

CHAPTER 12

Dual functional nano zeolites for CO₂ capture and conversion

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12.1 Introduction

As reported by the Intergovernmental Panel on Climate Change (IPCC), carbon dioxide (CO₂) in the atmosphere will reach 570 ppmv by the end of 21st century. This will increase average surface temperatures between 2°C and 6°C and mean sea level by 38 cm (Audus et al., 1995; Hasib-ur-Rahman et al., 2010; Stewart & Hessami, 2005). Massive CO₂ emissions come from anthropogenic activities like fossil fuel combustion for power generation, industrial manufacturing, and transportation (Heintz et al., 2008). Due to the increasing percentage of CO₂ emissions and the rising demand of energy, sustainable energy development (Carmo et al., 2013; Ursua et al., 2012), and CO₂ capture, storage, and consumption (Lackner, 2003) are two resourceful methods to battle the greenhouse gas emissions linked to climate change.

For a decade, carbon capture and sequestration (CCS) has been broadly reported as a means to mitigate the emissions of CO₂, employing systems based on amine solvents, ionic liquids, solid sorbents, advanced membrane, and porous materials (Alami et al., 2020; Yu et al., 2008). CCS facilitates CO₂ to be concentrated, liquefied, and stored to reduce the emissions from large-scale industrial processes. This strategy has a great potential to reduce CO₂ emissions up to 90% from conventional power plants (Yu et al., 2008). CCS systems are usually classified as precombustion, oxy-fuel combustion, and postcombustion, based on the location of emissions for the capture system (Alami et al., 2020). Major advances have been achieved toward the design of different adsorbent materials for CO₂ capture through physisorption at low temperature (25°C~150°C) and high pressure. Adsorbents, studied such as zeolites (Bezerra et al., 2014; Hudson et al., 2012), carbon-based materials (Sevilla & Fuertes, 2012; Wickramaratne & Jaroniec, 2013), and metal-organic framework (MOFs) structures (An & Rosi, 2010; Xue et al., 2013), etc. has shown promising results in several fields.

However, these materials frequently adsorb steam preferentially over CO₂, while possessing a lack of CO₂ adsorption capacity at atmospheric pressure and slightly eminent temperatures. Adsorption processes include temperature, pressure, and volume

swing adsorption. Furthermore, the thermal swing sorption method relies on temperature variations for carbon capture and subsequent regeneration of the sorbent. In addition, these methods are linking the transportation of compressed CO₂ gas and subsequent storage in geological formations. Thus making the processes highly energy intensive, high cost, and economically less viable.

To overcome these problems, hybrid processes together with sorption and chemical conversion of flue gas CO₂ on the same material could be potentially more feasible to minimize the CO₂ emission. CO₂ capture using heterogeneous adsorption on solids at elevated temperatures can be an attractive approach with lower cost and energy consumption. Dual function solid materials containing an adsorbent and a catalysts have been developed that can capture CO₂ from flue gas, and upon hydrogenation produce methane which is used at the inlet of the power plant, thereby reducing the overall utilization of natural gas. The CO₂ capture and conversion to CH₄ can occur in the same reactor at flue gas temperatures. Excess stored renewable H₂ is used for methanation of the captured CO₂ (Duyar, Ramachandran, et al., 2015; Duyar, Treviño, et al., 2015). The stored H₂ is produced by water electrolysis, which is one way of utilizing excess solar and wind energy for electricity. H₂ or CH₄/C₂H₆ (as in reforming reactions) flows to convert adsorbed CO₂. This approach reduces the CO₂ transportation and storage challenge connected with CCS processes. Furthermore, the in situ capture and conversion process of CO₂ reduces the high-energy penalty connected with the thermal swing regeneration process prevalent in other postcombustion capture schemes, such as amine scrubbing. It is also worth mentioning that together the chemisorption and physisorption properties of sorbents could affect the working capacity of dual function materials (DFMs), while the physisorption is associated with the occurrence of van der Waals forces and a low heat of adsorption (Berger & Bhowan, 2011). CO₂ undertakes a covalent chemical reaction with the sorbent sites and acquires a much greater heat of adsorption in the chemisorption. Physisorption processes are less selective, which leads to the adsorption of other gases such as N₂ that lowers the purity of the adsorbed CO₂. However, chemisorption could lead to the permanent binding of poisons, such as SO₂, hence decreasing the capacity of the sorbents. Nevertheless, the predominant chemisorption process in DFMs allows higher affinity to CO₂ in diluted streams. Parameters such as the weight loading of sorbent and catalytic species, ratio of sorbent to catalyst, temperature of adsorption and reaction, and composition of the feed gas streams are important in the design of DFMs, as they are essential in the optimization of the sorption capacity, reactive potential, and stability.

The extent of chemical reactions occurring at their large surface and much desired catalytic reactions take place on nanosized zeolites for CO₂ capture process. The nanosized zeolites are highly stable for CO₂ capture up to 1000°C due to the stabilization effect from the confinement of zeolites and the bimetallic synergistic effect. They possess large micropore volume, which can decrease crystal size and extends the possible

application of the zeolites for CO₂ capture. Catalyst can enhance the reaction rate during CO₂ conversion process. Ni catalysts typically involve high reduction temperatures and are inclined to oxygen-containing feeds for methanation of carbon dioxide on Ni-incorporated MCM-41 catalysts (Du et al., 2007). Ruthenium has been widely studied, because the catalysts have high activity and selectivity in the methanation of CO₂. Moreover, we can develop nanosized zeolites with catalysts (Ni, Ru, Rh, etc.) using several methods such as hydrothermal (Omodolor et al., 2020), solvothermal (Omodolor et al., 2020), simple chemical (Abrishamkar & Izadi, 2013), thermal treatment (Xu et al., 2012), co-precipitation (Abrishamkar & Izadi, 2013; Wang et al., 2017), and electrochemical (Xu et al., 2008) methods. The DFMs could be characterized by using XRD, FT-IR, SEM, TEM, BET surface area, TGA, etc.

The hydrocarbon resources are attained from shale gas, coal, biomass, solid wastes, and CO₂ emissions (Yarulina, Chowdhury, et al., 2018). Wide research is performed to the catalytic CO₂ conversion to hydrocarbons such as production of methanol, ethanol, methane, gasoline, light olefins, and even aromatics (Cai et al., 2016; Kang et al., 2016; Li et al., 2017; Lin et al., 2016; Liu et al., 2020; Schwach et al., 2017). All of these processes for hydrocarbons conversions from CO₂ emissions involve metals and metal oxide catalysts such as the single atoms, clusters, oxides, carbides, alloy nanoparticles (e.g., Ru, Rh, Fe₃O₄, Ni/NiO, and carbon-based catalysts), etc. However, these isolated metals and metal oxides catalysts suffer from harsh sintering as they are not confined by supports, which in turn results in a poor catalytic stability, selectivity of desired component, and decreased yields in the direction of their intention products. Thus the production of a precise range of hydrocarbons or oxygenates remains a difficult scientific and technical challenge. For overcoming the product selectivity limitations and improving catalyst stabilities, researchers are focusing on the design of efficient catalysts with the help of zeolites as dual functions materials (DMFs). Zeolites are a significant category of shape-selective catalysts with uniform micropores, tunable acidity, and high thermal and hydrothermal stabilities (Bande et al., 2015; Chen et al., 2017; Hasegawa & Matsumoto, 2017; Pham et al., 2014; Siriwardane et al., 2005). Fig. 12.1 shows the scheme of zeolites with nonzeolites (DFMs) for CO₂ capture and conversion processes for hydrocarbon production.

Over the past decade, the popularity of nanosized zeolites has increased (Chong et al., 2015; Li et al., 2014; Wakihara et al., 2011; Zhang et al., 2014). Furthermore, they have been applied to various industrially significant catalytic processes (Corma, 1995). The design and development of zeolites-based dual functional catalysts, which are the combination of zeolites with those of metal/metal oxides catalysts enhances the CO₂ conversion of zeolites to hydrocarbons (methanol, ethanol, methane, gasoline, light olefins, diesel fuels, and even aromatics) (Megías-Sayago et al., 2019) (Fig. 12.2). Moreover, the zeolites and zeolites-based metal/metal oxides DFMs can be synthesized via various methods.

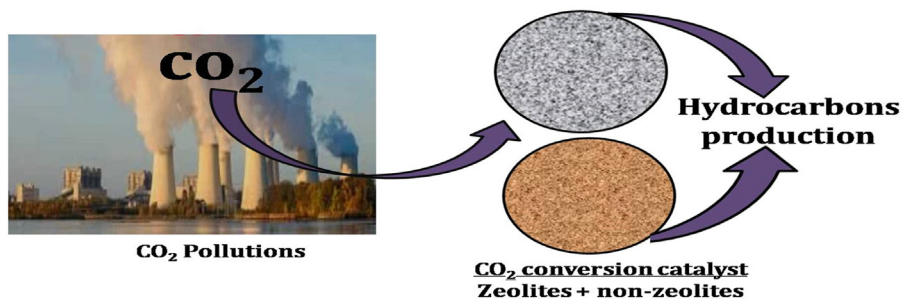


Figure 12.1 Zeolites with nonzeolites (DFMs) for CO_2 capture and conversion processes. *DFMs*, dual function materials.

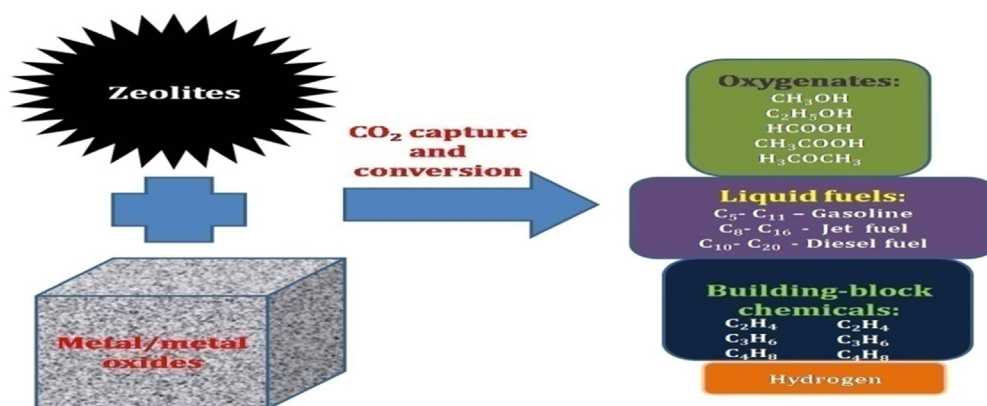


Figure 12.2 Schematic representation of CO_2 conversion to various hydrocarbon products using zeolite-based DMFs catalysts. *DMFs*, dual function materials.

In this book chapter, we describe the existent CO_2 capture processes, followed by relevant CO_2 adsorbents. Then, we focus on the development of various nanosized zeolite DMFs (Ni, Ru, and Rh) for CO_2 capture and conversion by different synthesis methodologies, application of various reaction extents, such as methanation, reverse water–gas shift, and dry reforming. We also elaborate on the challenges associated with these systems with an emphasis on the stability and regenerability of the materials. Finally, some future perspectives and possible areas of study are highlighted.

12.2 Synthesis methods of zeolites and metal/metal oxides containing zeolites

The synthesis of zeolites and zeolite with metal/metal oxides are carried out in the liquid phase, through various methods such as hydrothermal, solvothermal, and ionothermal (Zhang et al., 2020). For instance, Culfaz (2007) reported that ZSM-5 type zeolite was

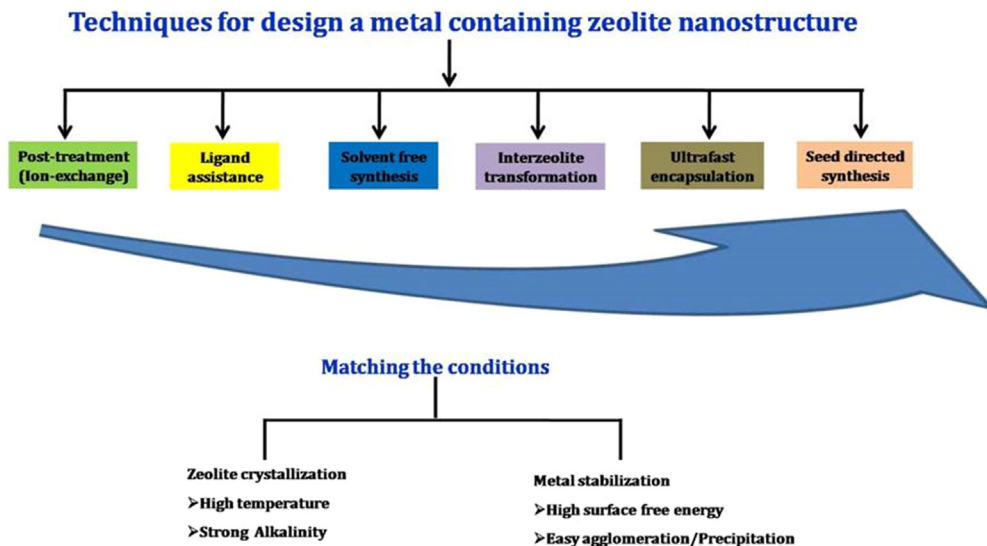


Figure 12.3 Various techniques for design a metal containing zeolite nanostructure.

prepared on seeded and unseeded macroporous α -Al₂O₃ disks from template-free batches using silicic acid, silica sol, and silica sol restrained methanol as stabilizer with different ratios of SiO₂/Al₂O₃. Ma et al. (2014) carried out the synthesis of 13X zeolite using low-grade natural kaolin through alkali fusion followed by hydrothermal treatment, exclusive of extra Si source or dealumination. The kaolinite, illite, and trace of quartz in kaolin sample were converted into zeolite followed by hydrothermal reaction with NaOH. Khan et al. (2009) synthesized metal oxides/zeolite core shell nanostructures via a simple secondary hydrothermal growth using tetrapropyl ammonium hydroxide fume silica and sodium hydroxide as source materials. For core-shell nanostructure formation, the absorption of zeolites nanoparticles on metal oxides particles by columbic attraction is followed via optimization of solution pH. Liu et al. (2019) found an approach for synthesis of isolated single metal catalysts in Y zeolite (Pt, Pd, Ru, Rh, Co, Ni, Cu) via in situ separation and confinement of a metal-ethanediamine complex into β -cages through the crystallization route followed by thermal treatment. Moliner (2012) reviewed the synthesis of metal-containing zeolites with precise functionalities and exclusive properties via a one-pot synthesis route. Currently, researchers are working on the design and functionalization of zeolites materials via different techniques (as shown in Fig. 12.3) as catalysts for various catalytic applications.

12.3 CO₂ conversion on zeolites-based catalysts

The CO₂ capture and catalytic conversion of CO₂ into hydrocarbons are much more eco-friendly, however their costs and efficacy varies. The zeolite-based thermocatalysts

(as DFMs) are used for CO₂ conversion process due to the unique properties of zeolites (high thermal stability and CO₂ selectivity). So far, several types of zeolites (zeolite Y, zeolite beta, metal-based zeolite (MFI)) have been investigated for CO₂ conversion into methane, alcohol, aromatics, gasoline, olefins, etc.

12.3.1 Dual functional materials for CO₂ conversion to methane

DFMs-based method can be executed isothermally; the exothermic nature of the methanation reaction supplies the required energy for CO₂ desorption from the adsorption sites (Wang et al., 2020). For each mole of CO₂ converted to methane, 165 kJ are generated and therefore available to support CO₂ desorption from the catalysts. Moderately mild reaction conditions can help to maximize the reaction conversion and to increase the selectivity of CH₄ which avoids the formation of by-products such as CO (Duyar, Treviño, et al., 2015). A variety of metals and zeolites composites as DFMs have been extensively studied for CO₂ methanation (Elliott & Lunsford, 1979; Quindimil et al., 2018). Zeolites act as supporting material for active metal. For example, Quindimil et al. (2018) demonstrated that Ni/La₂O₃/Na-zeolite Y formulation can perform 50% of CO₂ conversion to CH₄ at 350°C with high stability for more than 24 hours. In this work, the role of zeolite Y was to improve the CO₂ adsorption on Ni catalysts with its unique properties such as high specific surface area and it can modulate surface basicity with a combination of the alkaline earth metals. Costa-Serra et al. (2020) reported the ZSM-5 zeolite supported Ni catalyst for CO₂ conversion. The catalyst exhibits 68% of CO₂ conversion with 98% of CH₄ selectivity at 350°C. This work concludes that the zeolite has an influence on the hydrophobicity of Ni, and the enhances Ni reducibility and provides weak CO₂ adsorption sites in CO₂ conversion processes.

Elliott and Lunsford (1979) has carried out the methanation of CO₂ using Ru, Ru-Ni, Ru-Cu, and Ni Clusters in zeolite Y. They found that zeolite Y enhanced the stability of Ni catalyst at high temperature, and increased the kinetic reaction of Ru, Ru-Ni, Ru-Cu, and Ni clusters for CO₂ conversion due to pores of zeolite. Furthermore, Patzelová et al. (1984) reported on secondary pores of Y zeolite with Ni catalyst for CO₂ hydrogenation. The authors described that the secondary pores improved the stability and catalytic activity of Ni catalyst. According to the above report, the zeolites play a major role in the CO₂ hydrogenation (CO₂ adsorption, stability and active sides of catalysts) for CH₄ production.

12.3.2 Dual functional materials for CO₂ conversion to methanol

The major role of the zeolites (as support) is to increase and provide more surface for metal active sites for methanol production. The ZSM-5 zeolite holds unique properties such as Lewis and Bronsted acid sites, high resistance to water, and chemical and

thermal stability. An ideal catalyst is the one that can perform dual functions in a single reactor. Nowadays, much attention has been given to CuZnO/ZSM-5 zeolite which is the commercial catalyst for methanol synthesis from CO₂ and H₂ (Tariq et al., 2021). The catalysts should stabilize the desired intermediate (CO and H₂O) for high methanol yield by interacting strongly with CO₂. The commercial CuO/ZnO/Al₂O₃ attached with a zeolite membrane reactor can afford superior CO₂ conversion, methanol yield, and selectivity compared to a traditional reactor. The zeolite membranes act as support materials to increase CO₂ adsorption and preferentially permeate vapors in a gas–vapor mixture (Gallucci et al., 2004). A current study over on PdZn/ ZSM-5 zeolite also shows that CO produced through reverse water–gas shift facilitates the synthesis of methanol (Zhang et al., 2020). Ticali et al. (2021) demonstrated the Zr doped ZnO/H-ZSM-5 zeolite as a bifunctional catalyst for CO₂ hydrogenation to methanol production. The zeolite is one of the main factors for reducing coke and aromatic species formation via tuning of pore and channel dimensions.

12.3.3 Dual functional materials for CO₂ conversion to ethanol

Several zeolites have been tested with metal/metal oxides as DFMs to increase both the selectivity and activity to ethanol (Gao et al., 2020). Currently, the conversion of CO₂ to ethanol suffers from low selectivity or multiple processes with high energy consumption. The zeolites can improve the selectivity of CO₂ for metal catalysts. For example, Zhou et al. (2018) succeeded in the synthesis of ethanol using a three-component catalytic system composed of layer-by-layer ZnAl₂O₄, heterogeneous H-form mordenite zeolites, and ZnAl₂O₄ as a DFM, in which the selectivity of ethanol was ~50%. Ding et al. (2020) developed the beta zeolite and the active Cu nanoparticles to prepare DFMs in which the structure of copper is enclosed with particles of zeolite Beta, which showed excellent performance for CO₂ hydrogenation to ethanol. The ethanol yield in a single pass can reach up to 14% at 300°C, 12,000 mL gcat⁻¹ h⁻¹, and 2.1 MPa.

12.3.4 Dual functional materials for CO₂ conversion to olefins

Light olefins (C₂=–C₄=) are kinds of vital chemicals, and a number of studies have been carried out on research into the hydrogenation of CO₂ to light olefins using zeolites-based DFMs (Dang et al., 2018; Kim et al., 1998). Only a few research reports have been carried out on the selective hydrogenation of CO₂ to C₂–C₄ olefins using H-ZSM-5 zeolites-based catalytic materials (Hurtado et al., 2020). In addition, several research groups have been reported various promoters (porous cryptomelane-type octahedral molecular sieve manganese oxide (K-OMS-2) nanofiber (Hu et al., 2013), potassium) (Sathawong et al., 2015; Visconti et al., 2017; Wang et al., 2013) to modify with iron-based catalyst. Zeolite-based supports can enhance catalytic activity

to produce light olefins. For example, Wang, Wu, et al. (2019) also introduced the K to promote Fe-based catalysts (FeK—graphene/H-ZSM-5zeolite) for the CO₂ hydrogenation to light olefins. They found out that the lower olefins were the majority (92%) among other hydrocarbons when coupled with the beta zeolite. Gao et al. (2017) also found the beta zeolites with In₂O₃ acted as DFMs catalyst for CO₂ conversion to higher light olefins. The zeolite pores were support for formation of C-C coupling to produce C₅₊ range hydrocarbons.

12.3.5 Dual functional materials for CO₂ conversion to gasoline/diesel fuels

The highly efficient catalysts can support or be the key of this direct CO₂ hydrogenation process to gasoline. Wei et al. (2017) suggested a reaction scheme for CO₂ hydrogenation to produce hydrocarbons over Na—Fe₃O₄/H-ZSM-5 zeolite multifunctional catalyst as DFMs. During CO₂ hydrogenation, CO₂ is reduced to CO in the presence of H₂ using reverse water gas shift reaction (RWGS) on Fe₃O₄ sites, which is via a consequent hydrogenation of CO to α-olefins passing through RWGS on Fe₅C₂ sites. The olefin intermediates produced on the side of iron-based catalyst then diffuse into zeolite acid sites, on which the reactions undergo acid-catalyzed reactions. Hence, the gasoline type isoparaffins and aromatics are selectively produced and diffuse out of zeolite pores. In addition, CO₂ conversion and product selectivity are modulated by a changeable mass ratio of Na—Fe₃O₄ relative to H-ZSM-5 zeolite, which further supports the above assumption that Na—Fe₃O₄/H-ZSM-5 Zeolite catalyst is DFMs, and the reaction involves the intermediate migration surrounded by different active sites. This DFMs catalyst comprised Na—Fe₃O₄ nanocatalyst and nanocrystalline HZSM-5 zeolite (Na—Fe₃O₄/HZSM-5 catalyst) for the direct conversion of CO₂ to gasoline-range hydrocarbons. This catalyst exhibits confirmation of selectivity toward C₅—C₁₆ hydrocarbons (78%) (high gasoline range hydrocarbons) as well as low CH₄ and CO selectivity under industrially relevant conditions.

12.3.6 Dual functional materials for CO₂ conversion to aromatics

One of the hopeful energy conversions is CO₂ reductive valorization process. It consists of the composites of conservative metallic catalysts with ZSM-5 zeolites (Dokania et al., 2019). This permutation has been shown to permit the direct conversion of CO₂ to chemicals with selectivities for whichever light olefins (C₂—C₄) or aromatics over the drawback of classical Fischer—Tropsch synthesis (FTS), as shown by the Anderson—Schulz—Flory (ASF) sharing (Gao et al., 2018; Wei et al., 2017). In these DFM systems, the conversion of CO₂ to hydrocarbons can continue throughout two diverse routes: (1) the CO₂ transfer into CO by way of RWGS and consequent conversion of CO to hydrocarbons through the classical



Figure 12.4 Shows the reaction mechanism of CO₂ Hydrogenation over the DFMs catalyst, ZnZrO/ZSM-5. DFMs, dual function materials.

FTS mechanism (Ramirez et al., 2018), and followed by hydrocarbon cracking, isomerization, aromatization, etc. on the zeolites (Rahimi & Karimzadeh, 2011); (2) the conversion of CO₂ into methanol (Jadhav et al., 2014), followed by its conversion into hydrocarbons using the zeolite supports by the use of the classical methanol-to-hydrocarbons (MTH) mechanism (Yarulina, Chowdhury, et al., 2018; Yarulina, De Wispelaere, et al., 2018).

To defeat the boundaries of the above materials, different researchers prepared various new DFMs composites, which contain potassium superoxide-doped iron oxide and acidic zeolites. Further, potassium superoxide-doped iron oxide catalyst (Fe₂O₃@KO₂) (Ramirez et al., 2019) mixed with highly acidic zeolites to fine-tune the creation distribution to whichever light olefins (C₂–C₄=) or aromatics. Tandem catalytic system (as DFMs) with metal oxide and zeolite, where CO₂ activation and C–C coupling reaction can take place consecutively on two different types of active sites, and with CH₃OH as the intermediate for chain growth. Li et al. (2019) demonstrated the synthesis of a tandem catalyst ZnZrO/ZSM-5 for the conversion of CO₂ to aromatics along with aromatics selectivity of 73% in the presence of CO₂ by its conversion of 14%, and the selectivity of CO was suppressed down to 44% over a tandem catalyst ZnZrO/ZSM-5 as DFMs, which was constructed by a combination of ZnZrO solid solution and H-ZSM-5 zeolite. This type of reaction pathway is favorable for specific tailoring of the two independent active sites and the highly selective formation of the intention products via the shape-selective effect of the zeolite system. The hydrogenation of CO₂ to aromatics reaction mechanism on ZnZrO/ZSM-5 as DFM catalyst is shown in Fig. 12.4.

12.3.7 Dual functional materials for CO₂ conversion to other chemicals

The development as well as the reaction mechanisms of DFMs catalysts have been investigated over alkali metal halides (Gomes et al., 2008; Iwasaki et al., 2000; Kihara et al., 1993; Zhao et al., 1999; Zhu et al., 1996), organic bases (Barbarini et al., 2003;

Gomes et al., 2008; Shen et al., 2003), metal oxides (Bhanage et al., 2001; Yamaguchi et al., 1999; Yano et al., 1997; Yasuda et al., 2002) zeolites (Dorskocil et al., 1999; Srivastava et al., 2003; Yasuda et al., 2002), and metal complexes (Alexakis et al., 2000; Chen et al., 2007; Hoon et al., 2002; Jutz et al., 2008, 2009; Li et al., 2003; Paddock et al., 2004; Paddock & Nguyen, 2001; Shen et al., 2003; Srivastava et al., 2003) for CO₂ conversion to other chemicals. The tandem catalysts were made up with oxides and zeolites as DFMs (Cheng et al., 2016; Dokania et al., 2019; Jiao et al., 2016; Li et al., 2019; Ramirez et al., 2019; Wang, Tan, et al., 2019). Apart from those hydrocarbons, DFMs catalysts can be used in CO₂ conversion to other useful products such as urea, salicylic acid, formaldehyde, formic acid, cyclic carbonates, ethylene carbonate, di-methyl carbonate, etc. The activity of alkali metal-modified zeolites as DMFs has been investigated for the combination reaction of ethylene oxide and CO₂ to generate ethylene carbonate. The major role that zeolite plays is the water adsorbed in pores in ethylene carbonate formation by the electropositivity of the exchange cation (Dorskocil et al., 1999). Zeolite-Y has been synthesized with metal phthalocyanine complexes to show excellent catalytic performance for the cycloaddition of CO₂ to epichlorohydrin and propylene oxide yielding the corresponding cyclic carbonates (Srivastava et al., 2003).

12.4 Design of dual functional materials for CO₂ conversion

As a substitute for gathering CO₂ from whichever flue gases or the atmosphere followed by processes of transportation to utilization and storage sites, a two-step process focusing on combining the carbon capture, utilization, and storage (CCUS) units for power plants has been researched widely. The process would reduce the use of energy and cost inputs considerably. DFMs are intended as exclusive CO₂ capture and conversion materials to fuel solution as per the CCUS concept. The DFMs captures CO₂ from industrialized effluents and the adsorbed CO₂ is converted to liquid fuel and/or light hydrocarbons at the effluent temperature under catalytic conditions using renewable H₂. Furthermore, several methods are used to synthesize, design, and develop metal/zeolites-based DFMs such as solvothermal method, Pechini sol – gel, coprecipitation, sol – gel combustion, wet impregnation, and incipient wetness impregnation, etc.

12.4.1 Nickel (Ni) and zeolites-based dual functional materials

Ni-based catalysts are reported to show competitive catalytic activity with noble metals toward CO₂ methanation and are relatively low cost compared to noble metals (Bacariza et al., 2019; Guo et al., 2018; Le et al., 2017, 2018; Ratchahat et al., 2018). However, Ni-based catalysts are suitable at low temperature because of the aggregation of Ni nanoparticles and carbon deposition (Song et al., 2017). In recent research activities,

mesoporous zeolite molecular sieve as support is considered as an efficient mode to improve Ni dispersion, antisintering, and carbon-resistance, because they have large specific surface areas, tunable pore size, and high hydrothermal stability (Aziz et al., 2014). Kim et al. (2018) reported the dry reforming of methane using Ni/MgO/H-ZSM-5 containing a CaO, and H-ZSM-5 as sorbent. Al-Mam described the Ni-permeated CaO- and MgO-based double salts supported on H-ZSM-5 as DFMs for CO₂ capture and conversion to syngas via dry ethane reforming. Similarly, Ni – Na₂O/H-ZSM-5 DFMs were prepared for CO₂ capture. The Ni-based DFMs catalyst captured 9.55 mL of CO₂. The H-ZSM-5 zeolite was used as a support to enhance CO₂ adsorption over the Ni catalyst (Al-Mamoori et al., 2018; Yan et al., 2016).

12.4.2 Ruthenium and zeolites-based dual functional materials

Ruthenium (Ru) has been widely worked on due to its high activity, good stability, and selectivity in the methanation of CO₂ (Duyar, Ramachandran, et al., 2015; Garbarino et al., 2015; Janke et al., 2014; Kim et al., 2019). Ru has increased a lot of toehold because of its quick reversible redox behavior that permits it to straightforwardly transition between its metals and oxide states (Wang et al., 2017). These features have made this element an energetic substitute for Ni in the development of DFMs in spite of its high cost. However, the Ru has not enough capability for CO₂ absorption. Moreover, zeolites can support Ru to improve CO₂ gas absorption. For example, Khan et al. (2009) investigated the zeolite-based Ru DFMs for CO₂ capture and methanation. In this work, they made a remarkable contribution of CaO as a sorbent and Ru as a catalyst in the component and together were detached onto a MFI-type zeolite support. This DFM was prepared by conventional incipient wetness impregnation route using Ru(NO)(NO₃)₂ and Ca(NO₃)₂ as precursors. Similarly, another DFM was prepared by impregnating Ca(NO₃)₂ on 10 wt.% Ru/MFI-type zeolite, which was followed by an in situ reduction at 320°C to decompose Ca(NO₃)₂ into CaO (Wang et al., 2020). Optimization studies demonstrated that 5% Ru, 10% CaO/MFI-type zeolite was the best DFM put on show for the maximum methanation activity of 0.50 g-mol CH₄/kg.

12.4.3 Rhodium and zeolites-based dual functional materials

Rhodium (Rh) is a good catalyst for CO₂ conversion to various hydrocarbons. However, the Rh has less selectivity in CO₂ conversion to produce hydrocarbons. To overcome the drawback, the researchers are focusing on tuning the catalytic properties of rhodium on a MFI-type zeolite (as support), for the selectivity of CO₂ gas. The MFI-type zeolite is one of the best supports for CO₂ selectivity. Based on the idea, Wang et al. (2019) reported that the complexes firstly present as Rh(C₂H₄)₂ bonded to the zeolite HY catalyst for both the dimerization and hydrogenation of ethane and

CO₂ gas, with more selectivity for the former. Treatment of the catalyst to build rhodium clusters of only a few atoms each, on average, boosts the activity for hydrogenation. Rh-based DFMs have been one of the most studied for capture and conversion of CO₂. Wang et al. (2020) reported the Rh nanoparticles in proton-form ZSM-5 zeolite (Rh/HZSM-5) show methanation as a leading reaction, at the same time as the RhK/HZSM-5 catalyst exhibits CO selectivity higher than that of methane.

12.5 Effect of zeolite topology in the dual functional materials

The rational building of four-connected, particularly tetrahedrally connected, porous nanomaterials is associated with their topology and function to zeolites. The zeolites topology can be enlarged cavities and periodic intra- and/or extraframework DFMs functionality, which is an enduring synthetic challenge, and it is of outstanding scientific and technological interest. The large and extralarge cavities with pore size show enormous potential for novel applications (serving as nanoreactors, becoming a platform for a variety of alternative applications pertaining to large molecules, nanotechnology, optics, sensor technology, and medicine, for example), and enhancing the correlation between structure and function of DFMs. Zeolite topologies have the potential of providing precise and specific separation of gases, such as the removal of H₂O, CO₂, and SO₂ from low-grade natural gas streams. Zeolites topologies can build two main properties in DFMs such as adsorption and ion exchange with active sites and pores.

(Jiao et al., 2016, 2018; Ono, 1992) ZSM-5 had been chosen by many previous researchers, owing to the recognized capability of topology to produce aromatics and mordenite (MOR) due to its ethylene shape selectivity within the 8-membered ring (8Mr). The varying pore topology of both zeolites may give details in the distinction of carbocation stability. The zeolite pores size was previously shown to significantly affect the stability of alkene intermediates (Bučko & Hafner, 2015; Gounder & Iglesia, 2009, 2010; Moor et al., 2011; Nguyen et al., 2012; Sarazen et al., 2016; Sarazen & Iglesia, 2017). The stabilization of the involved intermediates can be subjective and the selectivity can be tuned toward the targeted product distribution by correctly choosing the zeolite topology.

12.6 Environmental impact of use of zeolites in the dual function materials for CO₂ capture and conversion technologies

As global warming is continuing at an growing rate, CO₂ abatement technologies systems are seriously required to mitigate climate change. Looking forward to the market need for CO₂ capture and conversion to valuable products (exploiting renewable hydrogen), the research keeps on improving the DFMs for both processes, capturing CO₂ and converting it into synthetic hydrocarbons at the site. The DFMs can be

infused with a high amount of CO₂-adsorbing zeolites. In the DFMs, zeolites have excellent capabilities for capturing carbon dioxide, and they can be combined with metal/metal oxides as catalysts, with strong mechanical and electrical properties.

However, the exposure of toxic metal-loaded zeolite-based catalysts could contaminate the environment and affect health. In particular, there are some specific limitations toward CO₂ conversion. The microporous nature of zeolite could limit the mass transfer of reactants which hinders the capturing and conversion of CO₂. Due to the large dipole moment of water, the capturing of CO₂ over zeolite under humid conditions is quite a challenge. The property of zeolite changes after the catalytic process, which may lead to diminishing catalytic activity. The formation of coke in the internal channel of the zeolite catalyst during the catalytic process may lead to catalytic deactivation. The pore structure and acidity of the zeolite are mainly influencing the coke formation. The great sensitivity of zeolites leads to deactivation by irreversible adsorption.

12.7 Future challenges and prospects for the use of zeolites as dual function materials catalysts for CO₂ capture and conversion technologies

Microsized crystallites of zeolites are at present used in catalytic, adsorption, and membranes applications. However, the “perfect zeolite” with superlative size, morphology, porosity, and structure for demanding applications in catalytic, adsorption and ion-exchange membranes is a challenge. A most important future challenge of zeolites is their external surface, which is insignificant compared to the large surface area developed in their internal micropores. The coverage of chemical reactions taking place at their external surface is therefore generally inconsequential in evaluation with the reactions taking place inside micropores, where their catalytic activity, adsorption, and membranes are maintained by the several facets of shape selectivity, whereby their normal microporous channel system sometimes brings strict mass transport limitations reducing their catalytic, adsorption, and membranes performance efficiency. When zeolite micropores are linked to mesopores and/or macropores, they are frequently referred to as “hierarchical.” At the engineering stage zeolites have been established to be thermally stable up to temperatures applicable for large-scale applications, but they are have conductivity for CO₂ conversion as DFMs catalysts. To overcome the catalytic activity the conductivity of zeolites is improved via composites with metal/metal oxides.

In view of the fact that the primary synthesized zeolite are accessible, efforts have been made to engineer the zeolites as DFMs in numerous aspects to improve their catalytic activities and performance as the catalysts. Approaches are formulated to improve the catalyst activity, increase the selectivity to the preferred product, and to recover catalyst stability and life time by optimizing the acidity, scaling down the crystal size, shifting the pore structure, and adjusting with heteroatoms. Herein the work of design

of zeolites has discussed subsequent new move toward engineering zeolites to change the porosity and acidity of the catalysts. The optimum results are in any future modification that may ultimately be applied to a multiplicity of industrial applications such as the catalytic conversion of CO₂ and its capture.

To design and develop the composites of zeolites is extremely an attractive topic particularly when they are utilized as catalysts. A noticeable catalytic synergism generally shows in the synthesized composite, no matter what type of structure it stands on. One of the new moves toward the synthesis of zeolite composite may change the zeolite chemical and physical properties and revise the catalytic performances to raise the zeolite of one type as a “shell” above an additional type of zeolite as the “core” crystals. In the core–shell structure, the preferred reactant or the intermediate with precise properties, proscribed by the shell layer, have a way into the core. The information has resolutely established that these core–shell structures not only change the morphology of the zeolite but also modify the acidity, pore size, adsorption–desorption behavior, in situ reaction mechanism, and distribution of the reaction products. Future prospective and strategy is that as prepared zeolites DFMs catalysts can be applied for CO₂ capture and conversion technologies.

Zeolite-based catalysts have been extensively used to capture and convert CO₂ into hydrocarbons. This is because of zeolites with abundant functionalization sites on its surface, tailorable pore structure, and shape selective characteristics that contribute to the enhanced activity toward the CO₂ conversion. Moreover, zeolite has a strong electric field, which favorably absorbs large dipole and quadrupole moments of CO₂. The presence of Lewis and Brønsted acid sites in the zeolite could absorb and activate the CO₂ and stimulate dissociation of water, thus improving the CO₂ conversion efficiency. The incorporation of amines into zeolite material could enhance the basic strength and CO₂ capture (Dinda et al., 2019). Also, metal-modified zeolite shows improved performance for capturing CO₂, which is mainly attributed to the molecular trapdoor effect (Shang et al., 2012). As compared to the random orientation of zeolite membrane, uniformly aligned zeolite-based membranes enhance CO₂-selective permeance (Zhou et al., 2014). The multiple active sites of zeolite-based catalysts induce the tandem reactions including reduction, hydrogenation, isomerization/aromatization/oligomerization (Wei et al., 2017). In addition, Brønsted acid sites in zeolites could enable the transfer of proton to the adsorbed on its surface and facilitates the formation of hydrocarbon (Wu et al., 2013). Besides, it shows potential to be the industrial catalyst for CO₂ conversion due to the remarkable stability for long time operation (Wei et al., 2017). Due to its high surface area (200–860 m² g^{−1}), the materials present improved stability, enhanced CO₂ capturing capacity, higher degree of oxygen defect, more electron trapping ability, better charge separation ability and capability to stabilize radical cations. Thus, the zeolites have been used as support of photocatalyst for photocatalytic conversion of CO₂ and thermochemical process, mainly attributed to the molecular scale pore structure. A few nanometers thickness

of zeolite nanosheets enhances the CO₂ adsorption, promotes the mass transport, and increases the surface excitation during CO₂ photoreduction (Lo & Taghipour, 2021; Tong et al., 2016). Furthermore, the oxygen vacancy of supporting functional material and porous structure of zeolites facilitates the generation of hydrocarbons with high octane number and suppresses the unintended reverse water–gas shift reaction (Gao et al., 2017).

So far, the studies on zeolites-based catalysts have focused on hydrogenation of CO₂, absorption of CO₂, photocatalytic conversion of CO₂, and solid oxides electrolysis for CO₂ conversion. Not much has been reported on photoelectrochemical (PEC) CO₂/H₂O cosplitting for the production of hydrocarbons (such as natural gas, alcohol, aromatics, gasoline, and olefins) using the zeolites as a photoelectrocatalyst. The researcher should focus on the synthesis of new zeolite-based photoelectrocatalysts and membranes by altering zeolite topology for efficient PEC CO₂/H₂O cosplitting applications. The prepared zeolite must have unique photoelectrocatalytic properties such as tunable band gap, which can be active in the UV–VIS range, and uniform pore size, which can improve ionic interactions and improve CO₂ uptake capacity, and the ability to stabilize radical proton and trap electrons within catalytic system (metal/metal oxides, membrane, etc.). The zeolite-based photoelectrochemical approach may overcome the drawbacks of other CO₂ conversion methods (thermochemical, solid oxides electrolyzer, etc.) and may help the production of high-purity hydrocarbons at lower cost, and in an eco-friendlier way. The PEC-based approach can also be used for the large-scale production of hydrocarbons, which is of great current interest to the industries.

12.8 Conclusions

Zeolites are exclusive materials with enhanced catalytic potential that when activated with metals or metal oxides act as adsorbents and membranes support materials in several applications. The unique physicochemical properties (shape/size selectivity, tunable solid acidity, large surface area, and porosity) of zeolite-based composites could offer the capturing and conversion of CO₂ into different hydrocarbons and chemicals such as methanol, ethanol, methane, olefins, gasoline, aromatics, and other products (formic acid, formaldehyde, urea, and carbonate). Some fundamental aspects have been pointed out as crucial for improving the efficiency: (1) the zeolites have to be stable in aqueous solution; and (2) the acid sites are required to be well-dispersed and of appropriate strength. The introduction of metal, metal oxide, and acidic components with the zeolite exhibits improves activity and stability. Different active metal or metal oxide catalysts (Ru, Rh, Cu, Ni, Co, Fe) and sorbents (K and Na) coupled with zeolite-based composites display outstanding performance for CO₂ adsorption and conversion. Besides, the surface functionalization significantly influences the CO₂ adsorption/desorption and product selectivity. Moreover, the physicochemical properties and activity of catalytic material mainly depends on the zeolite topology.

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