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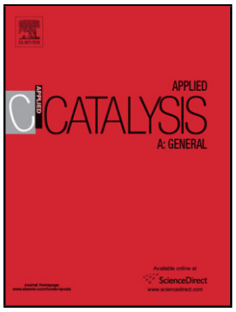

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Authors: Sreenivasulu Peta, Tao Zhang, Viktor Dubovoy, Katherine Koh, Maocong Hu, Xianqin Wang, Tewodros Asefa

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Template-free synthesis of highly selective amorphous aluminosilicate catalyst for toluene alkylation

Sreenivasulu Peta,[a,b,c] Tao Zhang,[b] Viktor Dubovoy,[a] Katherine Koh,[a] Maocong Hu,[d] Xianqin Wang,[d] Tewodros Asefa[a,b,c,*]

[a] *Department of Chemistry and Chemical Biology, Rutgers, The State University of New Jersey, 610 Taylor Road, Piscataway, New Jersey 08854, USA*

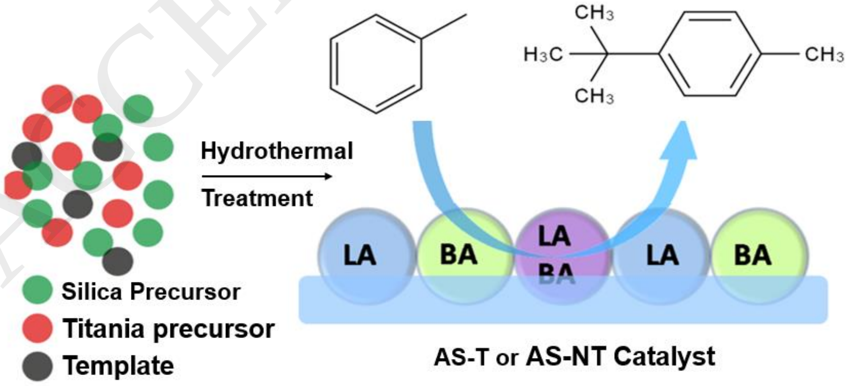
[b] *Department of Chemical and Biochemical Engineering, Rutgers, The State University of New Jersey, 98 Brett Road, Piscataway, New Jersey 08854, USA*

[c] *Institute for Advanced Materials, Devices and Nanotechnology, Rutgers, The State University of New Jersey, 607 Taylor Road, Piscataway, New Jersey 08854, USA*

[d] *Department of Chemical, Biological and Pharmaceutical Engineering, New Jersey Institute of Technology, University Heights, Newark, NJ 07102, USA*

[*] Corresponding author: E-mail: tasefa@chem.rutgers.edu (T. Asefa)

Graphical abstract



Highlights

- A simple template-free synthetic method to make aluminosilicate is reported.
- Amorphous, nanoporous aluminosilicate catalyst is synthesized.
- The catalyst shows high selectivity for toluene alkylation.
- The material catalyzes alkylation of toluene by *tert*-butyl alcohol under mild condition.
- The catalyst gives exclusively 4-*tert*-butlytoluene (*ca*. 99%) as product.

Abstract

In this paper, we report a simple template-free synthetic method that produces amorphous, nanoporous aluminosilicate and highly selective catalyst for toluene alkylation. The structure and composition of the material and its corresponding control material are characterized by various techniques. The material shows excellent catalytic selectivity for alkylation of toluene by *tert*-butyl alcohol in temperatures between 50-150 °C under atmospheric pressure, producing exclusively 4-*tert*-butlytoluene (*ca*. 99%).

Keywords: Aluminosilicate; Toluene Alkylation; Alkylation reaction; 4-*Tert*-butlytoluene; Selective catalyst

1. Introduction

Aluminosilicates have long been used as catalysts in many types of reactions in the chemical industries and refineries to produce various products [1]. Aluminosilicates have also been recognized as having potential applications as adsorbents/membranes for scavenging various pollutants from the environment and CO_2 from flue gases [2]. Aluminosilicates find these diverse applications due to their high surface areas, large pore volumes, uniform microporous channels, acidic sites, and remarkable hydrothermal and thermal stabilities [3-5]. For aluminosilicates to serve efficiently for many of these applications, particularly for catalysis of aromatic alkylation, isomerization and related reactions, which are widely carried out in petrochemical refinery processes, they need to have microporous structures and be shape/size selective. Therefore, many aluminosilicates are made using small molecules as templates [6]. However, the microporosity of aluminosilicates can sometimes compromise their performances in these applications, especially during catalytic reactions involving bulkier reactants, intermediates, and products. Consequently, their mesoporous counterparts—materials that allow better mass transport during catalysis—are sometimes sought and preferable [7, 8]. Hence, in recent years, there have been a great deal of interest to develop mesoporous and micro-mesoporous aluminosilicate materials, whose pores can let bigger reactants and products diffuse in and out of the catalytically active sites better and help reactants transform to products faster during various reactions [9-11].

Traditional method of synthesis of aluminosilicates is based on co-precipitation of silica precursors such as Na_2SiO_2 and aluminium salts and alkoxides. While this method has been used to produce various catalysts, it often involves costly and sometimes toxic, organic templates and energy-intensive solvothermal, microwave heating, and hydrothermal synthetic conditions [12, 13]. Furthermore, the materials need to be calcined at high temperature to remove the organic templates used to create the pores in the materials, which however produces harmful gaseous substances. Thus, facile, scalable synthetic methods that can produce mesoporous aluminosilicates are still needed. In particular, synthetic methods that can produce aluminosilicates with pores that allow reactants to undergo faster mass diffusion and catalytic transformations in the materials are desired [14, 15]. Thus, such materials are of huge importance for catalysis of many industrially relevant reactions.

4-*tert*-Butyltoluene (4-TBT) is a commercially important aromatic compound that is useful for making substances such as 4-*tert*-butylbenzoic acid, 4-*tert*-butylbenzaldehyde, etc., compounds that serve as precursors for various pharmaceuticals, alkylated resins and fragrances and as polymerization regulators for polyesters [16]. Currently, 4-TBT is produced in the chemical industry using homogeneous catalysts such as phosphoric acid, sulphuric acid, and boron triflouride. However, these catalysts have drawbacks associated with their extreme safety and handling requirements and corrosive wastes. To overcome these issues, new synthetic methods that can involve no/less toxic, costly organic templates and produce recyclable mesoporous aluminosilicate catalysts with better mass diffusion properties for reactants and products alike are needed [16].

To this end, herein we report a simple template-free, hydrothermal synthetic method that yields amorphous, mesoporous aluminosilicate material that exhibits excellent catalytic selectivity toward toluene alkylation reaction with *tert*-butyl alcohol. The material is found to catalyze the reaction, selectivity yielding (99 %) 4-TBT as product. To the best of our knowledge, this degree (99 %) selectivity to a 4-TBT product by this reaction has never previously been achieved with laboratory-synthesized aluminosilicate catalysts.

2. Experimental Section

2.1. Chemicals and Reagents

Aluminium isopropoxide, tetraethyl orthosilicate (TEOS), tetrapropylammonium bromide (TPABr), 2-propanol, toluene, and *tert*-butanol (TBA) were purchased from Sigma-Aldrich, and they were all used without further purification.

2.2. Synthesis of Mesoporous Aluminosilicates

In typical synthesis of the mesoporous aluminosilicate catalyst, 0.4 g (2 mmol) of aluminium isopropoxide was mixed with 20 mL of 2-propanol, and the mixture (without any template) was then stirred overnight at 40 °C. Into the resulting solution was added 10.4 g (50 mmol) of TEOS and 25 mL water. The solution was transferred into an autoclave, sealed, and heated at 130 °C for 3 days. It was then filtered, and the solid product was separated and washed with copious amount of ethanol. After letting it to dry in oven at 100 °C, it was calcined at 550 °C for 5 h. The resulting solid material was denoted as AS-NT (to represent "aluminosilicate synthesized using no template"). By using 0.2 g TPABr as template under otherwise similar procedure, aluminosilicate, which was denoted as "AS-T" to represent "aluminosilicate synthesized using template", was produced. This material was used for comparative studies.

2.3. Catalytic Toluene Butylation

In a typical procedure for catalytic toluene butylation reaction, 1 mL of toluene was mixed with 2 mol of TBA and 0.2 g of catalyst. The mixture was placed in a 25 mL Teflon-lined Parr reactor and sealed tightly. The reaction was conducted at 50, 100 or 150 °C for 9 h. At the end of the reaction, the catalyst was separated by filtration, and the products were analyzed with gas chromatography (GC, Agilent 6850) instrument equipped with HP-1 capillary column (30 m length, 0.32 mm id, 0.25

μm film thickness) and flame ionization detector (FID). The products were identified using GC-MS 5890 Series II equipped with 5972 Series mass selective detector. After completion of the reaction, the catalyst was recovered from the reaction mixture *via* centrifugation and washed thoroughly with acetone, and its reusability was tested in multiple cycles.

3. Results and Discussions

The aluminosilicate materials obtained above were both characterized by various analytical techniques. Elemental analysis as well as elemental mapping on the materials conducted using energy dispersive X-ray (EDX) spectroscopy (Figure S1 in Supporting Information) showed that both materials had Al, O and Si atoms. The morphologies of both AS-NT and AS-T materials were analyzed using field emission scanning electron microscopy (FE-SEM) (Figures 1a and 1b). The images showed that both materials possessed sub-micron-sized particles and nanoporous structures. The transmission electron microscopy (TEM) images (Figures 1c and 1d) showed that both materials had some pores that appeared to be due to the void spaces created in between the particles. Further characterization of the materials by wide-angle X-ray diffraction (XRD) indicated that the frameworks in the materials were amorphous (Figure 2a).

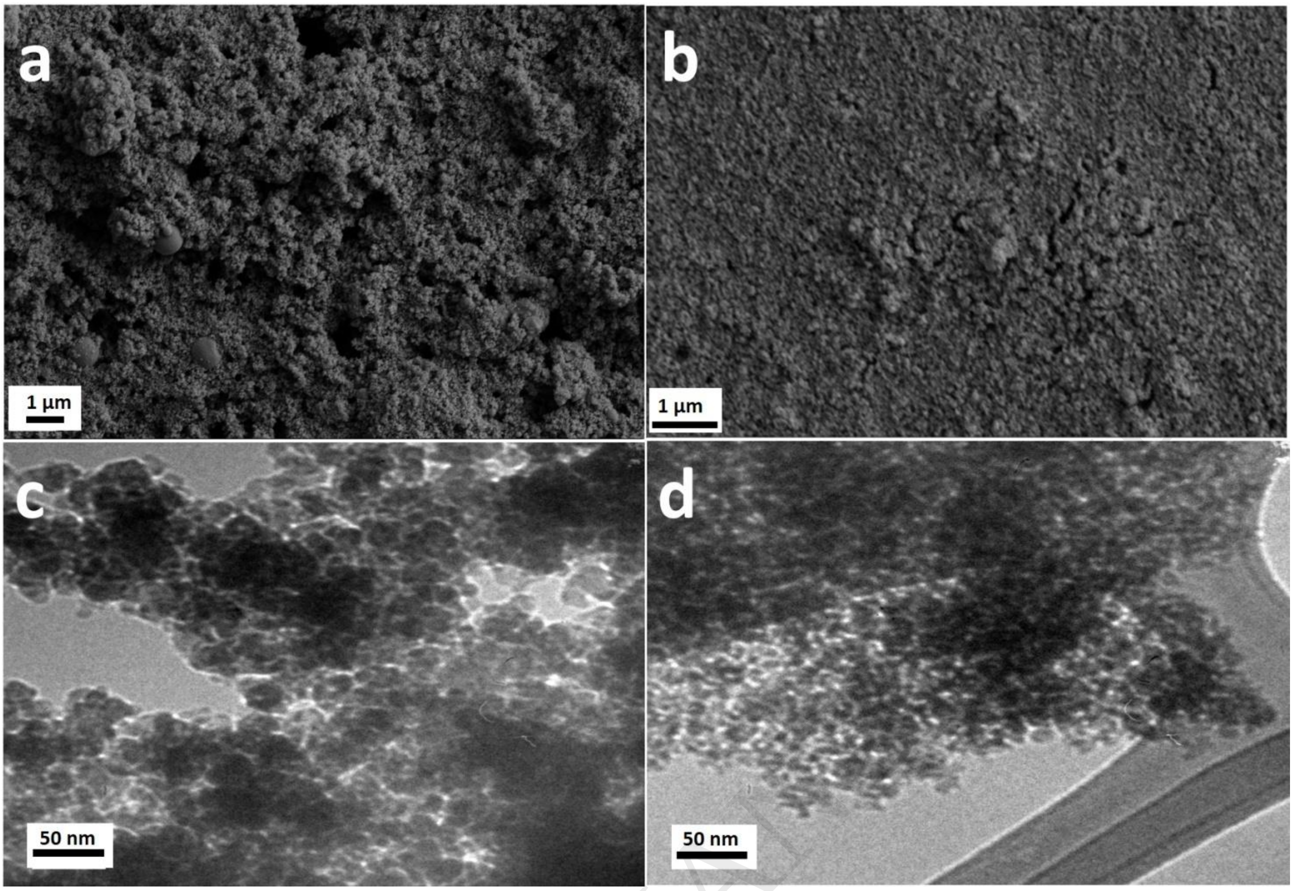


Figure 1. (a, b) FE-SEM of AS-T and AS-NT, respectively, and (c, d) TEM images of AS-T and AS-NT, respectively.

Nitrogen adsorption/desorption measurements were carried out for both AS-T and AS-NT materials, and using the data the corresponding isotherms were plotted (Figure 2b). The isotherm for AS-T appeared to be Type II/IV with H3-type hysteresis loop, and the one for AS-NT seemed to be Type II/III with H3-type hysteresis loop. The isotherms of both materials also indicated the presence of monolayer-multilayer type adsorption and capillary condensation processes at higher relative pressures. The curves qualitatively revealed the presence of, at least, some mesoporous and macroporous structures in both materials. Pore size analysis showed that both AS-T and AS-NT had mesoporosity, with their average diameters centered at *ca*. 31 nm and *ca*. 13 nm, respectively (see

Figure S1 in Supporting Information). The large mesopores in both materials may have to be due to the inter-particle pores formed from the close packing of the particles in the materials, as discussed earlier. Interestingly, the material made without a template showed relatively more monodisperse pores and greater porosity than the one made using a template. The total pore volumes of AS-T and AS-NT were found to be 0.66 $cm^3\ g^{-1}$ and 0.92 $cm^3\ g^{-1}$, respectively, and their corresponding BET surface areas were found to be 100 $m^2\ g^{-1}$ and 323 $m^2\ g^{-1}$, respectively (Table S1, in Supporting Information section).

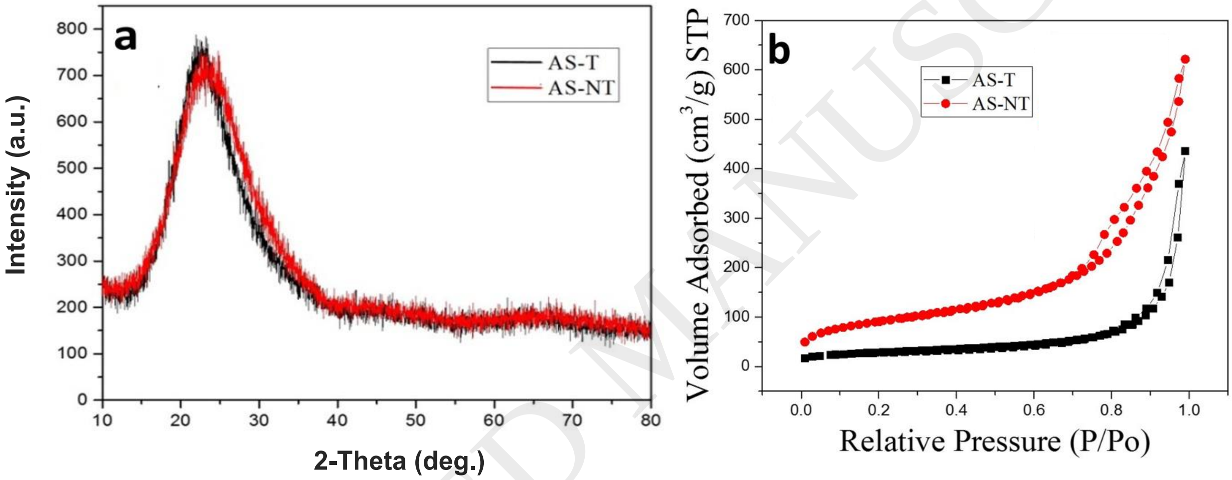


Figure 2. (a) Wide angle XRD patterns and (b) N_2 adsorption-desorption isotherms of AS-T and AS-NT materials.

The large difference in total pore volume, pore diameter and surface area between the two materials seemed to be due to the differences in size and morphology of their particles and the types of inter-particle void spaces the particles generated. Based on the TEM image in Figure 1c, the bigger and different-sized particles in AS-T appeared to have packed in random manner, giving different pore sizes (Figure S2). This is most likely related to TPABr template and the possibility that TPA^+

cations interact differently with the silicate and the aluminate anions formed from the hydrolysis of TEOS and aluminium isopropoxide, respectively, during the self-assembly process. On the other hand, the smaller particles in AS-NT appeared to have packed together more closely, forming smaller and more uniform inter-particle void spaces (Figure 1d). The higher surface area and pore volume in AS-NT could be the result of the formation of more pores from the necking of many smaller sized particles, as can be seen in the TEM image in Figure 1d.

The acidity of the materials measured by TPD (Figure 3) indicated the presence of two NH_3 desorption peaks in both materials: a broad peak centered at *ca.* 100 °C and a second one at *ca.* 400 °C corresponding to weak and strong acid sites, respectively. The intensity of the strong acid peak is relatively higher in the case of AS-T, which indicates the presence of strong acid sites *via* the interaction between the template with Al and Si moieties in this material. The nature of acidity was further characterized using FT-IR spectroscopy of pyridine that was adsorbed on the materials (Figure 4). The IR band between 1540–1548 cm $^{-}$1 and that between 1440–1460 cm^{-1} in aluminosilicates are commonly related to Brønsted (B) and Lewis (L) acid sites, respectively. The IR spectra given in Figure 4 indicated the presence of both Brønsted and Lewis acidic sites in both AS-T and AS-NT materials. Both Brønsted (at ~1540) and Lewis (~1440) acid peaks were higher in intensity in the case of AS-T compared with those in AS-NT. A band at ~1490 related to both Brønsted and Lewis acid sites [17] was also observed in both the samples.

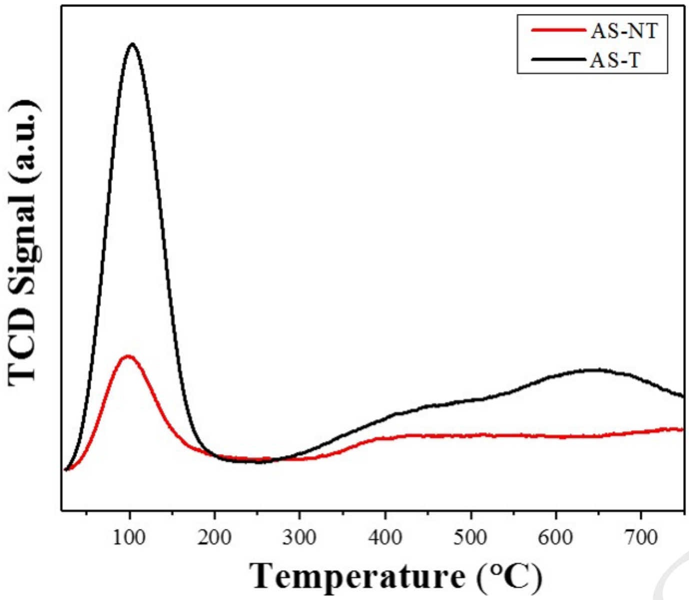


Figure 3. TPD spectra of the AS-T and AS-NT materials.

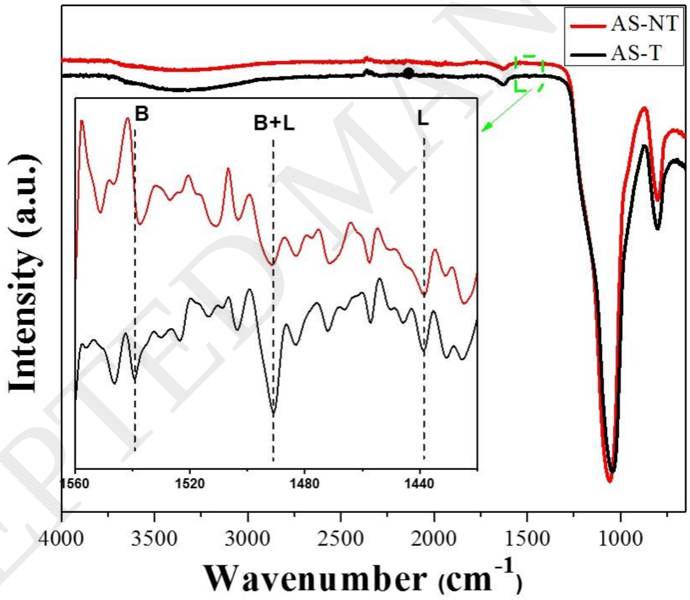


Figure 4. FT-IR spectra of pyridine adsorbed on AS-T and AS-NT materials.

The catalytic properties of the two materials toward toluene alkylation were then investigated. During acid-catalyzed alkylation of toluene with TBA using a solid acid catalyst, the reaction is generally known to take place through the formation of a carbocation intermediate from the alcohol, followed by the electrophilic substitution of a proton in the phenyl ring of toluene by the carbocation, as in Scheme S1 in Supporting Information. So, the reaction has the potential to form at least three possible products: 4-TBT, 3-*tert*-butyltoluene (3-TBT), and 2-*tert*-butyltoluene (2-TBT) [18, 19]. The relative ratio of these three products often depends on the types of catalysts and their catalytic properties as well as the specific reaction conditions employed for the reaction.

In our experiment, the reaction was conducted using AS-NT and AS-T as catalysts without any solvent under atmospheric pressure and at different temperatures, namely 50, 100, or 150 °C. The results, compiled in Table 1, indicated that, at 150 °C, AS-T catalyzed the reaction with ~32 % conversion of toluene and 99% selectivity to 4-TBT product whereas AS-NT catalyzed the reaction with ~24 % conversion of toluene and with 99% of 4-TBT product. AS-T gave a higher conversion of toluene than AS-NT most likely because it has a higher amount of Brønsted and Lewis acid sites [16a]. It is worth emphasizing though that both AS-NT and AS-T gave 99% selectivity to 4-TBT despite their differences in catalytic conversion values; this is most likely because they both have optimal amount of Brønsted and Lewis acids [16a]. Furthermore, raising the reaction temperature from 50 to 150 °C did not alter the selectivity, except making both materials to give slightly higher catalytic conversions (Figure S3 in Supporting Information). In fact, both materials gave only 4-TBT as product under all the reaction conditions in which their catalytic properties were evaluated in this work (Figure S4 in Supporting Information). The possible reasons as to why the catalysts reported herein favored this product are discussed further below.

It is known that acid-catalyzed alkylation of toluene by TBA goes through the dehydroxylation of TBA, whose carbocation product can be in equilibrium with isobutylene [20]. The carbocation subsequently alkylates toluene through electrophilic substitution. Thus, the mechanism by which toluene alkylation proceeds over the catalytically active aluminosilicate materials is likely to be the same as the one illustrated in Scheme S1 in Supporting Information.

Table 1. Catalytic conversion and selectivity of AS-T and AS-NT for toluene butylation with *t*-butanol (TBA) after 9 h-long reaction.

Catalyst	Conversion of Toluene (%)	Product Selectivity to 4-TBT (%)
AS-T[a]	28	~99
AS-NT[a]	22	~99
AS-T[b]	29	~99
AS-NT[b]	22	~99
AS-T[c]	32	~99
AS-NT[c]	24	~99
AS-T[d]	29	~99
AS-NT[d]	21	~99

[a] T = 50 °C, Toluene = 1 mL, *t*-Butyl alcohol (TBA) = 2 mL, and Catalyst = 0.2 g; [b] T = 100 °C, Toluene = 1 mL, TBA = 2 mL, and Catalyst = 0.2 g; [c] T = 150 °C, Toluene = 1 mL, *t*-Butyl alcohol = 2 mL, and Catalyst = 0.2 g.

[d] Used catalyst, recovered 9 h after the reaction and re-tested at T = 150 °C, Toluene = 1 mL, *t*-Butyl alcohol = 2 mL, and Catalyst = 0.2 g.

Notably, the selectivity of the aluminosilicate catalysts, especially the one synthesized without using any template, to give only one product in the toluene alkylation reaction, is interesting. For this reaction as well as other related ones (*e.g.*, disproportionation of toluene to xylenes and benzene) catalyzed by most aluminosilicate catalysts possessing acidic sites, the types of products forming from reaction are often dependent on whether or not the isomerization of some of products within the catalysts is possible. This, in turn, can be affected by various structural factors present in the catalyst, such as the pore sizes of the catalyst, the diffusion coefficient of reactants, intermediates and products within the pores of the catalysts and their thermodynamic stability, the reaction conditions, the acidity of the active sites of catalysts, and possible coke deposition [21]. For example, shorter reaction time during toluene alkylation has been reprted to favor 4-*tert*-alkylated products because isomerization of these products to 2- and 3-*tert*-alkylated isomers in the pores of the catalysts is unlikely [22]. Hence, pore narrowing within aluminosilicate material by silylation of the pore walls using tetraalkoxysilanes or other agents is sometimes carried out to favor the selectivity of the reactions to 4-*tert*-alkylated products [23]. The external surface area and the crystal size of the catalyst have also been reported to favor the selectivity of the reaction to 4-*tert*-alkylated products [24]. This is because an increase in the size of the catalyst particles reduces the total external surface area of the catalyst and limits the number of available acidic sites on the external surface of the catalyst; this in turn impedes the transformation of 4-*tert*-alkylated toluene to 2- or 3-*tert*-alkylated toluene product *via* isomerization [25]. In our case, the major factor appears to be the presence of optimal density of Brønsted and Lewis acid sites in the catalyst, rather than the selectivity of the pores. The right relative amount of Brønsted and Lewis acid sites has been reported to favor TBT product [16a].

Finally, the recyclability and reusability of the catalysts were evaluated by recovering them after the end of each reaction. Both AS-T and AS-NT catalysts were found to maintain their catalytic selectivity and give 99% of 4-TBT as product (Tables S2 in Supporting Information).

4. Conclusions

In conclusion, we have shown a simple template-free synthetic method involving only heat treatment at moderate temperature that can give amorphous nanoporous aluminosilicate that exhibits high catalytic selectivity for toluene alkylation with *tert*-butanol almost exclusively giving 4-TBT as product. The reported synthetic method is simple and potentially applicable for making other types porous titanium silicate and metal oxide catalysts. The finding of high catalytic selectivity observed for the catalyst suggests that the reported material can have strong industrial relevance, given the importance of selective toluene alkylation reaction in the chemical industry.

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