediate elements as it cannot detect trace elements. Moreover, unlike XRF it needs only a pinch of sample for the chemical composition. Sometimes it becomes of utmost importance to know the trace elements of zeolites when they are used for highly specific purposes; in that case atomic absorption spectroscopy becomes very important. However, it does not have any other role in the identification and confirmation of the zeolites.

Another X-ray-based technique is the X-ray diffraction technique, which helps in the identification of various mineral phases of the synthesized zeolites. In addition, it also helps characterize the amorphous or crystalline nature of the zeolites. There are certain zeolites that cannot be identified by any of the above techniques, therefore, XRD plays an important role. Sodalities, Na-X zeolites, etc., could be identified with the help of peaks obtained by XRD. When it comes to zeolites, porosity is an important factor that determines their further application since the porosity and porous nature of zeolites decide their water holding or any other liquid holding capacity. The pore size and surface area of zeolite particles could be obtained by using a surface area analyzer or Brunauer–Emmett–Teller (BET) analyzer. Some authors have also reported the importance of SS-nuclear magnetic resonance (NMR) in the identification of class and conformation of shape of the synthesized zeolites [67].

Furthermore, several other techniques that may further help the identification and classification of the synthesized zeolites with more ease and comfort, are shown below in Figure 3. Yadav et al. reported the synthesis of cuboidal-shaped zeolites from ISA of size 80–180 nm by FESEM, which is shown in Figure 4.



**Figure 3.** Different characterization techniques for zeolites.



**Figure 4.** FESEM micrograph of zeolite particles synthesized from ISA [66] [Patent no 202011036844 A].

## 7. Conventional and Emerging Applications of Zeolites

Zeolites are mainly used for wastewater treatment, environmental cleanup [68], etc. As zeolites are porous in nature [69] they are the most preferred candidates for the remediation of heavy metals, dyes, organic and inorganic toxic pollutants and pesticides from the

contaminated water [70]. If zeolites are synthesized from waste materials such as incense sticks ash, fly ash or red mud, then the synthesis of zeolites becomes cheaper and eco-friendly [19]. When these zeolite particles are used for wastewater treatment, the removal cost will be much less as the starting material is a waste product. They are frequently used as molecular sieves in chemistry laboratories for research. They are also used in the industries for catalyzing petrochemical products. There are only a few reports for ISA-based zeolite applications, due to the lack of work done by investigators in this field. Earlier, investigators have used incense sticks ash directly (without any processing) for the remediation of Victoria blue dye from the wastewater under laboratory conditions and they obtained quite satisfactory results. Therefore, the potential of ISA has been known as an economical adsorbent for the remediation of pollutants from the wastewater. However, that research work did not provide information about the potential of ISA-synthesized value-added material for remediation purposes. Furthermore, Yadav et al. (2021) used ISA-based calcium-rich zeolite for the remediation of heavy metals (Cu, Co, Ni, Pb, Cr, Zn and Cd) and alkali metals (Ca, Mg, Ba and Al) from simulated wastewater from fly ash. There was a significant decrease in the concentration of all the heavy metals and alkali metals after two hours by using 9 mg of zeolites in 150 mL of 20% fly ash aqueous solution.

From both the above-reported works, it was found that both ISA and zeolites derived from them have the potential to act as an efficient and economical adsorbent. As a result, the utilization of ISA or zeolites synthesized from them can act as an alternative adsorbent for the remediation of dyes, heavy metals and alkali metals. The utilization of ISA waste as an emerging material in the field of adsorbents can minimize the burden on the current zeolite industry. Moreover, the utilization of overlooked waste as a value-added mineral will help minimize the solid waste arising from ISA. The suggested utilization of ISA could save a huge amount of the money used for the removal of the hardness of the water. Moreover, it will also create awareness among the devotees involved in the disposal of ISA into the rivers.

## 8. Conclusions

Zeolites have become the basic demand for all the petroleum, chemical and water softening industries. To meet the demand, the load on the existing industries has increased and the material cost has become expensive due to expensive precursor materials. Incense sticks ash, being rich in calcium, silica and alumina, could open new horizons in the field of synthesis of zeolites for our daily needs at a much economical cost. The unawareness and lack of research in the field regarding the utilization of incense sticks ash is a major hurdle. The changing trends in the field of overlooked incense sticks have drawn the attention to ISA for the recovery of value-added materials. The major factor will be controlling the parameters, which contribute towards desirable and eco-friendly zeolites. In the future, it could be utilized as a carrier for biofertilizers to the plants, which would minimize the toxic effect of pesticides and insecticides on the soil and ultimately on human beings. The use of incense stick ash as a zeolite could prove an alternative and low-cost material for the zeolites-based industry. Moreover, incense sticks ash-based recovery of zeolites will minimize the solid waste which is being dumped in the rivers and other water bodies.

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