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1-Butene Cracking to Propene on High Silica HMCM-22: Relations Between Product Distribution and Feed Conversion Under Various Temperatures

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Abstract HMCM-22 zeolite with a high Si/Al_2 ratio (158) was applied to the 1-butene cracking reaction in a fixed-bed reactor. On basis of the product distribution, the main reaction pathways of C_4 alkenes catalytic transforming on HMCM-22 were proposed. Also, the preferences of the reaction pathways influenced by the temperature and conversion level were discussed, which could provide good guidance on enhancing the propene production.

Keywords High silica MCM-22 · Butene · Cracking · Propene

1 Introduction

MCM-22 zeolite, a novel microporous material invented by Mobil [1], has a unique structure which consists of two independent pore systems. One is defined by two-dimensional sinusoidal 10-membered-ring (MR) channels (4.1 Å × 5.1 Å), and the other is characteristic of 12-MR supercages (inner diameter 7.1 Å; height 18.2 Å) accessible by 10-MR windows (4.1 Å × 5.5 Å) [2]. Besides the two pore apertures inside the MCM-22 crystals, there are 12-MR pockets corresponding to half supercages (inner diameter 7.1 Å; height 7.0 Å) on the external surfaces [3, 4]. Due to its unique structure and acid distribution, MCM-22 catalyst has been widely used in many hydrocarbon processing reactions [5–9].

Recently, high silica MCM-22 (Si/Al_2 ratio > 50) was found to be rather active and selective in the process of butene cracking [10, 11], which showed good prospects in meeting the propene supply-demand gap. However, MCM-22 zeolites could only be synthesized within the Si/Al_2 range of 15–80, preferably 20–50 with conventional synthesizing methods [9, 12–15]. Impurities including ZSM-5 and kenyaite often appear in the products with further increasing Si/Al_2 ratios in the synthetic gels [9, 15]. In our previous work, a boron-containing way for one-pot synthesis of high silica MCM-22 was developed [16]. When the zeolites with various Si/Al_2 ratios were used as catalysts for 1-butene cracking to propene, the best performance was obtained on a sample with Si/Al_2 ratio of 158 due to its suitable acidity [16].

As proved in many hydrocarbon transformation processes, the preferences of the reaction pathways and thus the product distribution were not only influenced by the nature of catalysts but also by the reaction conditions. In the butene cracking processes, the formation of the desired product (propene) is accompanied by a series of by-products, and propene could also undergo secondary reactions at the same time [6, 10, 16, 17]. Thus propene is a kind of intermediate products, which means that high conversion in this reaction does not definitely bring about high propene yield. On a given catalyst, there must be an optimal reaction condition, both preferred in thermodynamic (reaction temperature) and in kinetic (contacting time) aspects, for enhancing the production of propene.

In this study, we have carried out 1-butene cracking reaction over high silica HMCM-22 (Si/Al_2 = 158). The main purpose of this study is to obtain an optimal operating

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condition for maximizing the propene production on the catalyst. For this purpose, evolution of the product distribution with the feed conversion was studied. The reaction pathways, as well as the influences of temperature and conversion level (contacting time) on the preferences of the reaction pathways, were also discussed, which could provide good guidance on enhancing the production of propene.

2 Experimental

2.1 Materials

Synthesis of the high silica MCM-22 zeolite was according to literature [16]. A detailed procedure was as follows: NaOH (96%, AR), $Al(NO_3)_3 \cdot 9H_2O$ (99.0%, AR), colloidal silica (25.6 wt% SiO_2, 0.06 wt% Al_2O_3), H_3BO_3 (99.5%, AR), and hexamethyleneimine (HMI) (95%, CP) were well mixed with molar composition of 0.1 Na_2O:1.0 SiO_2:0.0067 Al_2O_3:0.5 B_2O_3:1.0 HMI:40.0 H_2O. The mixture was stirred thoroughly for half an hour to form a homogeneous gel, which was transferred into a 250 mL stainless steel autoclave. The autoclave together with the gel was heated to 175 °C and held at this temperature for 168 h while being rotated at 60 rpm. After the crystallization was completed, the autoclave was cooled by tap water and the as-synthesized MCM-22 was obtained by filtrating, washing and drying.

The as-synthesized sample was calcined at 550 °C for 5 h in air to burn off the occluded organics. Then the as-calcined zeolite was ion exchanged with 0.8 mol/L NH_4NO_3 solution triply to give an NH_4-form sample, which was followed by calcination at 550 °C for 2 h to give HMCM-22. Finally, the HMCM-22 sample was pelletized, crushed and sieved to 20–40 mesh particles before application in the catalytic cracking of 1-butene.

2.2 Characterizations of the High Silica MCM-22

X-ray diffraction (XRD) pattern of the MCM-22 sample was recorded on an X Pert Pro X-ray diffractometer using Cu-Kα radiation, operating at 40 kV and 40 mA. The data were collected in 2θ range of 2.5°–50°. XRD results (not shown) indicated that the synthesized sample was quite pure MCM-22.

Chemical composition of the MCM-22 was analyzed on a Magix 601X X-ray fluorescence (XRF) spectrometer. The Si/Al_2 ratio was 158 in the as-synthesized sample and the same value in HMCM-22. That was quite close to the Si/Al_2 ratio (150) in the synthesis mixture.

Nitrogen adsorption measurement was carried out at liquid nitrogen temperature (−196 °C) on a Micrometrics ASAP 2010 equipment. The total surface area, microporous surface area and micropore volume of the calcined sample were 477, 339 m^2/g, and 0.162 cm^3/g, respectively, indicating that the high silica MCM-22 catalyst possessed good microporous structure.

The coking amount of the used catalysts was determined by thermo-gravimetric (TG) analysis on a Perkin–Elmer TG 1700 instrument. The experiments were carried out in a temperature range of 25–850 °C, with a heating rate of 10 °C/min in flowing air (100 mL/min).

2.3 Catalytic Evaluations

1-Butene cracking reactions were carried out at ambient pressure in a fixed-bed reactor with inner diameter of 7 mm. Before every evaluation, 1 g of the catalyst was pretreated at 500 °C for 1 h in a constant flow of N_2 (20 mL/min). Then 1-butene (99.5 wt%, Qilu Petrochemical Co. Ltd) or mixture of 1-butene with nitrogen was passed through the reactor at desired temperatures. Blank experiments were carried out with 1 g of quartz sand placed in the center of the reactor using the same reaction conditions in order to study the pyrolysis of the feed stocks.

The products were analyzed by an on-line Varian 3800 gas chromatography (GC) equipped with both a flame ionization detector (FID) and a thermal conductivity detector (TCD). A 100 m PONA column was used to separate the components of the products. The hydrocarbons were detected by the FID and in some runs hydrogen was also detected by the TCD. The quantitative determinations were carried out by using the corrected area normalization. All the butene isomers in the products were considered to be unconverted feed stocks. The conversion of feed stocks (X) and the selectivity (S) of a particular product C_xH_y were calculated as follows [16, 18]:

$$X = (C_4H_8\% \text{ in the feed } - \ C_4H_8\% \text{ in the products})/ (C_4H_8\% \text{ in the feed}),$$

$$S\left(C_xH_y\right) = \left(C_xH_y\% \text{ in the products}\right)/ \left(\Sigma C_iH_j\% \text{ in the products}\right),$$

where all the percentages were in weight percent.

3 Results

In this study, variations of the feed conversion under defined temperatures were obtained via adjusting the space velocity and/or the partial pressure of the feed stocks, thus the experiments need to be carried out on catalysts with comparable status. In our previous study, the performance of low silica HMCM-22 catalysts changed greatly during the initial 1–2 h, followed by a quasi-plateau stage [6]. However, the situation on high silica HMCM-22 has not been investigated yet. Therefore, catalytic evaluation of the high silica HMCM-22 (Si/Al$_2$ = 158) was first performed along with the time on steam (TOS).

3.1 Evolution of the Catalytic Performance with TOS on High Silica HMCM-22

The catalytic performance of HMCM-22 was evaluated under the following conditions: reaction temperature $T = 580$ °C, 1-butene partial pressure $P = 0.1$ MPa, and the weight hourly space velocity of 1-butene WHSV = 5 h^{-1} [16]. The results were presented in Fig. 1.

As can be seen from Fig. 1, the catalytic performance of the high silica HMCM-22 changed significantly during the first 2 h, which was similar to that of the low silica HMCM-22 [6]. The conversion of the feed stocks decreased obviously, accompanied by a rapid increase of the selectivities of propene and C_5^+ (non-aromatic hydrocarbons with carbon number ≥ 5), and a marked decrement in the selectivities of propane and aromatics. The selectivity of ethene showed a more moderate variation than the other products, it increased a little at the first half an hour, and then decreased slightly with further prolonged TOS. After 2 h of TOS, the product distribution came into a quasi-plateau stage, indicating a comparable status of the catalyst was achieved. In the following experiments, the conversion variations were conducted after 2–3 h of the initial pre-coking stages.

3.2 Evolution of Product Distribution with Feed Conversion Under Various Temperatures

For an on-purpose propene-enhancing process, flexible temperatures and high selectivity for propene with acceptable conversion level are desired. As has been mentioned above, there must be an optimal conversion level on a given catalyst for enhancing the propene production. In order to investigate the preferences of the reaction pathways and thus obtain the optimal reaction conditions, studies on the evolution of the product distribution with the conversion level under various temperatures were carried out, as presented in Fig. 2.

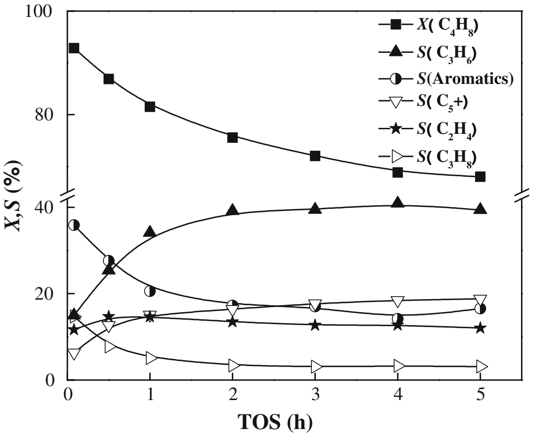


Fig. 1 Infulence of time on stream (*TOS*) on the catalytic performances of high silica MCM-22 (Si/Al$_2$ = 158) in 1-butene cracking. Reaction conditions: $T = 580$ °C, $P = 0.1$ MPa, WHSV = 5 h^{-1}, 1-butene (99.5 wt%) as feed stock

As can be seen from Fig. 2a, evolutions of the propene selectivity with the conversion showed rather similar tendencies under different reaction temperatures. At any defined temperature, the values increased gradually before an inflexion, and then decreased rapidly with further increasing of the conversion level. The conversion corresponding to the inflexion was about 70% at 500 °C, and those at higher temperatures were about 60%. Among the selected temperatures, the lowest propene selectivity was obtained at 500 °C while the highest was obtained at 580 °C at any defined conversion level. Thus 580 °C is the optimal temperature for enhancing propene. From the changing tendency of the propene selectivity, the optimal operating conversion range is 50–65% at 580 °C. If the conversion exceeds 65%, the selectivity of propene is unsatisfying; when the conversion is less than 50%, although the selectivity of propene is still acceptable, the process may be uneconomical as the single-pass propene yield is low.

As the growth of propene demanding has outpaced that of ethene in the global market, the propene/ethene ratio has become an important index for on-purpose propene-producing processes. The evolutions of propene/ethene ratios with the conversion level on HMCM-22 were given in Fig. 2b. As can be observed, the propene/ethene ratios decreased almost linearly with the increasing conversion at any defined temperature, and they decreased obviously with the increasing of temperature at any specified conversion. From the specific values in Fig. 2b, when the conversion was lower than 65% at 580 °C, propene/ethene ratios could be larger than 3.2, which is a high value among C_4^+ alkene cracking processes for boosting propene [19, 20].

The selectivity evolutions of two groups of main byproducts, i.e., aromatics and light alkanes (propane plus butanes), with the conversion level were depicted in Fig. 2c, d, respectively. Figure 2c demonstrated that the selectivity of aromatics increased with the increasing conversion at a certain temperature, and a more rapidly increasing tendency was observed when the conversion was higher than 60%. At any defined conversion level, higher aromatics selectivities were observed at higher temperatures. From view of inhibiting the formation of aromatics, relatively low conversion level and low reaction temperature are desirable. From the values in Fig. 2c, the selectivity of aromatics could be lower

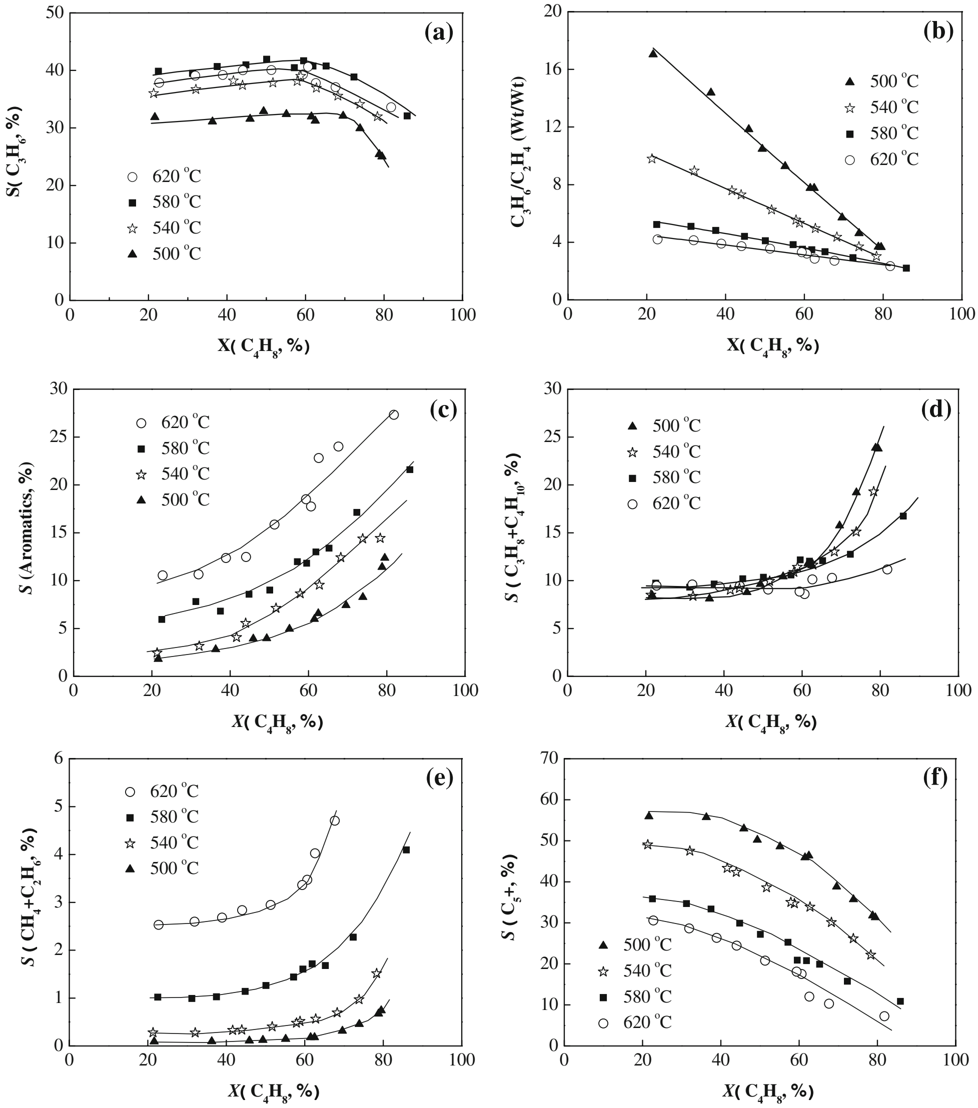


Fig. 2 Evolution of product distribution with the conversion in the reaction of 1-butene cracking on high silica MCM-22. **a** selectivity of propene. **b** weight ratio of propene/ethene. **c** selectivity of aromatics. **d** selectivity of propane plus butanes. **e** selectivity of methane plus ethane. **f** selectivity of C_5^+

than 14% when the conversion level is less than 65% at 580 °C. However, from another point of view, the co-production of aromatics, a kind of valuable chemicals, is also attractive in this process.

As can be observed from Fig. 2d, the selectivity of propane plus butanes increased slightly with the increasing conversion (<60%), and more rapidly when the conversion exceeded 60%. The selectivities of propane plus butanes at different temperatures maintained an equivalent level of about 10% when the conversion is less than 60%. However, they differentiated from each other upon higher conversion levels, and lower values were obtained at higher temperatures. If the conversion is controlled below 65% at 580 °C, the selectivity of propane plus butanes would be no larger than 13%.

Figure 2e presented the selectivities of methane plus ethane under various temperatures. As can be seen, the changing tendencies were very similar to those of aromatics (Fig. 2c). If the conversion is controlled below 65% at 580 °C, the selectivity of methane plus ethane would be less than 1.5%, whose influence on the propene production would be negligible.

Figure 2f exhibited similar evolution curves of the C_5^+ selectivities with the conversion under various temperatures. As can be seen, the selectivity of C_5^+ decreased more and more rapidly with the increment of conversion at any defined temperature, and it also decreased obviously with the increasing temperature at any specified conversion level. A selectivity of C_5^+ between 30 and 20% could be obtained if the conversion was controlled between 50 and 65% at 580 °C.

As the amount of alkanes could be reflected directly by the alkane/alkene ratios in the products, the ratios of the C_5 products (as representative of the C_5^+ species) under various temperatures were studied and presented in Fig. 3. As can be seen, the C_5 alkane/alkene ratios increased smoothly when the conversion was less than 65% and more rapidly beyond that. At any given conversion level, the C_5 alkane/alkene ratios under various temperatures exhibited almost the same values when the conversion was lower than 65%, and they differentiated from each other when the conversion exceeded 65%. Relatively lower ratios were obtained at higher temperatures. When the conversion was lower than 65% at all temperatures, the C_5 alkane/alkene ratios would be less than 0.08, demonstrating that more than 92 wt% of the C_5 products are C_5 alkenes, which could be recycled as good cracking feed stocks to further enhance the propene production.

4 Discussions

4.1 The Main Reaction Pathways Involved in 1-Butene Cracking

From the product distribution described above, the main reaction pathways were summarized in Scheme 1. The hydrogen atoms in the molecular formula of the hydrocarbons and intermediates were omitted for simplicity. As depicted in Scheme 1, there are mainly seven groups of reactions involved in this process:

1. Isomerization. The feed stocks (1-butene) are protonated on acid sites to form [C_4^+] intermediates, which could desorb as butene isomers (via routes 1a, b).
2. Oligomerization. The [C_4^+] ions oligomerize with alkenes $C_x^=$ (such as $C_3^=$, $C_4^=$ and $C_5^=$) to form larger carbenium ions [C_n^+]. As the feed stocks consist mainly of 1-butene (99.5 wt%), the oligomerization step proceeds mainly as the dimerization of butenes.
3. Cracking. The [C_n^+] intermediates are catalytically cracked to propene and ethene, which are denoted as routes (3a) and (b), respectively.

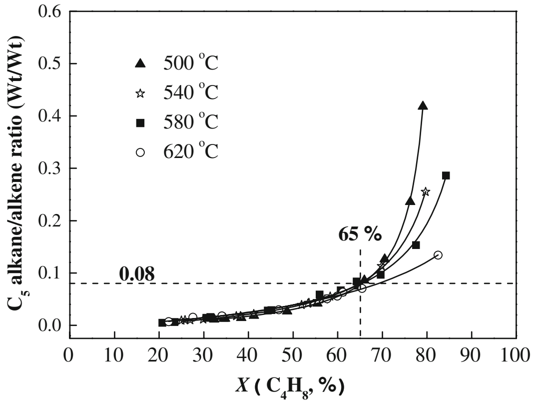
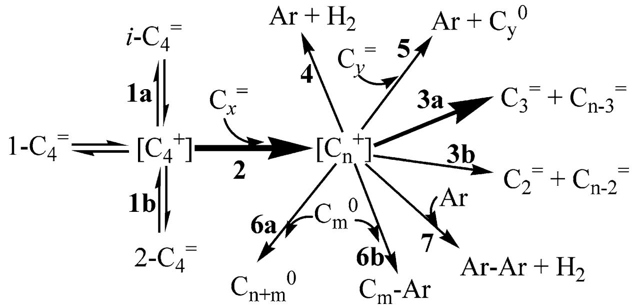


Fig. 3 Evolution of C_5 alkane/alkene ratios with the conversion under various temperatures in the reaction of 1-butene cracking on high silica MCM-22



Scheme 1 Main reaction pathways of 1-butene cracking on high silica MCM-22. Note: $C_i^=$, alkenes with carbon number of i; [C_n^+], carbenium ions with carbon number of n; C_j^0, alkanes with carbon number of j; Ar, aromatics; C_m–Ar, alkylated aromatics; Ar–Ar, polycyclic aromatics

4. Dehydro-aromatization. Although H_2 could be produced in this process, the amount of H_2 was usually less than 0.02 wt% in the products when pre-coked high silica HMCM-22 were used as catalysts, thus H_2 was usually not calculated.
5. Hydrogen transfer reaction. This is the main pathway for the formation of the aromatics and light alkanes (propane and butanes).
6. Alkylation. An alkane molecular C_{n+m}^0 is formed when the [C_n^+] is olefinic (route 6a). If the [C_n^+] is an aromatic carbenium, the product would be an alkylated aromatic (route 6b).
7. Coking. The polycyclic aromatics (Ar–Ar) are considered as coke precursors and the repetition of this reaction leads to coke deposition. In this study, coking amount was usually less than 0.3 wt% of the total products, thus it was usually not calculated, similar with the treatment of the hydrogen.

4.2 Influence of TOS on the Catalytic Performance of HMCM-22

As described in Sect. 3.1, the feed conversion over high silica HMCM-22 decreased rapidly, accompanied by great changes in the product distribution at the initial stage (Fig. 1). This could be explained by rapid coke formation (reaction 7) over acid sites in the 12-MR supercages [6] and part of 12-MR pockets on the external surfaces. During the quasi-plateau stage, the reactions would mainly take place in the 10-MR channels and the undeactivated 12-MR pockets where the coking precursors are not easily formed and deposited [4, 6]. That may be also one reason for the product distribution on HMCM-22 is intermediate between those on HZSM-5 and HY zeolites [18]. The results also demonstrated that acid sites in different pore apertures would bring about different selectivity and stability, in accordance with the results on zeolites with various topolgies in butene cracking process [18].

4.3 Influence of Reaction Temperature on the Product Distribution

As chemical equilibrium of the butene isomers can be easily achieved under the cracking conditions [17, 21], the isomerization reactions (1a) and (b) would have little influence on the product distribution. Then the product distribution is determined mainly by the other 6-group reactions.

In general, the cracking reactions, i.e., (3a) and (b), which account for the formation of propene and ethene, proceed mainly via β-scission of the carbenium ions. Comparatively, the latter is less energetically favorable, owing to the formation of unstable primary carbenium ions [22]. Thus the formation of ethene is less favored at lower temperatures. However, in parallel reactions, the one which has higher activation energy would be more accelerated by raising temperature [23]. So reaction (3b) is promoted to a larger extent with the increasing of the reaction temperature, which brings about lower propene/ethene ratios at higher temperatures (Fig. 2b).

As has been addressed in Sect. 4.1, the formation of aromatics involves mainly two pathways. One is the dehydrogenation route, i.e., reaction (4), which is a strongly endothermic reaction [24]. This reaction is favored at elevated temperatures, which results in the higher selectivities of aromatics at higher temperatures (Fig. 2c). The significant formation of the aromatics at 620 °C also caused a decrement of the propene selectivities compared with those at 580 °C (Fig. 2a) in that large amount of [C_n^+] intermediates were consumed by reaction (4). Unlike reaction (4), the other pathway for the formation of aromatics, i.e., the hydrogen transfer reaction (5), is weakly exothermic and thermally unfavored [24]. Thus this reaction does not contribute to the high selectivities of aromatics at high temperatures.

Besides the contribution to the aromatics, reaction (5) is also the main pathway for the formation of the alkanes. As it is unfavored at higher temperatures, the selectivities of propane plus butanes (Fig. 2d) and the C_5 alkane/alkene ratios (Fig. 3) exhibit lower values at higher temperatures.

C_5^+ species are mainly formed by incompletely cracking of the oligomeric intermediates (3a, b), of course, the contribution of the oligomerization (2) and alkylation (6a) reactions could not be excluded. The cracking reactions are endothermic while the latter two reactions are exothermic [24], thus the increasing of the reaction temperatures promoted further cracking of C_5^+ species (via routes 3a, b) and also inhibited the reactions (2) and (6a), which accounts for the lower C_5^+ selectivities at higher temperatures (Fig. 2f).

Besides catalytic cracking routes, pyrolysis of the intermediates may also exist in the process. Blank experiments carried out in this study indicated that the pyrolysis conversion was usually lower than 10% at 620 °C and no more than 3% at other selected temperatures. Such low conversions mean that catalytic reactions are the main reactions in the process, and that is why pyrolysis reactions are not included in the main reactions discussed in Sect. 4.1. But it gives a clue that pyrolysis would occupy some proportion at relatively high temperatures, which may contribute to the higher selectivities of methane plus ethane at higher temperatures (Fig. 2e) [25].

Besides pyrolysis of the intermediates, the formation of methane and ethane is also related with the dealkylation of the alkylated aromatics [18]. Thus the evolutions of the selectivities of methane plus ethane with the conversion level (Fig. 2e) show obvious similarity with those of the aromatics (Fig. 2c).

From the discussions above, the product distribution is strongly affected by the reaction temperature. The optimal temperature for propene production has to be the one that promotes the catalytic cracking and at the same time inhibits the side reactions to the greatest extent. Combined with the results in Sect. 3.2, 580 °C is undoubtedly the best choice.

4.4 Evolution of Product Distribution with the Conversion Level

In the consecutive reactions of propene formation and consumption, viz. C_4 alkene oligomerization (2), [C_n^+] cracking (3a) and hydrogen transfer reactions (5), propene is an intermediate product. For consecutive reactions, there must exist an optimal contacting time for maximizing the intermediate product on a given catalyst under a defined temperature [23]. In this study, the contacting time is controlled by variations of the space velocity and partial pressure of the feed stocks. As the catalyst status is comparable, long contacting time of the feed stocks and the catalyst corresponds to high conversion level. When the conversion is low (<50%), reaction (3a) proceeds incompletely as the contacting time is too short, leading to a low selectivity of propene (Fig. 2a) and high selectivity of C_5^+ species (Fig. 2f). With the increasing conversion level (contacting time), the cracking of the intermediates is promoted, thus the propene selectivity increases when the conversion level is lower than 65% (Fig. 2a). With further increasing of the contacting time (conversion level > 65%), the chance of bimolecular hydrogen transfer reaction (5) is increased to a larger extent, resulting in a noticeable decrement of propene selectivity (Fig. 2a) and an increment of propane selectivity (Fig. 2d).

Besides the consecutive reactions, there are also some parallel reactions, which have a common characteristic that they consume the intermediates [C_n^+]. Among them, reactions (3b, 4, 5) are the main ones competing with the propene-producing reaction (3a), while other reactions (6a, b) would also have some minor influences.

As has been addressed in Sect. 4.2, the catalytic cracking of C_4^+ alkenes to ethene (3b) is less energetically favorable. However, under relatively higher reaction severity (longer contacting time), more ethene could be formed via further cracking of $C_4^=$ and $C_5^=$ in the 10-MR channels of MCM-22 zeolite [6], resulting in the lower propene/ethene ratios at higher conversion levels (Fig. 2b).

The dehydro-aromatization route for the formation of aromatics, i.e., reaction (4), is essentially a multi-step process, which involves multi-step dehydrogenation and cyclization reactions. Thus the prolongation of the contacting time (increment of the conversion level) directly favors the ultimate formation of aromatics. As has been discussed above, hydrogen transfer reactions (5) are also favored at prolonged contacting time. These reactions account for the high selectivities of aromatics (Fig. 2c) and propane plus butane (Fig. 2d) as well as high C_5 alkane/alkene ratios (Fig. 3) at high conversion levels.

Although the alkylation reactions (6a, b) would also be accelerated by increasing of the contacting time as they are secondary reactions, the reactions in their opposite directions, i.e., the cracking and dealkylation, are promoted to even larger extents. That is incarnated by the decrease of C_5^+ selectivity (Fig. 2f) and the increase of the methane plus ethane selectivity (Fig. 2e) with the increment of conversion level. Thus these reactions do not contribute much to the product distribution.

From the results in Fig. 2, when the conversion level is lower than 60%, the main reactions involved can be divided into three groups, namely the oligomerization/alkylation (2, 6), the cracking (3), and the hydrogen transfer (5) reactions. As the products of hydrogen transfer reactions showed smooth tendencies along the conversion level (see Figs. 2c, d, 3), the selectivity of propene is mostly influenced by the cracking severity of the intermediates. Thus the increasing of contacting time favors the formation of propene. If the conversion exceeds 60%, the decrement of the propene selectivity with the increasing conversion level can be ascribed to the increase of the side reactions (4, 5). Thus the optimal conversion range for enhancing propene would be 50–60% on the high silica HMCM-22.

5 Conclusions

The product distribution and reaction pathways in 1-butene cracking on high silica HMCM-22 zeolite were investigated in detail. It can be concluded that high temperature could promote the cracking reactions and inhibit the hydrogen transfer processes; however, it could also accelerate side reactions such as aromatization and pyrolysis. Deduced from the evaluation data, the most suitable temperature would be 580 °C, the suitable range of the feed conversion would be 50–60% for enhancing the propene production. Under the optimal conditions, the reaction intermediates are well cracked and the main side reactions are inhibited to the greatest extent, and a high selectivity of propene (about 40 wt%) could be obtained.

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References

1. Rubin MK, Chu P (1990) US 4954325
2. Leonowicz ME, Lawton JA, Lawton SL, Rubin MK (1994) Science 264:1910
3. Lawton S, Leonowicz ME, Partridge R, Chu P, Rubin MK (1998) Micropor Mesopor Mater 23:109
4. Matias P, Lopes JM, Laforge S, Magnoux P, Russo PA, Ribeiro Carrott MML, Guisnet M, Ramôa Ribeiro F (2008) J Catal 259:190
5. Corma A, Gonzalezalfaro V, Orchilles AV (1995) Appl Catal A Gen 129:203
6. Zhu XX, Liu SL, Song YQ, Xie SJ, Xu LY (2005) Appl Catal A Gen 290:191
7. Asensi MA, Corma A, Martinez A (1996) J Catal 158:561
8. Laforge S, Martin D, Paillaud JL, Guisnet M (2003) J Catal 220:92
9. Corma A, Corell C, Perezpariente J (1995) Zeolites 15:2
10. Zhu XX, Liu SL, Song YQ, Xu LY (2005) Catal Commun 6:742
11. Zhao GL, Teng JW, Xu N, Tang Y, Xie ZK, Yang WM, Chen QL (2005) Chem J Chin U 26:1140
12. Okumura K, Hashimoto M, Mimura T, Niwa M (2002) J Catal 206:23
13. Guray I, Warzywoda J, Bac N, Sacco A (1999) Micropor Mesopor Mater 31:241
14. Cheng MJ, Tan DL, Liu XM, Han XW, Bao XH, Lin LW (2001) Micropor Mesopor Mater 42:307
15. Liu ZY, Liu ZM, Qi Y, Xu L, He YL, Yang Y, Zhang YY (2004) Chin J Catal 25:542
16. Xu GL, Zhu XX, Niu XL, Liu SL, Xie SJ, Li XJ, Xu LY (2009) Micropor Mesopor Mater 118:44
17. Liu JT, Zhong SQ, Xu CM, Xie ZK, Yang WM (2005) Petrochem Technol 34:9
18. Zhu XX, Liu SL, Song YQ, Xu LY (2005) Appl Catal A Gen 288:134
19. Johnson DL, Nariman KE, Ware RA (2001) US6222087-B1
20. Dath JP, Vermeiren W (2005) EP1508555 A1
21. Xu GL, Zhu XX, Yu HW, Niu XL, Liu SL, Xie SJ, Xu LY (2007) Natur Gas Chem Ind 32:5
22. den Hollander MA, Wissink M, Makkee M, Moulijn JA (2002) Appl Catal A Gen 223:85
23. Fu XC, Shen WX, Yao TY, Hou WH (2005) Physical Chemistry. Higher Education Press, Beijing
24. Zhu XX, Song YQ, Li HB, Liu SL, Sun XD, Xu LY (2005) Chin J Catal 26:111
25. Li L, Gao JS, Xu CM, Meng XH (2006) Chem Eng J 116:155