

## Research Article

# Designing Hierarchical ZSM-5 Materials for Improved Production of LPG Olefins in the Catalytic Cracking of Triglycerides

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LPG olefins (propene and butenes) are key building blocks in the petrochemical industry whose demand has been expanding steadily in recent years. The use of FCC (fluid catalytic cracking) units for conversion of triglycerides is a promising option for the future to boost production of LPG olefins. However, a need for innovative cracking catalysts is rising due to the different nature between petroleum and biomass-derived feedstocks. In this study, series of hierarchical ZSM-5 materials, namely, mesoporous ZSM-5, nanosized ZSM-5, and composite ZSM-5 were prepared, aiming to enhance the production of LPG olefins along with transportation fuels. Mesoporous ZSM-5 materials were synthesized by the postsynthetic modifications involving base treatment and subsequent acid washing, whereas nanosized ZSM-5 and composite ZSM-5 were synthesized by the direct-synthetic routes for a comparative purpose. The obtained materials were characterized by XRD, FTIR, N<sub>2</sub> sorption, TEM, AAS, ICP-AES, and NH<sub>3</sub>-TPD, and their catalytic performance was assessed in the cracking of triolein as a representative of triglycerides under FCC conditions. It was found that the subsequent strong acid washing step of alkaline treated ZSM-5 for removal of aluminum debris and external acid sites is needed to improve the catalytic performance. The resulting mesoporous ZSM-5 material shows higher yields of the desired products, i.e., gasoline and LPG olefins than its parent, commercial ZSM-5 at the almost complete conversion (ca. 90 wt.%). The selectivity toward LPG olefins is also enhanced over all the hierarchical ZSM-5 materials, particularly high for composite ZSM-5 (ca. 94 wt.%). The improved diffusion and lowered acidity of the hierarchical ZSM-5 materials might be responsible for their superior catalytic performance.

## 1. Introduction

LPG (liquefied petroleum gas) olefins including propene and butenes are important raw materials for the petrochemical industry to produce various valuable products such as PP (polypropylene), acrylonitrile, propene oxide, MTBE (methyl tert-butyl ether) ETBE (ethyl tert-butyl ether), etc. [1, 2]. Steam crackers and FCC units have accounted for the majority of LPG olefins, which are coproducts in the production of ethene and gasoline, respectively. However, the shale gas revolution has made ethane as a preferred feedstock for steam crackers, lowering the yield of LPG olefins by an order of magnitude [2, 3]. The shortage supply of LPG

olefins along with their strong global demand has provided driving forces for FCC units to enhance the production of LPG olefins along with transportation fuels. To achieve this, most of FCC units relies on the use of ZSM-5 either as separate particles or incorporating them in the FCC base catalyst [3, 4].

On the other hand, biomass in particular triglycerides has been regarded as the only sustainable source that can be able to replace partially petroleum feedstock in a mid- and long-term perspective [5, 6]. Thus, for application of ZSM-5 as cracking catalysts for efficient conversion of triglycerides to liquid fuels and LPG olefins, a high demand for research occurs due to the different nature of fossil and biogenic

materials. Over the years, ZSM-5 zeolites have been extensively investigated for the catalytic cracking of triglyceride-based feedstocks [5–10]. The medium pore zeolite ZSM-5 has been found to be very selective to  $C_5$ - $C_{10}$  hydrocarbons which correspond to gasoline fraction [9, 10]. However, the small pore size of ZSM-5 often causes intracrystalline diffusion limitations, reducing the selectivity to LPG olefins. Hence, intensified efforts have been focused on development of hierarchical ZSM-5 materials in different forms including mesoporous ZSM-5, nanosized ZSM-5, and composite ZSM-5 because they enable to couple the intrinsic catalytic properties of ZSM-5 with improved diffusion [11–13]. Consequently, hierarchical ZSM-5 materials have shown superior catalytic performance compared to their purely microporous counterparts in the catalytic cracking of triglycerides [14–19]. However, most of available studies have performed on a single type of hierarchical ZSM-5 materials with the focus on production of fuels, particularly gasoline. The design of hierarchical ZSM-5 materials aimed to enhance production of LPG olefins along with gasoline is relatively unexploited.

In the previous study [20], we successfully prepared mesoporous ZSM-5 materials via base-acid treatments of commercial ZSM-5 for the cracking of triglycerides to gasoline and light olefins ( $C_2$ - $C_4$  olefins). Compared to the untreated ZSM-5 material, the resulting mesoporous ZSM-5 material showed significant improvements in the conversion, yield, and selectivity of the desired products, i.e., gasoline and light olefins at the mild reaction temperature, i.e., 450°C. However, the selectivity to light olefins (light olefinicity) decreased sharply when rising the reaction temperature to 500–550°C that is relevant for industrial FCC processes. It is likely that the external acid sites generated by the postsynthetic treatments have promoted further transformation of primary light olefins via hydrogen transfer reactions under severe cracking conditions [21]. In the present study, it is planned to remove further external acid sites by applying a stronger acid washing step to improve the selectivity of LPG olefins at the elevated temperature. The resulting mesoporous ZSM-5 materials are thoroughly characterized, and their catalytic performance was evaluated in the cracking of triolein as a model feed for triglycerides under the FCC conditions. In addition, two other types of hierarchical ZSM-5 materials, i.e., nanosized ZSM-5 and ZSM-5 composite synthesized by using our reported protocols are also included for comparison to find suitable catalysts for cracking of triglycerides to gasoline and LPG olefins.

## 2. Materials and Methods

**2.1. Synthesis of Hierarchical ZSM-5 Materials.** Mesoporous ZSM-5 materials were prepared by the alkaline and subsequent acid treatments of the parent, commercial ZSM-5 (Zeocat PZ-2/25, Zeochem AG) as previously reported [20], but with some modifications. The parent ZSM-5, coded as P-Z, was first treated in the optimized NaOH aqueous solution (0.5 M) to generate mesopores. Then, the alkaline-treated sample, coded as M-Z-AT, was further treated in a

stronger HCl aqueous solution (1.0 M) to completely remove aluminum debris, which was formed and deposited on the external surface during the base treatment. The resulting mesoporous ZSM-5 material upon alkaline and acid treatments is labelled as M-Z-AAT. Composite nanosized ZSM-5/SBA-15 analogs, coded as C-Z, was synthesized from ZSM-5 precursors by using our previous recipe [19, 22]. In the first step, the precursor solution was synthesized by heating mixture of 6.0 g of tetraethyl orthosilicate (TEOS, 99%, Aldrich), 10.0 g of tetrapropylammonium hydroxide (TPAOH, 20% in water, Aldrich), 0.19 g of aluminum isopropoxide (AIP, 98%, Aldrich), and 2.0 g of distilled  $H_2O$  in a reflux system at 90°C for 24 h to partially form intact ZSM-5 nanocrystals. The resulting precursor solution was then added to the surfactant solution (triblock copolymer pluronic P123, Aldrich) to convert unreacted precursors, which have not been consumed in the first step into ordered mesoporous SBA-15 analogs, resulting in a composite nanosized ZSM-5/SBA-15 analogs. For the synthesis of nanocrystalline ZSM-5 (coded as N-Z), the precursor solution used for synthesis of C-Z was heated for a longer time (120 h) for complete crystallization [22]. Before characterization and catalytic testing, all the studied materials were transformed into the protonated form by two consecutive ion exchanges in 0.5 M  $NH_4NO_3$  solution at 80°C for 4 h.

**2.2. Characterization Methods.** The X-ray diffraction (XRD) studies were performed on a theta/theta diffractometer (X'Pert Pro from Panalytical). The Fourier transform infrared spectroscopy (FT-IR) measurements were carried out on an ALPHA-FTIR spectrometer (Bruker) using the ATR (Attenuated Total Reflection) sampling technique. The nitrogen physisorption measurements were conducted at  $-196^\circ C$  with an ASAP 2010 apparatus (Micromeritics). The transmission electron microscopy (TEM) micrographs were recorded at 200 kV with a JEM-ARM200F (JEOL). The temperature-programmed desorption of ammonia ( $NH_3$ -TPD) studies were performed in a home-made setup using a quartz tube reactor. The Al and Si contents were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES; 715-ES, Varian) and atomic absorption spectroscopy (AAS; Analyst 300, Perkin Elmer), respectively. The detailed characterization methods employed in this study are reported elsewhere [19, 20, 22].

**2.3. Catalytic Test Experiments.** The catalytic cracking of triolein was carried out on a fully automated single receiver short-contact-time microactivity test unit (SR-SCT-MAT, Grace Davison). The detailed experimental setup and testing procedure of the SR-SCT-MAT unit are described in the previous works [19, 20]. Technical triolein was used as a model feed for triglycerides since its acid compositions, i.e., oleic acid and linoleic acid are dominant in most plant oils such as rapeseed oil, soybean oil, or sunflower oil [9, 19]. The testing conditions were ambient pressure, 550°C, a catalyst-to-oil (CTO) mass ratio of 0.4 ( $g \cdot g^{-1}$ ), and a reaction time of 12 s.

The cracking products consisted of predominant hydrocarbons along with oxygenated compounds (mainly water, CO, and CO<sub>2</sub>) and coke. The gaseous hydrocarbon fraction is separated into dry gas (hydrogen, methane, ethane, and ethene) and liquefied petroleum gas (LPG; propane, propene, butenes, and butanes). The liquid hydrocarbons are lumped in terms of boiling ranges: C<sub>5+</sub> gasoline (<221°C), light cycle oil (LCO; 221–360°C), and heavy cycle oil (HCO; >360°C). These cracking products were analyzed by specialized analytical instruments as reported previously [19, 20]. The yield toward different products (Y<sub>i</sub>, wt.%) is defined as gram of product *i* per gram of the feed. The standard MAT conversion is defined as 100%—(Y<sub>HCO</sub> + Y<sub>LCO</sub>). The selectivity to propene is defined as a fraction of propene per total C<sub>3</sub> hydrocarbons (C<sub>3</sub> olefinicity). The selectivity to butenes is defined as a fraction of butenes per total C<sub>4</sub> hydrocarbons (C<sub>4</sub> olefinicity).

### 3. Results and Discussion

**3.1. Characterization of the Studied ZSM-5 Materials.** Figure 1 depicts the XRD patterns and IR spectra of the studied materials. As shown in Figure 1(a), the parent ZSM-5 (P-Z) and alkaline treated ZSM-5 (M-Z-AT) exhibit identical XRD patterns. This result indicates that the crystalline framework of ZSM-5 has been maintained under the optimized base treatment [20]. The subsequent acid treatment in a strong acidic medium (1.0 M HCl) does not affect the crystalline structure as could be judged from the XRD pattern of M-Z-AAT compared to that of M-Z-AT. It is expected since the framework structure of MFI zeolites has been proved to be stable in strongly acidic media [23]. Compared to the treated ZSM-5 samples, the composite material C-Z shows the same reflections, indicative of crystalline ZSM-5 phase present. However, the relatively high baseline along with the broadening X-ray diffraction peaks means that this sample has a lower fraction of crystalline ZSM-5. Among the studied materials, nanosized ZSM-5 (N-Z) shows the most resolved reflections, implying its highest crystallinity, being consistent with the previous observations [18, 22].

The crystalline structure of the studied ZSM-5 materials is also ascertained from their framework IR spectra (Figure 1(b)). The absorption band at ca. 548 cm<sup>-1</sup> is typical for asymmetric stretching mode of pentasil units in crystalline ZSM-5 zeolites [24]. N-Z exhibits the strongest absorption band ca. 548 cm<sup>-1</sup>, confirming its highest crystallinity, being in line with the result of XRD study. With respect to N-Z, the postsynthetic series P-Z, M-Z-AT, and M-Z-AAT display the same absorption band, but less pronounced, indicating their lower ZSM-5 crystallinity degree. For C-Z, the lowest crystallinity among others is evidenced by the least resolved absorption band at ca. 548 cm<sup>-1</sup> in its IR spectrum.

Figure 2 presents the N<sub>2</sub> adsorption/desorption isotherms of the studied ZSM-5 materials. All the materials show combined type I-IV isotherms. A high nitrogen adsorption at low relative pressures ( $p/p_0 < 0.01$ ) corresponds to micropore filling while a hysteresis loop occurs at high relative pressures ( $p/p_0 > 0.7$ ), indicative of mesopores

present. Commercial ZSM-5 (P-Z) and nanosized ZSM-5 (N-Z) exhibit a H4 hysteresis loop, typical for narrow slit-like, intercrystal mesopores [22]. On the other hand, M-Z-AT and M-Z-AAT show a H1 hysteresis loop, suggesting the formation of cylindrical mesopores. With respect to the treated ZSM-5 materials, the composite sample C-Z displays a more resolved hysteresis loop. This might result from the presence of ordered mesoporous SBA-15 analog phase in this sample. The detailed textural parameters are given in Table 1. As expected, all the hierarchical ZSM-5 materials exhibit an enlargement in their external/mesoporous surfaces compared to that of commercial ZSM-5 (P-Z). The enlarged external/mesoporous surface is particularly prominent for M-Z-AAT, N-Z, and C-Z which could have positive effects on their catalytic performance.

The TEM images of the representative ZSM-5 materials are depicted in Figure 3. The generation of intracrystal mesopores in M-Z-AAT can be clearly visible (Figures 3(a) and 3(d)). The TEM image of N-Z (Figure 3(b)) evidences the formation of nanosized crystals in the range of 50–100 nm. The lattice fringe of the ZSM-5 crystal phase is evidenced through its TEM image with high magnification (Figure 3(e)). The TEM micrographs of C-Z show the presence of spherical ZSM-5 nanoparticles embedded in mesoporous SBA-15 analogs, forming a composite structure with well-dispersed ZSM-5 nanocrystals (Figures 3(c) and 3(f)) [22].

The acidic properties of the studied ZSM-5 materials were investigated by NH<sub>3</sub>-TPD. The results are presented in Figure 4 and Table 1. From Figure 4, two NH<sub>3</sub> desorption peaks are observed at ca. 215°C and ca. 420°C which are generally attributed to weak and strong acid sites, respectively. The acid amount can be estimated from the peak area of TPD profiles (Table 1). Commercial ZSM-5 (P-Z) possesses a high amount of acid sites (1.24 mmol NH<sub>3</sub>/g) caused by its high aluminum content (Si/Al ratio = 11) [20]. The alkaline treatment (M-Z-AT) induces an increase in the acid amount (1.26 mmol NH<sub>3</sub>/g) and aluminum content (Si/Al ratio = 7). This can be explained by the selective extraction of Si from the ZSM-5 framework in alkaline media as well documented in the literature [11, 20]. The subsequent strong acid washing (M-Z-AAT) drastically reduces the acid amount (0.56 mmol NH<sub>3</sub>/g) and the aluminum content as well (Si/Al ratio = 27). The observation might suggest that the strong acid treatment applied has completely removed the aluminum debris deposited on the external surface, thus the external acid sites because the framework aluminum of ZSM-5 has been proved to be stable in strongly acidic media [23]. Nevertheless, M-Z-AAT has still a greater acid amount than N-Z though both the samples have similar Si/Al ratios of 27–28. It is likely that the postsynthetic treatment has created more defect sites such as terminal silanol groups, thus the greater acid amount for M-Z-AAT. With respect to the previous ZSM-5 samples, C-Z has the lowest acidity because of its lowest zeolite crystallinity as reported earlier [22].

**3.2. Catalytic Cracking of Triolein.** It is believed that catalytic cracking of triglycerides first undergoes thermal decomposition of triglyceride molecules to primarily form fatty

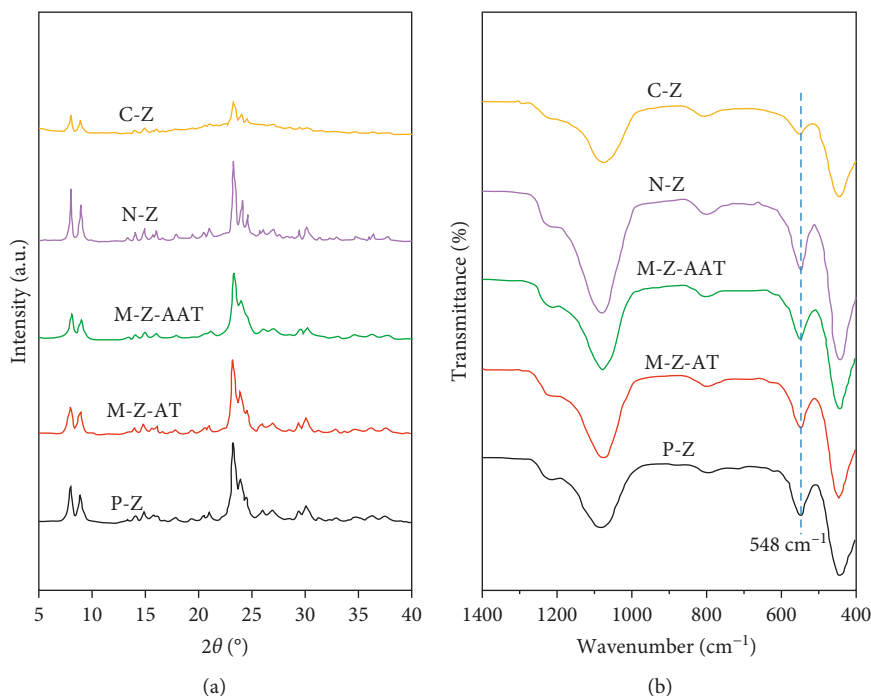


FIGURE 1: XRD patterns (a) and FTIR spectra (b) of the studied ZSM-5 materials.

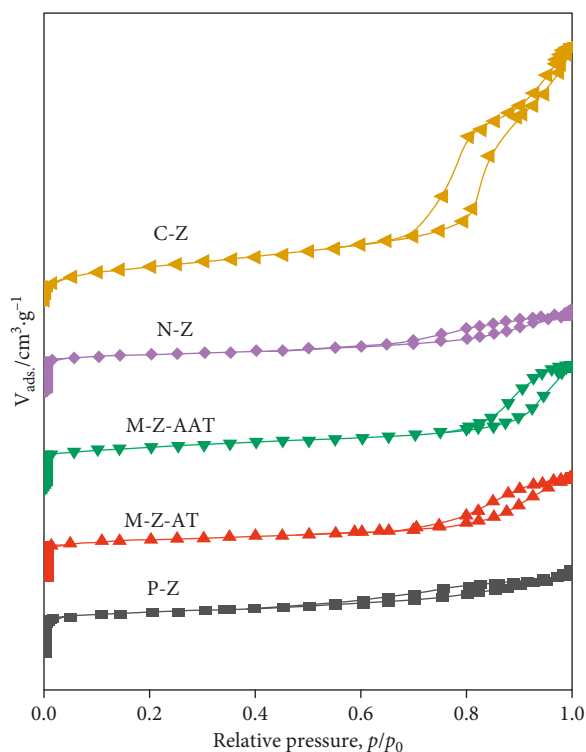


FIGURE 2: Nitrogen isotherms of the studied ZSM-5 materials.

acid by means of radical reactions, which is fast and independent of reaction temperature and catalyst characteristics [18, 20, 25]. The formed fatty acids are then converted over acid zeolite catalysts to produce a mixture of heavy hydrocarbons and oxygen-containing products (mainly CO,

CO<sub>2</sub>, and water). It is assumed that further conversion of these heavy hydrocarbons follows traditional catalytic cracking chemistry over zeolite catalysts which ends up in a wide range of products including dry gas, liquefied petroleum gas (LPG), gasoline, LCO, HCO, and coke as well documented in the literature [18–20, 25, 26]. In this study, gasoline and LPG olefins are desired products because the separation of ethene from the gaseous products is undesired from economic point of view because of its low concentration [4]. The catalytic performance of the studied ZSM-5 materials was assessed in the catalytic cracking of triolein as a representative of triglycerides under the optimized conditions (550°C, a CTO mass ratio of 0.4 (g·g<sup>-1</sup>) and a reaction time of 12 s) which were defined in the previous studies [19, 20]. The results are given in Table 2.

As shown in Table 2, the parent, commercial ZSM-5 (P-Z) delivers a high conversion (88.7 wt.%) and yields of the desired products, i.e., gasoline (37.3 wt.%) and LPG olefins (19.7 wt.%). It is expected since the medium pore zeolite ZSM-5 has been proved to be an effective catalyst for conversion of triglycerides to the desired products. However, it should be noted that the C<sub>3</sub> and C<sub>4</sub> olefinicity is relatively low, ca. 64–66 wt.%, suggesting the extensive occurrence of hydrogen transfer reactions that saturate LPG olefins to LPG paraffins (Table 2), being in line with the reported works [18, 20]. Alkaline treatment diminishes the catalytic performance. In fact, the conversion and yields of the desired products over M-Z-AT decline slightly while the coke yield increases appreciably (Table 2). The observed catalytic behavior can be linked to the formation of aluminum debris deposited on the external surfaces. These aluminum species partially block the pore mouth of micropores of ZSM-5. This prevents the access to acid sites in

TABLE 1: Physicochemical properties of the studied ZSM-5 materials.

Sample	Si/Al <sup>a</sup>	Si/Al <sup>b</sup>	$S_{\text{meso}}^c$ (m <sup>2</sup> /g)	$S_{\text{BET}}$ (m <sup>2</sup> /g)	$V_{\text{micro}}^c$ (m <sup>3</sup> /g)	$V_t$ (m <sup>3</sup> /g)	Acid amount <sup>d</sup> (mmol NH <sub>3</sub> /g)
C-Z	30	30	233	361	0.059	0.780	0.34
N-Z	30	28	142	394	0.124	0.264	0.41
M-Z-AAT	—	27	282	465	0.128	0.763	0.56
M-Z-AT	—	7	131	411	0.110	0.520	1.26
P-Z	—	11	110	373	0.113	0.219	1.24

<sup>a</sup>In the starting solution; <sup>b</sup>in the final products analyzed by AAS and ICP; <sup>c</sup>t-plot method; <sup>d</sup>NH<sub>3</sub>-TPD.

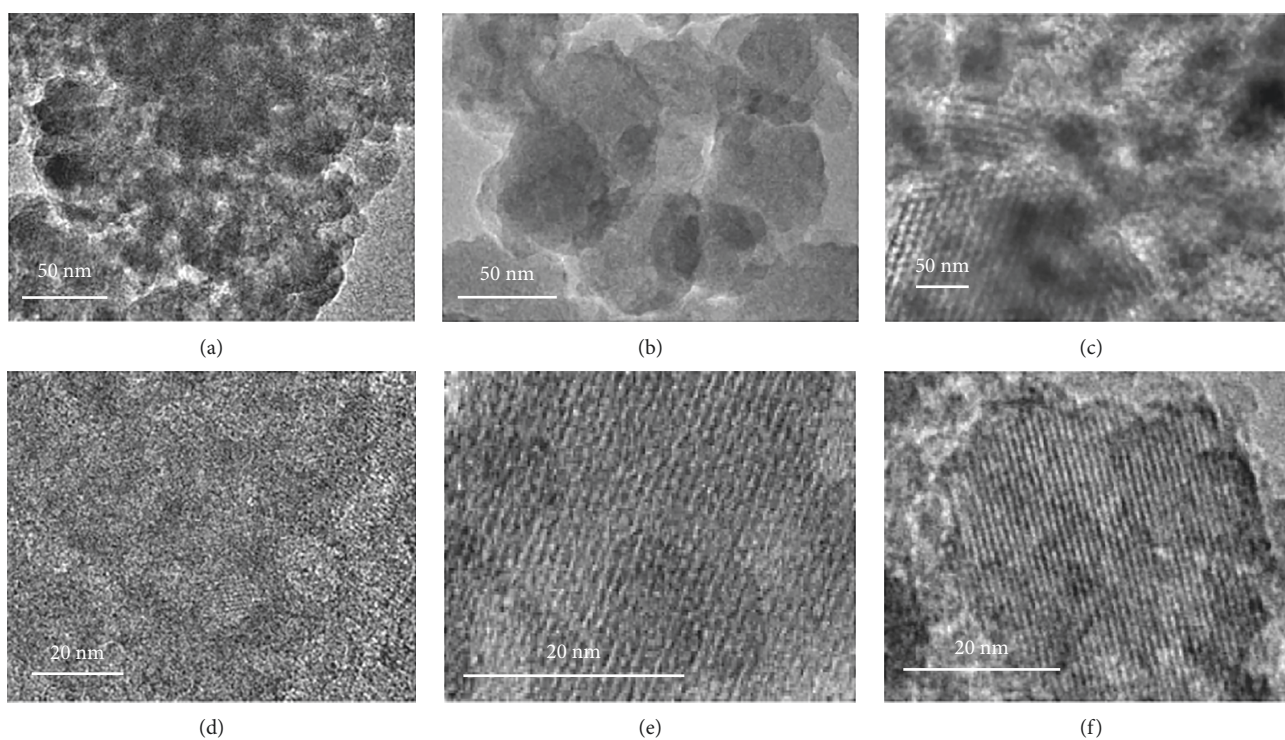


FIGURE 3: TEM images of the representative hierarchical ZSM-5 materials: M-Z-AAT (a, d); N-Z (b, e); and C-Z (e, f).

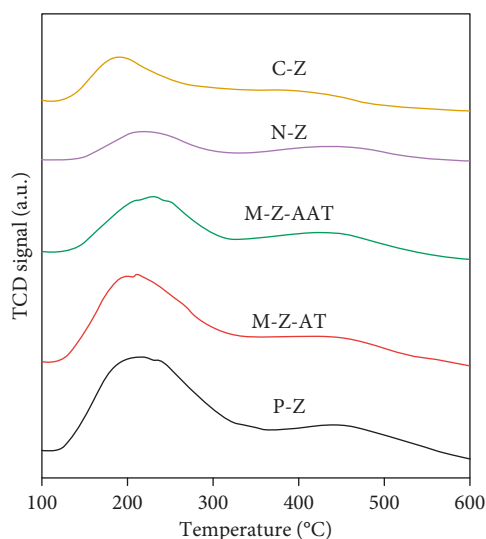


FIGURE 4: NH<sub>3</sub>-TPD profiles of the studied ZSM-5 materials.

the micropore channels to some extent and the consequent poor catalytic performance. On the other hand, such aluminum debris has been proved to mainly generate Lewis acid sites which promotes the formation of coke as reported elsewhere [20, 21].

Upon the acid treatment for removal of the aluminum species and consequent external acid sites, the resulting M-Z-AAT material exhibits the outstanding catalytic performance. With respect to M-Z-AT, the alkaline-acid treated sample M-Z-AAT gives a higher conversion of 88.2 wt.%, being comparable to that of commercial ZSM-5 (88.7 wt.%) despite its lower acid amount. The yields of gasoline and LPG olefins are also enhanced over M-Z-AAT, reaching the highest values (38.3 wt.% and 25.2 wt.%, respectively) among the postsynthetic series. These remarkable achievements can be rationalized by the presence of substantial, interconnected mesoporosity which enhances the accessibility of acid sites, thus the increased conversion. Simultaneously, it might suppress the undesired secondary reactions of reactive gasoline and light olefin molecules by shortening the

TABLE 2: Catalytic performance of the studied ZSM-5 materials in the cracking of triolein.

Sample	C-Z	N-Z	M-Z-AAT	M-Z-AT	P-Z
Conversion (wt.%) <sup>a</sup>	70.9	85.9	88.2	86.4	88.7
Product yields (wt.%) <sup>a</sup>					
Gas	28.0	41.6	42.2	41.3	43.3
Dry gas	2.3	6.4	5.8	7.6	7.8
LPG	18.0	28.7	31.0	28.3	30.5
Propane	0.6	3.4	3.0	5.8	6.3
Propene	7.8	12.2	14.2	10.2	11.9
Butanes	0.5	3.1	2.7	4.8	4.5
Butenes	9.2	10.0	11.0	7.5	7.8
LPG paraffins	1.1	6.5	5.7	10.6	10.8
LPG olefins	16.9	22.2	25.2	17.7	19.7
CO/CO <sub>2</sub>	7.6	6.5	5.4	5.5	5.0
Gasoline	35.2	37.0	38.3	36.9	37.3
LCO	18.7	9.4	7.4	7.2	6.7
HCO	10.4	4.7	4.4	6.4	4.6
Coke	1.6	1.0	1.3	1.8	1.2
Water	6.1	6.3	6.4	6.3	6.9
Olefinicity (wt.%) <sup>b</sup>					
C <sub>3</sub> /total C <sub>3</sub>	92.9	78.2	82.4	63.5	65.3
C <sub>4</sub> /total C <sub>4</sub>	94.8	76.6	80.2	61.3	63.7

<sup>a</sup>wt.% on feed basis; <sup>b</sup>wt.% on product basis.

diffusion path lengths, thus the higher yields of the desired products.

Compared to M-Z-AAT, the nanosized ZSM-5 sample (N-Z) shows a lower conversion (85.9 wt.%) and lower yields of gasoline (37.0 wt.%) and LPG olefins (22.2 wt.%). This can be attributed to its lower acid amount and mesoporosity. Similarly, the composite ZSM-5 (C-Z) gives the lowest conversion (70.9 wt.%) and lowest yields of gasoline (35.2 wt.%) and LPG olefins (16.9 wt.%) among the studied ZSM-5 materials. However, C-Z shows the highest C<sub>3</sub> and C<sub>4</sub> olefinicity, reaching ca. 93–95 wt.% that are considerably greater than those of M-Z-AAT (ca. 80–83 wt.%), N-Z (ca. 77–78 wt.%), P-Z (ca. 64–66 wt.%), and M-Z-AT (ca. 62–64 wt.%). It is worthy to note that the C<sub>3</sub> and C<sub>4</sub> olefinicity over M-Z-AAT is remarkably higher than that of P-Z and M-Z-AT, suggesting that the stronger acid treatment applied for complete removal of external acid sites is beneficial to the formation of LPG olefins. The catalytic behavior can be rationalized by the fact that LPG olefins are primary cracking products produced from end-cracking reactions of heavier compounds. Subsequently, they can be transformed into other types of hydrocarbons such as aromatics and light paraffins via hydrogen transfer reactions because of their high reactivity. These secondary reactions are largely suppressed over hierarchical ZSM-5 materials, i.e., M-Z-AAT, N-Z, and C-Z because of their reduced acidity and shortened diffusion path lengths. The latter results in the faster diffusion, avoiding the undesired secondary reactions by rapid transport of valuable LPG olefins out of zeolite crystals [18]. On the other hand, Ishihara et al. [15, 16] studied the cracking of soybean oil over composite zeolites and found that the introduction of a mesoporous matrix to zeolite crystals could also hinder these hydrogen transfer reactions since it reduces the possibility for reactive molecules exiting

a zeolite crystal to encounter another zeolite one. Taking these findings into account, the remarkably high selectivity to LPG olefins over C-Z might originate from its moderate acid amount coupled to its unique composite structure which effectively prevents these valuable LPG olefins from further transformation.

## 4. Conclusions

We have shown that the production of LPG olefins along with gasoline in catalytic cracking of triglycerides under the FCC conditions can be realized by proper design of hierarchical ZSM-5 materials. For the postsynthetic modification of commercial ZSM-5, the strong acid treatment step for removal of aluminum debris and consequent external acid sites is key to improve its catalytic performance. Compared to commercial ZSM-5, the alkaline-acid treated ZSM-5 (mesoporous ZSM-5) delivers higher yields of gasoline and LPG olefins at the almost complete conversion of ca. 90 wt.%. Nanocrystalline ZSM-5 is slightly less active, producing lower yields of gasoline and LPG olefins than mesoporous ZSM-5. However, both mesoporous ZSM-5 and nanosized ZSM-5 exhibit a comparable selectivity toward LPG olefins of ca. 80 wt.%. The advantage of composite ZSM-5 over the other hierarchical ZSM-5 materials is the remarkably high selectivity toward LPG olefins, reaching ca. 94 wt.%. These findings might be helpful for rational design of hierarchical ZSM-5 materials to enhance LPG olefins in catalytic cracking of triglycerides.

## Data Availability

The data used to support the findings of this study are included within the article.

## Conflicts of Interest

The authors declare that there are no conflicts of interest.

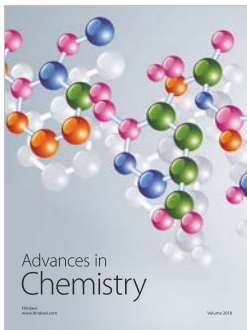
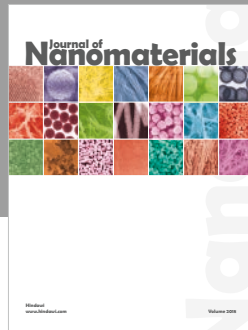
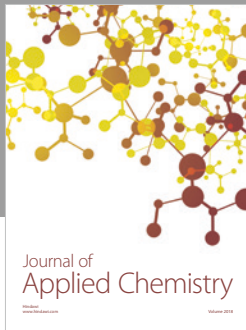
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