ISOMERIZATION OF 1-BUTENE CATALYZED BY SURFACTANT-MODIFIED, Al$_2$O$_3$-PILLARED CLAYS

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Abstract—Recent studies indicate that a template method for creating Al-pillared clays, in which surfactant micelles foster the creation of a homogeneous mesoporous network within the pillar, effectively enhance catalyst performance and adsorbent properties. No studies, however, have described the relative effects of the surfactant concentration and Al content on the textural and acidic properties and on the catalytic activity of the Al-pillared clays. The purpose of the present study was to fill this gap, using the isomerization of 1-butene as the test process for catalytic activity. Modified pillared clays (MPC) were prepared from a synthetic clay, TS-1, using different amounts of a non-ionic surfactant (Igepal CO-720) and a fixed concentration of a solution containing the Al polycation [Al$_{13}$O$_4$-(OH)$_{24}$]$^{7+}$. MPC with a fixed amount of surfactant and different amounts of Al were also prepared. The catalysts were characterized by X-ray diffraction (XRD), X-ray fluorescence (XRF), temperature-programmed desorption of ammonia, ^{27}Al magic-angle spinning nuclear magnetic resonance (^{27}Al MAS NMR), and N$_2$ adsorption/desorption isotherms. Isomerization of 1-butene at 250ºC was used to test the catalytic activity. Analyses by XRD and XRF showed that the synthesized solids were amorphous and that the amount of pillaring by Al increased with the amount of Al complex used. Interestingly, the surface area and pore volume were directly proportional to the amount of surfactant employed and decreased with increasing amounts of Al pillaring. All solids showed activity for 1-butene isomerization, with a maximum conversion of ~75%. Only *cis*- and *trans*-2-butene were observed. The absence of isobutene suggested that acid sites of moderate strength were formed, in agreement with the results obtained from the desorption of ammonia.

Key Words—Isomerization of 1-butene, Mesoporous Clays, Modified Pillared Clay (MPC), PILC.

INTRODUCTION

The use of clay minerals as catalyst supports is of much interest because of their great abundance, small cost, and specific properties (Gil *et al.*, 2000; Corma, 1997; Vaccari, 1998, 1999). Acid activation and exchange with inorganic, organic, or organometallic cations are the main methods employed to obtain a wide range of materials with potential applications in adsorption processes or catalysis. The incorporation of large inorganic cations into the interlamellar space of the clay leads to the formation of metal oxide pillars after calcination, which improves the surface area available and the structural integrity. The materials have the added advantage that the pillars themselves may be catalytically active. In particular, montmorillonites and its pillared derivatives have been used in hydrogenation and dehydrogenation reactions (Gil *et al.*, 2000; Vaccari, 1999; Moronta *et al.*, 2006).

Templated synthesis of mesoporous materials is of interest because it facilitates the formation of a continuous, homogeneous network of mesopores in a solid matrix. It can be achieved by first adding a surfactant to an aqueous medium, which forms micelles that orient into a well-defined structure or template, then by adding a source for a solid matrix, such as alumina or silica, which condenses around the micelles and generates a matrix embedded within the organic surfactant or template. Usually the organic template is removed by calcination to form a continuous and homogeneous network that mimics the size and shape of the micellar template. The advantages of this synthesis are that the pore volume is controlled by the volume fraction of the template constituents and the pore size is controlled by the size of the surfactant micelles, leading to materials with large surface area, uniform mesoporous structure, and large pore volume (Hwang *et al.*, 2001; Zhu and Lu, 2001).

Using the template approach, two new families of mesoporous solids synthesized from clays have been reported in recent years. Polverejan *et al.* (2000, 2002), Benjelloun *et al.* (2001), Vansant and Cool (2001), and Chmielarz *et al.* (2006, 2007) reported a new material referred to as 'porous clay heterostructures' (PCH), synthesized by treating a clay prepared using cetyltrimethylammonium bromide with tetraethyl orthosilicate (TEOS), employing dodecylamine as co-surfactant and co-solvent. This group of porous materials is characterized by combined micro and mesopore structure, a very large surface area, large porosity, and good thermal stability. The second class of materials, referred to as

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"composite clay nanostructures," was reported by Zhu and Lu (2001) and by Zhu *et al.* (2002). These are synthesized in a similar way to that of pillared clays (PILC), employing non-ionic surfactants to orient a well-defined structure. Their apparent advantage over the PCH is the use of low-cost surfactants and the greater versatility of the preparation method for the incorporation of other metals. However, few studies in the literature have reported on either of these types of materials.

The large surface areas and the pore structure of the clay composites provide significant potential for these solids to be used as catalysts and adsorbents. In this regard, Ni catalysts supported on clay nanocomposites have shown good catalytic activity (conversions of 90–95%) for the carbon dioxide reforming reaction with methane to synthesis gas (Hwang *et al.*, 2001; Moronta *et al.*, 2005a). In addition, good catalytic activity (conversion of 50%) was obtained for the dehydrogenation of ethylbenzene to styrene over Pt catalysts supported on clay nanocomposites (Morán *et al.*, 2007).

Skeletal isomerization of linear butanes to isobutylene is an interesting reaction from an industrial point of view because the product is used in the production of octane-enhancing fuel additives. Zeolites and microporous molecular sieves, such as ZSM-22 and SAPO-11, are good catalysts for the selective skeletal isomerization of *n*-butanes (Méariaudeau and Naccache, 1999). This is due to the sufficient Brönsted acid function and shape-selective pore structure. The product distribution of the catalytic 1-butene isomerization is widely used to evaluate the acidity of heterogeneous catalysts. The product distribution is divided into the following groups: (1) double-bond isomerization (*cis*-2-butene and *trans*-2-butene); (2) skeletal isomerization products (isobutene); (3) hydrogenated products (*n*-butane and isobutene); (4) cracking products (C4-) (methane, ethane, ethylene, propane, and propene); and (5) surface carbon residues and greater-molecular-weight compounds. Modified natural clays are also good catalysts for this reaction. Moronta *et al.* (2002) reported that 1-butene gives good yield to *cis*- and *trans*-2-butene over acid-activated organoclays when they are first exchanged with the tetramethylammonium chloride (TMACl) and then acid activated (maximum conversions of 70%), whereas a decrease in the catalytic activity was observed by employing the inverse procedure (maximum conversions of 51%). Such behavior was due to the fact that the organo-cation content was greater than the proton population in the interlayer space. Moronta *et al.* (2005b) isomerized *cis*-2-butene and *trans*-2-butene over two commercial, acid-activated clays exchanged with different cations (Al^{3+}, Cr^{3+}, Fe^{3+}, and Ni^{2+}). The order of activity for acid- and other exchanged forms was $H^+ > Al^{3+} \cong Ni^{2+} > Cr^{3+} > Fe^{3+}$ with a maximum yield of 75% for isomerization of *cis*-2-butene and 53%

for isomerization of *trans*-2-butene. However, no study to date has been published on the isomerization of 1-butene over surfactant-modified, Al_2O_3-pillared clays. The objective of this work, therefore, was to study the effect of Al and surfactant on the textural and acidic properties and on the catalytic activity in the isomerization of 1-butene of Al_2O_3-pillared clays.

MATERIALS AND METHODS

Preparation of surfactant-modified, Al_2O_3-pillared clays

A synthesized smectite-type clay TS-1 (a tri-octahedral Mg clay, referred to hereafter as TS), obtained from CO-OP Chemical Co., Inc., Japan, was used as received.

The Al complex $[Al_{13}O_4(OH)_{24}(H_2O)]^{7+}$ was synthesized by basic hydrolysis of $AlCl_3$ using the procedure described by Bradley *et al.* (1992).

The surfactant-modified, Al_2O_3-pillared clays were prepared using a similar procedure to that described by Zhu and Lu (2001). 2.5 g of TS clay was dispersed in 40 mL of de-ionized water and stirred for 1 h. Next, 0, 1, 3, or 5 g of surfactant (Igepal CO-720, supplied by Aldrich) was added to the clay suspension. Stirring was maintained for 2 h to allow sufficient mixing. To this mixture, 25 mL of the Al complex solution was added, drop-wise, with continuous stirring for 2 h. Samples with a fixed amount of surfactant (5 g) and different amounts of the Al complex solution (12.5 mL and 50 mL) were also prepared. The suspension was transferred to an autoclave and kept at 100ºC for 48 h. A solid precipitate was recovered from the mixture by centrifugation and washed with de-ionized water. The solid collected was dried at 75ºC overnight and calcined at 500ºC for 20 h ($\beta = 2$ºC/min). The samples were identified according to the amount of Al and surfactant added, as follows: MPC-0S25Al, MPC-1S25Al, MPC-3S25Al, MPC-5S25Al, MPC-5S12.5Al, and MPC-5S50Al, *e.g.* MPC-1S25Al indicates that this sample was prepared using 1 g of surfactant and 25 mL of the Al-complex solution.

Characterization techniques

The surface areas of the starting TS clay and the surfactant-modified, Al_2O_3-pillared clays were measured by N_2-adsorption isotherms at −196ºC, conducted on a Micromeritics ASAP 2010 porosimeter. Samples were outgassed at 300ºC and 0.13 Pa for 2 h before taking measurements. The surface area was calculated using the BET method (S_{BET}). The total pore volume (V_t) was calculated from the amount of adsorption at a relative pressure close to unity.

The bulk chemical composition was measured by XRF using a Shimadzu EDX-700 HS. The XRD measurements of the modified pillared clays were recorded on a Bruker D8 Focus diffractometer, operating at 40 kV and 40 mA at a scan rate of 2º/min, using CuKα radiation. The crystalline phase was identified using the

tabulated powder diffraction files of the International Center Diffraction Data (ICDD) *d*-value files.

Solid-state NMR spectra were recorded on a Bruker Avance 300 spectrometer. The ^{27}Al MAS NMR spectra were measured at 78.2 MHz, with radio frequency pulses of 4 μs, equivalent to a 90° pulse angle, a spinning rate of 5000 rpm, and 1 s recycle delays. Chemical shifts are quoted in parts per million (ppm).

The acidity was evaluated by the adsorption of ammonia at room temperature in samples pre-treated at 300ºC to compare the acid strength of the catalysts prepared with the conversion of 1-butene. For this experiment, 50 mg of sample was placed in a U-shaped quartz reactor and dried at 120ºC in N_2 flow. Afterwards, pulses of ammonia were sent up to saturation, and the thermal desorption was carried out from 35 to 600ºC. A thermal conductivity detector was used to measure the ammonia removed.

The catalytic activity of the surfactant-modified, Al_2O_3-pillared clays was evaluated by monitoring the isomerization of 1-butene (Linde, CP grade) as a probe reaction. The experiments were carried out in a stainless steel reaction line. The reactor consisted of a U-shaped Pyrex tube of 6 mm outer diameter and 4 mm inner diameter. Products analysis was carried out on-line using a gas chromatograph (HP series II) equipped with a flame ionization detector and a capillary column (30 m × 0.053 mm inner diameter Altech AT-alumina). The column was kept at 80ºC. A loop of 0.025 cm^3 was used as a calibrated volume. The reactor was loaded with 100 mg of catalyst and heated to 250ºC for 1 h at 10ºC/min under a flow of 30 cm^3/min of dry He; then, pulses of the 1-butene (in He flow) were sent to the reactor at 250ºC.

RESULTS AND DISCUSSION

The chemical composition of the starting TS clay, as determined by XRF, was 67.7% SiO_2, 28.0% MgO, and 4.3% Na_2O (Table 1). Chemical compositions also confirmed that, by increasing the Al content in the synthesis gel and keeping the amount of surfactant constant, the Na cations were displaced by Al cations to form Al_2O_3-pillared clays after calcination. The incor-

poration of Al_2O_3 increased from 6.6 to 20.6 wt.%. When Al was kept constant and the surfactant content varied, the amount of Al_2O_3 incorporated changed slightly to 11.7 wt.% and 13.5 wt.%. When the cation concentration in the synthesis gel was increased, a greater degree of cation exchange took place, as shown by the percentage of Al_2O_3 incorporated, while a greater concentration of surfactant does not affect the cation-exchange process. These results are in agreement with those reported by Deng *et al.* (2003), who stated that the cation exchange capacity (CEC) of the clay was not affected by the incorporation of non-ionic surfactant molecules, and only an interaction of the clay layers with the ethylene oxide groups exists, keeping the same availability of exchange sites.

Variation in the amount of Al and surfactant affected the textural properties of the synthesized solids (Table 1) by increasing the surface area by 20−30% and pore volume by 67−79% compared with the raw TS clay. Varying the Al content while holding the amount of surfactant constant (5 g), however, led to a maximum surface area of 616 m^2/g. When using the smallest amount of Al, the surface area decreased slightly by 4−8%, possibly due to the formation of large polyoxyl Al ions which congest the interlayer space of the intercalated clay. Similar behavior was observed by Zhu and Lu (2001).

The d_{001} value from the XRD pattern (Figure 1) of the raw TS clay calcined at 500ºC was 16 Å. Other well-defined XRD peaks corresponded to the 110, 201, and 060 planes of MgO-SiO_2 species. In the pillared form of TS (MPC-0S12.5Al), a noticeable increase in the d_{001} value to 19 Å was observed, indicating the formation of Al_2O_3 pillars in the interlayer space. Surfactant-modified, Al_2O_3-pillared clays showed no clear d_{001} value, which is attributed to the lack of long-range order in the structure of these solids (Zhu and Lu 2001; Moronta *et al.*, 2005a). Similarly, Bisio *et al.* (2008) prepared a series of synthetic saponites and attributed the absence of the d_{001} signal, at temperatures close to 500ºC, to the loss of laminar order. The structural collapse and smaller particle sizes observed in synthetic *vs.* natural clays may account for the absence or low intensity of the d_{001}

Table 1. Chemical composition, surface area, and pore volume of the starting TS clay and prepared catalysts.

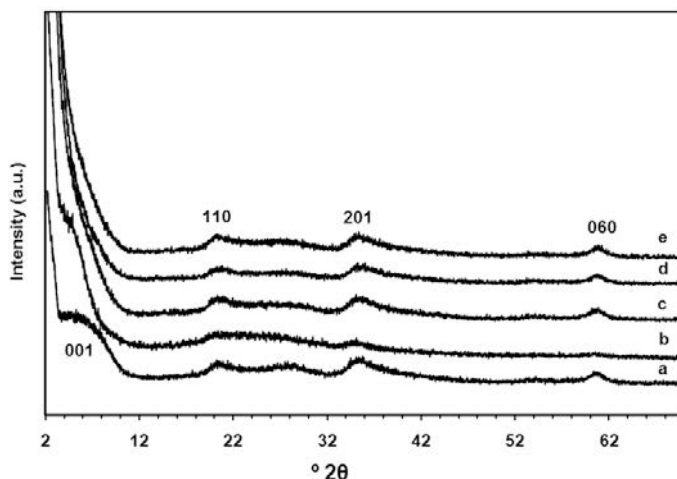| Catalysts	SiO_2 (wt.%)	MgO (wt.%)	Al_2O_3 (wt.%)	Na_2O (wt.%)	S_{BET} (m^2/g)	V_t (cm^3/g)
TS	67.7	28.0	0	4.3	488	0.288
MPC-0S25Al	63.1	25.2	11.7	0	477	0.374
MPC-1S25Al	62.5	25.3	12.2	0	546	0.495
MPC-3S25Al	63.0	25.0	12.0	0	553	0.482
MPC-5S25Al	62.5	24.0	13.5	0	591	0.515
MPC-5S12.5Al	66.5	26.9	6.6	0	616	0.488
MPC-5S50Al	57.4	22.0	20.6	0	565	0.585

Figure 1. XRD patterns for (a) TS calcined at 500ºC, (b) MPC-0S12.5Al, (c) MPC-3S12.5Al, (d) MPC-5S12.5Al, and (e) MPC-5S25Al catalyst.

diffraction peak. In the present case, the incorporation of alumina and surfactant molecules in the interlayer space may also contribute to the absence of the d_{001} signal, because they favor the formation of highly disordered solids. Zhu and Lu (2001) and Zhu *et al.* (2006) ascribed the absence of diffraction signals to instrumental limitations because of the small particle size of the materials studied.

N$_2$ adsorption-desorption isotherms of the starting TS clay and its derived MPC (Figure 2) revealed that the volume of N$_2$ adsorbed for TS is less than that of the modified Al$_2$O$_3$-pillared clays calcined at 500ºC for 48 h. The removal of the surfactant contributes to a noticeable increase in the porosity of the MPC and, thus, the volume of N$_2$ adsorbed. The starting TS clay exhibits a type IV isotherm with an hysteresis loop of type H4, while MPC-1S25Al, MPC-3S25Al, and MPC-5S25Al also produced isotherms of type IV with much greater volumes of adsorption by mesopores. For samples prepared with variable amounts of Al and fixed amounts of surfactant, no significant changes in the adsorption

isotherm were observed as the Al content was increased in the MPC (Figure 3). The hysteresis loop seems to be of type H4, usually found in solids consisting of aggregated or agglomerated particles. The mesopores for the starting TS clay are widely attributed to slit-shaped pores (plates or particle edges like cubes), while the new mesopores (in the modified clay with surfactant) are due to the space generated by removal of surfactant during calcination, in which the length of the chain of the surfactant molecule controls the pore size and the amount of surfactant is proportional to the total pore volume in the solids synthesized.

The pore size of the MPC tends to be ~2.5 nm and increasing the amount of surfactant increases the pore-size distribution slightly (Figure 4). The slight increase in pore size is attributed to a greater degree of agglomeration of the surfactant micelles; no marked effect of the surfactant concentration on the pore-size distribution was observed and the number of pores with diameters between 4 and 13 nm increased only sightly, in contrast to the findings of Zhu and Lu (2001). Large
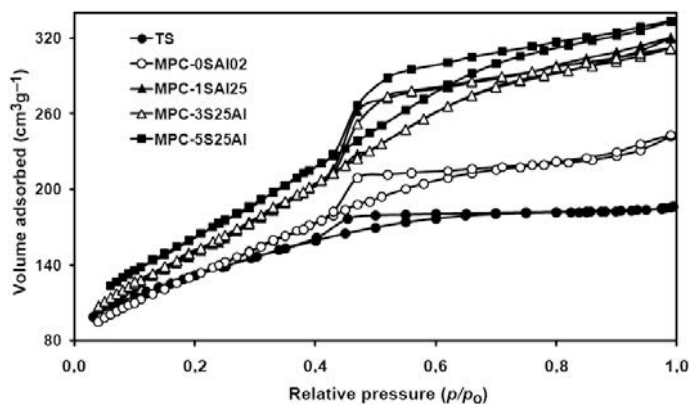

Figure 2. Nitrogen adsorption-desorption isotherms of TS and its derived MPC synthesized with different amounts of surfactant and a fixed Al content.
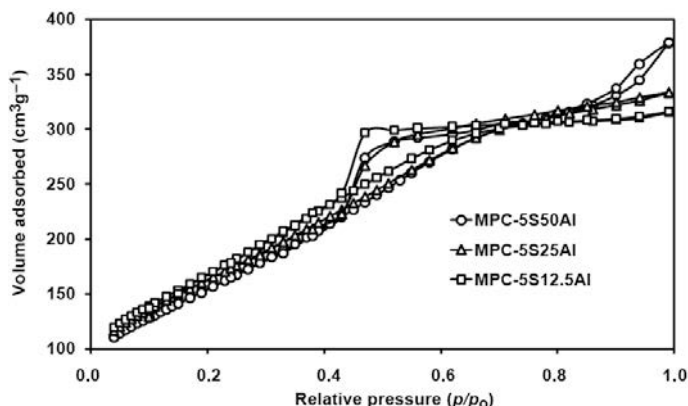
Figure 3. Nitrogen adsorption-desorption isotherms of MPC prepared with a fixed amount of surfactant and different Al contents.

surfactant concentrations guarantee the swelling of the clay layers to form Al_2O_3-pillared clays, but involve more costly experimental conditions during the removal of surfactant. However, relatively small concentrations of surfactant improve considerably the textural properties of the materials synthesized and allow clean surfaces to be obtained. The increase in the surface area, even at small surfactant concentrations, is an indirect measurement of the surfactant-clay interaction. Zhu and Lu (2001) observed a decrease in the surface area as the number of ether groups (n) of the surfactant increased and ascribed this trend to the fact that the miscibility of the surfactant in water increases with n, favoring greater dispersion in water and greater susceptibility to surfactant loss during the washing process. The amount of large-n surfactant remaining in the intercalated solid before calcination is, therefore, less than that of the surfactant with smaller n. The hydrophile-lipophile balance (HLB) is expected to play a key role in determining if surfactant dispersion stabilizes it in solution by forming micelles and/or micelle aggregates, or by interaction with the clay particles. In spite of the small concentrations used in this work, the increase in the surface area is attributed to the fact that the nonyl phenyl group is very voluminous with a high hydrophilic character and, thus, favors micelle formation and allows ethyl chains to interact with water molecules and the clay layers.

The most plausible mechanism for the synthesis of surfactant-modified, Al_2O_3-pillared clays appears to be that the surfactant molecules form micelles in the interlayer region because of the poor solubility in water and the strong affinity for the clay surface. The Keggin ions replace the interlayer Na ions and polymerize further to form larger pillars which are separated by the micelles. The micelles act as templates, preventing the intercalated framework from collapsing during the dehydration process. Meanwhile, the framework hardens, and removal of the surfactant at 500ºC results in a disordered mesoporous structure of alumina-pillared clay (Figure 5).

The ^{27}Al MAS NMR spectra for MPC-5S12.5Al, MPC-5S25Al, and MPC-5S50Al (Figure 6) consisted of peaks attributed to Al in tetrahedral and octahedral coordination, respectively, at 60 and 0 ppm. Another small peak at ~120 ppm was ascribed to a spinning side band. The spectrum for starting TS clay contained no such peak because this sample contains no Al in the
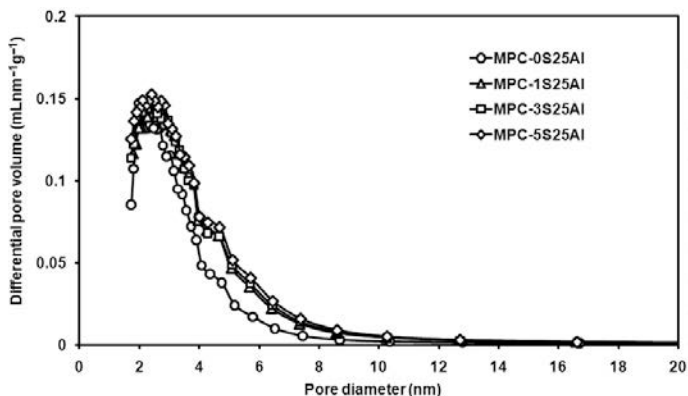

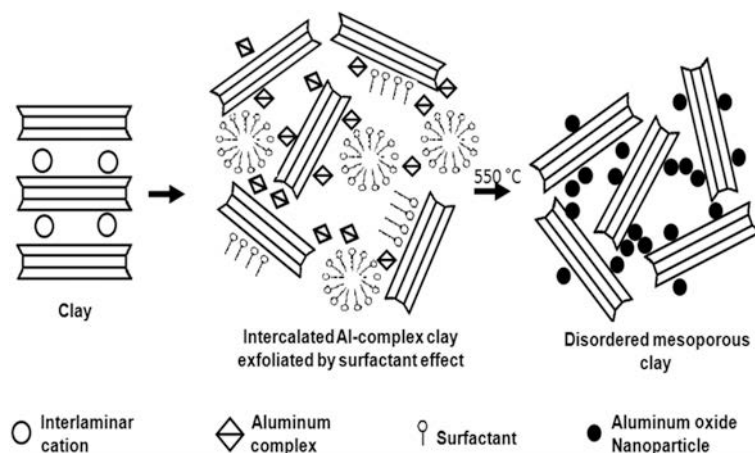Figure 4. Pore-size distribution for MPC prepared with different surfactant contents.

Figure 5. Schematic representation of the structure of modified, porous Al_2O_3-pillared clay.

structure (Table 1). Samples prepared with varied amounts of surfactant and fixed amounts of Al gave similar ^{27}Al MAS NMR spectra (not shown). The intensity of the Al peak in tetrahedral coordination was five times greater than that of the Al peak in octahedral
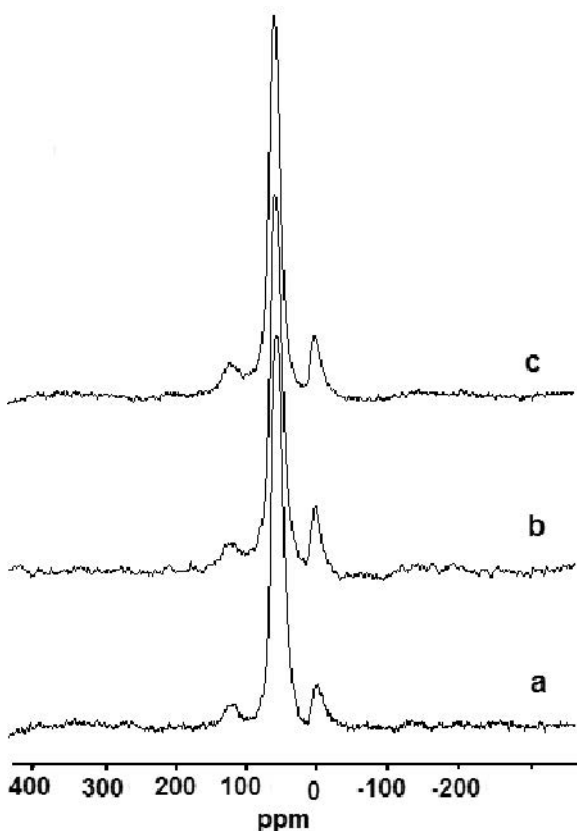

Figure 6. ^{27}Al MAS NMR spectra of MPC prepared with a fixed amount of surfactant and different Al contents: (a) MPC-5S12.5Al, (b) MPC-5S25Al, and (c) MPC-5S50Al.

coordination. The Al clearly is mainly located in exchange sites and grafted onto the silica surface. The Al in octahedral coordination is incorporated by substitution of Mg in the octahedral sheet (Sanz, 2006).

Temperature-programmed desorption (TPD) of ammonia (Figure 7) from the catalysts prepared with fixed amounts of surfactant and varied amounts of Al clearly revealed that an increase in the Al content modifies the surface acidity. Two characteristic desorption peaks were observed: one at low temperature ($120-160ºC$) (LT), and the other at high temperature ($160-350ºC$) (HT). The HT is related to NH_3 desorption from Brönsted acid sites (Si-OH/Al) (Topsøe *et al*., 1981). The LT peak is due to the physical desorption of ammonia along with interaction of the base with weak acid sites, *i.e.* terminal silanol groups, extraframework Al cations, and cation residues. Using the intensity of the HT peak, the order of acid strength was MPC-5S50Al > MPC-5S25Al > MPC-5S12.5Al. No significant differences were detected in the TPD profiles (not shown) for samples prepared by varying the surfactant content, because the Al incorporated was almost the same in all catalysts (Table 1) and no adsorption/desorption of ammonia was detected in the starting TS clay.

The activity for the isomerization of butanes is believed to be due to the presence of acid sites on the catalyst surface. Significant catalytic activity can result from (1) a large number of acid sites, or (2) the acid strength of the site (Pines and Haag, 1960). Isomerization of 1-butene was, therefore, performed at $250ºC$ to test the acid strength of the catalysts prepared. Figures 8 and 9 show the product distribution for isomerization of 1-butene over MPC prepared by varying the surfactant and the Al contents; the total conversion is also included. Only isomers *cis*-2-butene and *trans*-2-butene were detected, indicating the formation of acid sites of moderate strength, in agreement with the results obtained by TPD of ammonia. The starting TS
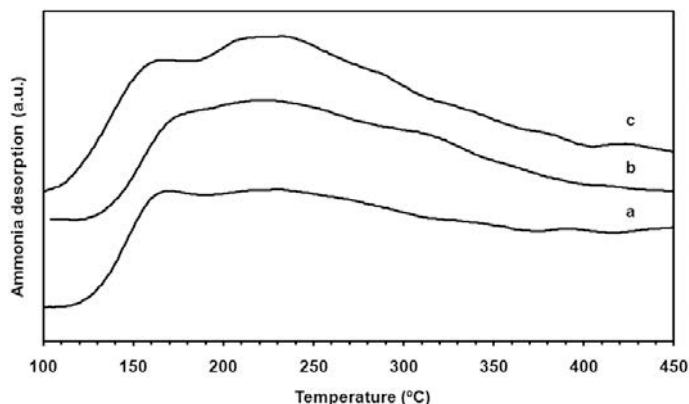
Figure 7. Ammonia TPD profiles for MPC prepared with a fixed amount of surfactant and different Al contents: (a) MPC-5S12.5Al, (b) MPC-5S25Al, and (c) MPC-5S50Al.

clay was inactive with respect to the isomerization reaction. The order of activity for the MPC prepared with different amounts of surfactant and fixed amounts of Al was: MPC-1S25Al > MPC-3S25Al > MPC-5S25Al > MPC-0S25Al, while the order in samples obtained using different amounts of Al and a fixed amount of surfactant was MPC-5S12.5Al < MPC-5S25Al < MPC-5S50Al. The performance for the latter samples is correlated with the acidity obtained after calcination of the modified pillared clays. For samples prepared at small Al contents, the selectivity for cis-2-butene and trans-2-butene isomers was similar, though for samples prepared with greater Al contents, the selectivity increased toward the trans-2-butene product. Moronta et al. (2005b) studied the isomerization of 1-butene over two commercial, acid-activated clays exchanged with Al^{3+}, Cr^{3+}, Fe^{3+}, and Ni^{2+} and observed a greater selectivity toward the cis-2-butene than for trans-2-butene, in line with the thermodynamic stability of the trans isomer. The fact that more sites are available in the MPC with large Al contents could favor the isomerization of cis-2-butene over trans-2-butene. The MPC

prepared here have a better catalytic performance than that reported for the same reaction at higher temperatures over pure alumina (34.8%), silicated alumina (43.5%), silica alumina (40.5%), H-ferrierite (FER, 55.4%) (Trombetta et al., 1998), and acid-activated metakaolin (Lenarda et al., 2007), but they are less active than zeolite HZSM5 (87.7%) (Trombetta et al., 1998).

The total conversions for MPC-1S25Al and MPC-3S25Al were greater than those of MPC-0S25Al and MPC-5S25Al. The greater catalytic activity for the solids synthesized with surfactant is ascribed to the increase in surface area. Even though the catalyst MPC-5S25Al presented a greater surface area than that of MPC-0S25Al, their activities were similar. The low activity observed in MPC-5S25Al may be due to the fact that this particular catalyst was prepared with the largest amount of surfactant, which leads to the formation of carbonaceous deposits on the surface after calcination. To prove this statement, a portion of this sample was calcined again at 500ºC for 8 h under a flow of air (30 mL/min) and an increase in the amount converted in
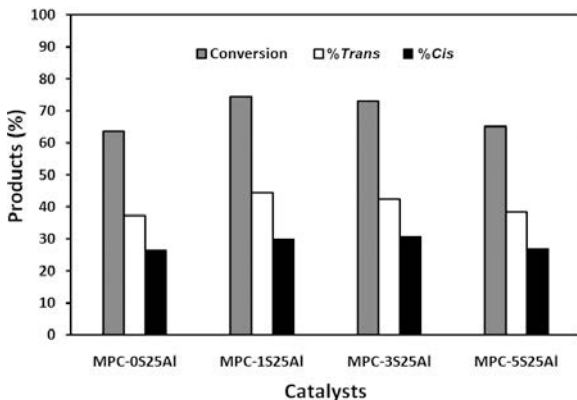

Figure 8. Product distribution for isomerization of 1-butene over MPC prepared with a fixed amount of Al and different surfactant contents.
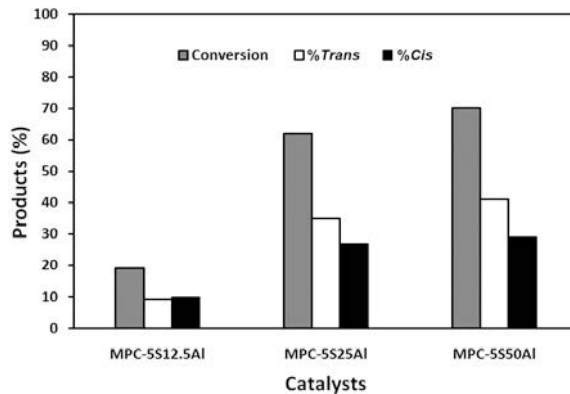

Figure 9. Product distribution for isomerization of 1-butene over MPC prepared with a fixed amount of surfactant and different Al contents.

the first reaction pulse was detected, indicating that a proportion of active sites are deactivated quickly at the beginning of the reaction. To support this statement further, Moronta *et al.* (2005a) carried out thermogravimetric analysis of the TS sample modified with MgO and 5 g of surfactant (Igepal CA720) calcined at 500ºC for 48 h. The desorption/breakdown of the surfactant in the 200−420ºC region disappeared completely after calcination and only those peaks attributed to loss of physisorbed water (produced by rehydration) at 35−280ºC and dehydroxylation combined with CO_2 evolution (at 430−720ºC) were observed. In the latter peak, the CO_2 was generated by carbonaceous deposits not desorbed after calcination, and the weight loss in the region 430−720ºC was 2.8%. Therefore, a smaller weight loss in samples prepared with 1 g and 3 g of surfactant would be expected, confirming that the carbonaceous phase affects the catalytic activity.

CONCLUSIONS

Mesoporous solids with excellent textural properties were synthesized from a synthetic clay and a non-ionic surfactant. In the preparation of modified Al_2O_3-pillared clays, large concentrations of polymeric cations led to diminution of the surface area by oversaturation of the interlamellar space, as in the synthesis of PILC. The use of small concentrations of surfactant caused an increase in the surface area as well as the pore volume. A wide distribution of acid sites of moderate strength was observed in the desorption profiles of ammonia and correlated with the conversion of 1-butene. The use of large amounts of surfactant produced surfaces with carbonaceous deposits that reduced the catalytic performance.

ACKNOWLEDGMENTS

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