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Synthesis of a silicotungstic acid SBA-15 catalyst for selective monoglyceride production

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Abstract The esterification reaction between glycerol and lauric acid was conducted in the presence of commercial Amberlyst IR-120 and synthesized silicotungstic acid (STA)/SBA-15 at 393, 413 and 433 K. The results indicated that STA/SBA-15 was more convenient than Amberlyst IR-120 by means of monoglyceride production. Higher selectivity to monoglyceride obtained in the presence of STA/SBA-15 was due to its mesoporous structure, which induced shape selectivity in favor of monoglyceride. Reaction experiments in the presence of recovered catalysts showed an increase of monoglyceride selectivity, which was due to adsorbed lauric acid inside the mesopores. The selectivity was shown to be altered towards monoglyceride by simply changing the feed ratio and applying washing procedure on the catalyst. Characterization studies conducted on recovered catalysts validated the preservation of catalyst structure and the stability of the catalyst even after utilization for 7 days.

Keywords Esterification · Lauric acid · Glycerol · Silicotungstic acid

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Introduction

Heteropoly acids (HPA) have emerged as a catalyst alternative in acid catalyzed reactions following their first utilization in the production of 2-propanol [1]. Despite high activity obtained in homogeneous reactions, their solubility in the presence of polar reactants created separation problems [1–3]. Low specific surface area ($<10\ m^2\ g^{-1}$) [2, 4–7] is another limitation of these catalysts which could be solved by dispersing them onto a support material with large surface area [1–7]. Activated carbon, silicon dioxide, clay, titanium and zeolites are commonly used supports [1, 6]. However, the heteropoly acid remained on the surface of these materials as a result of their pore sizes, which was smaller than 1 nm. Due to weak surface interactions of the active HPA and the support, liquid phase applications in the presence of these catalysts resulted in leaching of the active material to reaction mixture [8].

Molecular sieves were also used as support materials in catalyst synthesis and the use of these materials has been investigated since the discovery of mesoporous MCM-41 [9, 10]. SBA-15, the latest discovered mesoporous silicate has thicker pore walls, wider pore sizes and higher hydrothermal stability compared to MCM-41 and hence a better alternative for use as a support material [4–6]. Their uniform tubular channels with a varying pore diameter from 50 to 300 Å enable incorporation of active HPA inside and provide a sheltered medium for HPA against leaching [5]. The catalysts synthesized with SBA-15 as a support material could have easily been regenerated as the molecular structure of SBA-15 remained intact even after washing and calcination [7].

The main idea in the synthesis of supported HPAs was to obtain an active catalyst with combined properties of favorable catalytic activity of HPA and physical properties of the support material SBA-15 [2]. One way to achieve this combination was the use of direct impregnation of HPA to already synthesized SBA-15 followed by evaporation of the HPA solution [2–8, 11, 12]. However, partial leaching of the active material due to the weak interaction with the support was the main drawback in impregnation, resulting in activity losses in repeated use [13, 14].

In the present study, the direct hydrothermal method that requires simultaneous addition of the active material and the silicate source to an acidic surfactant solution [8, 15] was applied in the synthesis of the STA/SBA-15 catalyst. It was thought that better dispersion of the active material inside the pores of the support could have been achieved by applying this method. This method, in our opinion, would also enable maintaining high activity with much lower loading amounts and prevent leaching of the active material. Silicotungstic acid (STA) was selected as the active material due to its superior properties in acid catalyzed reactions [16, 17]. HPA/SBA-15 catalysts have been used in a variety of reactions such as oxidation of benzaldehyde to benzoic acid [3], esterification of acetic acid and ethanol [4], alkylation of phenol [5], isopropylation of naphthalene with isopropanol [6], benzylation of phenol [18] and esterification of free fatty acids [11, 12, 15].

Esterification of glycerol with lauric acid was selected as the model reaction to test the activity of the STA/SBA-15 catalyst and the results were compared with the results of reaction experiments conducted in the presence of commercial Amberlyst

IR-120 catalyst. Monoglycerides, the primary products of the reaction between glycerol and lauric acid, have been utilized in pharmaceutical formulations, drug delivery systems and oil well drilling operations [19–21].

Investigations on the esterification of glycerol with lauric acid mainly focused on process parameters such as reaction temperature [15, 21], feed combination [21], solvent type [22] and the type and amount of the catalyst used [23]. The mesoporous molecular sieve with active material inside their structure was an important catalyst alternative for use in esterification reaction with lauric acid. The high monoglyceride yield was due to shape selectivity of the catalyst [23].

Commercial Amberlyst IR-120 was known as an effective catalyst and utilized in a variety of esterification reactions [24–26]. The acidities of Amberlyst IR-120 and silicotungstic acid were previously determined as 2.5 and 1.3 mmol H^+/g, respectively [27]. The behavior of these catalysts in esterification reactions are very different. Amberlyst IR-120 has sulfonic acid groups donating proton in the course of reaction, on the other hand the protons in heteropoly acid structure completely dissociate during reaction. Although these catalysts indicate different behavior in the esterification reaction, it is imperative that STA/SBA-15 catalyst, synthesized in the present study, should indicate an activity comparable to Amberlyst IR-120. Based on the survey conducted by the authors of this study, there are no investigations on the esterification of glycerol and lauric acid in the presence of Amberlyst IR-120 catalyst and we believe that utilization of this catalyst in glycerol esterification with lauric acid would provide an important contribution to the literature. The HPA loading applied in synthesis was 5 % which was a very low value compared to those reported in literature [1, 2, 18, 19]. Hence, the present study aimed to obtain a commercial grade activity in the presence of STA/SBA-15 catalyst with much lower loading amount.

Attaining high and sustainable activity with recovered catalysts was the main objective of the present study. The catalysts recovered at the end of first run was washed, dried and reused in the second run with identical conditions in order to achieve this objective. The stability of the catalysts was also tested with a reaction experiment conducted in identical conditions for 7 days.

Experimental

Synthesis of STA/SBA-15 catalyst

The direct hydrothermal method was applied in the synthesis of STA/SBA-15 catalyst. The synthesis procedure was a modification of a previously applied procedure [28]. The initial step in the synthesis was to prepare a surfactant solution which was achieved by dissolving 4 g of Pluronic P123 (Sigma-Aldrich) in 144 mL of 1.7 M HCl at 313 K. The resultant solution was stirred for 4 h to maintain complete dissolution. Tetraethyl orthosilicate (TEOS, Merck) used as silica source and silicotungstic acid (STA, Sigma-Aldrich) used as the active material were simultaneously added to the surfactant solution according to direct hydrothermal method [8, 15]. STA was used as the active material based on its superior properties

in acid catalyzed reactions [16, 17]. It was also anticipated that Si, present in the structure of STA, would have acted as an additional silica source and facilitated incorporation of the active material inside the pore structure. The amount of TEOS was determined as 8 g based on the weight ratio of TEOS/Pluronic P123, which was selected as 2 in the course of syntheses. The resulting solution was further stirred for 2 h and aged at 373 K for 48 h in order to achieve completion of consecutive hydrolysis and condensation of TEOS. The crystalline sample obtained at the end of aging process was washed with deionized water, dried at 353 K for 12 h and calcined at 523 K for 5 h in a tubular furnace for the removal of the surfactant. The calcination temperature was determined according to thermal stability of STA. The amount of STA was determined as 0.07945 g, which corresponded to 5 % loading based on weight ratio of tungsten (W) of STA to Silicium (Si) of TEOS solution.

Reaction experiments and reusability studies

Mono-di and tri glyceride production was achieved via series of reactions given below:

$$\text{Glycerol} + \text{Lauric acid} \rightarrow \text{Monolaurin} + H_2O$$

$$\text{Lauric acid} + \text{Monolaurin} \rightarrow \text{Dilaurin} + H_2O$$

$$\text{Lauric acid} + \text{Dilaurin} \rightarrow \text{Trilaurin} + H_2O$$

Experiments were conducted in a batch reactor system operated with autogenic pressure. The reaction system consisted of a Teflon autoclave situated inside a steel reactor and a magnetic stirrer which was operated at 1000 rpm to minimize mass transport limitations. The effects of various reaction parameters on lauric acid conversion and product selectivity were investigated through experiments conducted with varying catalysts, catalyst amounts, temperature and feed ratio. Ethanol was added to the reaction mixture as a solvent to enhance gradual decrease of the viscosity of reaction solution and increase homogeneous distribution of lauric acid

Table 1 Experimental conditions applied in esterification reactions

Catalyst	Loading amount (g)	T (K)	Feed molar ratio	Run no
Amberlyst IR-120	1	363	6/3/1	1
Amberlyst IR-120	1	383	6/3/1	1
Amberlyst IR-120	1	393	6/3/1	1
STA/SBA-15	1	393	6/3/1	2
STA/SBA-15	1	413	6/3/1	2
STA/SBA-15	1	433	6/3/1	2
STA/SBA-15	0.5	433	6/3/1	1
STA/SBA-15	0.5	433	3/3/1	1
STA/SBA-15 (7 days)	0.5	433	6/3/1	1

[29, 30]. Glycerol/ethanol/lauric acid amounts utilized in reaction experiments were selected as 30/15/5 and 15/15/5 mL. 5 M concentration of each reactant with stated volumes were taken as basis which corresponded to molar ratios of 6/3/1 and 3/3/1. The reaction time was determined as 6 h in the course of experiments in order to minimize experimental error emanating from sample withdrawal. The experimental conditions applied in the experiments are given in Table 1 and the scheme of apparatus used for esterification are shown in the supplementary material.

Reusability studies were conducted under identical conditions in the presence of catalysts recovered from the reaction mixture. Recovered catalysts were washed with water and dried at the temperature of reaction. Stability of STA/SBA-15 catalyst for long term use was tested with reaction experiments conducted at 433 K for 7 days. A sample from the reaction solution was withdrawn in every 48 h starting from the first day to minimize experimental error.

Product analysis

Samples from the reaction mixture were collected hourly and analyzed by gas chromatography using Shimadzu GC-2010 device equipped with a capillary column (Restek Rtx-1 30 m × 0.32 mm × 0.1 μm) according to the analysis method [21]. Conditions applied in the gas chromatograph are summarized in Table 2. The temperature of the detector and injector were held at high values due to high boiling point of the reactants.

Product analyses were conducted with 100 μL samples collected hourly from the reactor. Samples were diluted by adding 100 μL of water and methyl acetate. The mixture was centrifuged to collect the organic phase. 20 μL of the organic phase was again diluted by adding 480 μL acetone and 100 μL 0.2 M pentadecanoic acid and 0.02 μL of this solution was injected to the gas chromatograph for analysis. The conversion was determined based on the amount of lauric acid (C_A) reacted to form mono (C_M), di (C_D) and triglycerides (C_T). The conversion and selectivity to products were determined according to the formulas [21]:

$$\text{Conversion } \% = [(C_M + 2C_D + 3C_T)/(C_M + 2C_D + 3C_T + C_A)]\text{x}100$$

$$\text{Monoglyceride selectivity } \% = \left[\frac{(C_M)}{(C_M + 2C_D + 3C_T)}\right]\text{x}100$$

Table 2 Operating conditions of the gas chromatograph applied in the esterification reaction [21]

Column	Restek Rtx-1 capillary column 30 m × 0.32 mm × 0.1 μm
Detector	Flame ionization detector (FID)
Carrier gas (N_2)	1.5 mL/min
Temperature program	353 K (1min) ⟶ 288K/min603 K (2 min)
Detector temperature	653 K
Injector temperature	553 K

$$\text{Diglyceride selectivity } \% = \left[\frac{(2C_D)}{(C_M + 2C_D + 3C_T)}\right] x100$$

$$\text{Triglyceride selectivity } \% = \left[\frac{(3C_T)}{(C_M + 2C_D + 3C_T)}\right] x100$$

Characterization studies

XRD patterns of recovered STA/SBA-15 catalysts were obtained on a Panalytical Empyrean HT-XRD instrument using Cu K_α radiation with 0.066° step size and over the range of $0° < 2\theta < 10°$. Adsorption–desorption isotherms and pore size distributions of recovered STA/SBA-15 catalysts were obtained by nitrogen physisorption analyses conducted in ASAP2020 instrument. Samples were degassed at 523 K for 3 h prior to analyses. SEM images of catalysts were obtained with Zeiss SUPRA V 40 device. EDX analyses were applied to determine the extent of active material leached during reaction experiments.

Results and discussion

Reaction experiments and reusability studies

The results of reaction experiments conducted in the presence of Amberlyst IR-120 and STA/SBA-15 at 393 K and with 1 g catalyst loading are illustrated in terms of lauric acid conversion and mono and diglyceride selectivity (DGS) in Fig. 1. The conversion value obtained with Amberlyst IR-120 reached 98 % after 3 h and the change occurred in conversion values from this point was negligible. Comparison of lauric acid conversions (LAC) in the presence of Amberlyst IR-120 and STA/SBA-15 catalysts revealed similar values after 3 h of the experiment. Although high conversion values were obtained in the presence of Amberlyst IR-120, it was interesting to observe 100 % DGS with this catalyst. Usually, in the esterification of glycerol with lauric acid, high MGS is anticipated and the necessity of the design of catalysts that could show shape selectivity was pointed out in the literature. STA/SBA-15 catalysts have a pore diameter of 9.1 nm which provides a large area for the adsorption of the molecules during reaction. Compared to the kinetic diameter of glycerol [31] (0.63 nm) it is clear that the synthesized catalysts would show shape selectivity towards monoglyceride. In other words, catalysts with mesopores favored monoglyceride formation [15, 21, 23]. Amberlyst IR-120 is an ion exchange resin with a surface area of 0.3 m^2/g and essentially no porosity. Merchant et al. [25] reported that in the presence of Amberlyst IR-120, Eley–Rideal mechanism best correlated the kinetic data for the esterification reaction of ethanol with acetic, propanoic and pentanoic acids, which, in our opinion, was the case for glycerol esterification with glycerol and water adsorbed on the surface of the catalyst. Comparison of LAC values for both catalysts implied that the synthesized STA/SBA-15 catalyst were suitable for use in industrial applications due to similar

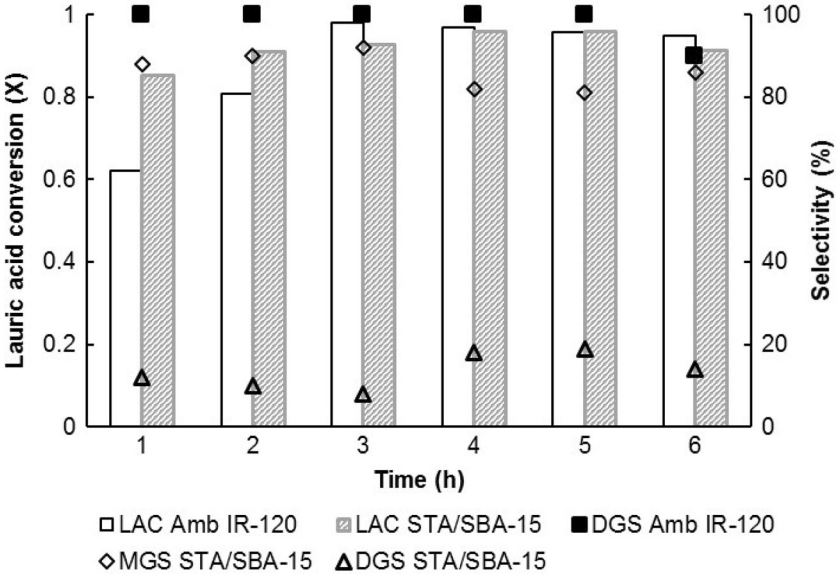

Fig. 1 Lauric acid conversion (LAC) and selectivity values obtained in the presence of Amberlyst IR-120 and STA/SBA-15 catalysts (Catalyst loading: 1 g; T = 393 K)

activities obtained under identical experimental conditions. Nevertheless, only diglyceride formation was observed in the presence of Amberlyst IR-120, which was less favorable compared to monoglyceride and as far as the type of the product was concerned, STA/SBA-15 was more convenient than Amberlyst IR-120 for the applied experimental conditions (Fig. 1).

The effect of temperature on LAC and monoglyceride selectivity (MGS) obtained in the presence of STA/SBA-15 is shown in Fig. 2. A decrease in LAC was observed with increasing temperature, which was due to the competitive adsorption of water obtained as side product in the reaction. It was thought that water competed with glycerol for the adsorption sites, which resulted in the decrease of conversion values. MGS higher than 80 % was obtained in reaction experiment conducted at 393 K. However, a decrease was observed in MGS with further increase of temperature. The decrease in MGS was due to diglyceride and triglyceride formation. Diglyceride and triglyceride formation at elevated temperatures was thought to be due to the increase in kinetic energy of lauric acid with increasing temperature (Fig. 2).

Experiments were conducted in the presence of 0.5 g STA/SBA-15 catalysts at 433 K with glycerol/ethanol/lauric acid molar ratios of 6/3/1 and 3/3/1 in order to determine the effect of solvent on LAC and product selectivity. An increase was observed in LAC with the decrease of glycerol (3/3/1) in feed mixture. The increase in LAC might be due to sudden decrease of viscosity of the reaction media with decreasing glycerol amount. In other words, glycerol, used as excess in reactions, probably covered the active sites and prevented adsorption of lauric acid during the

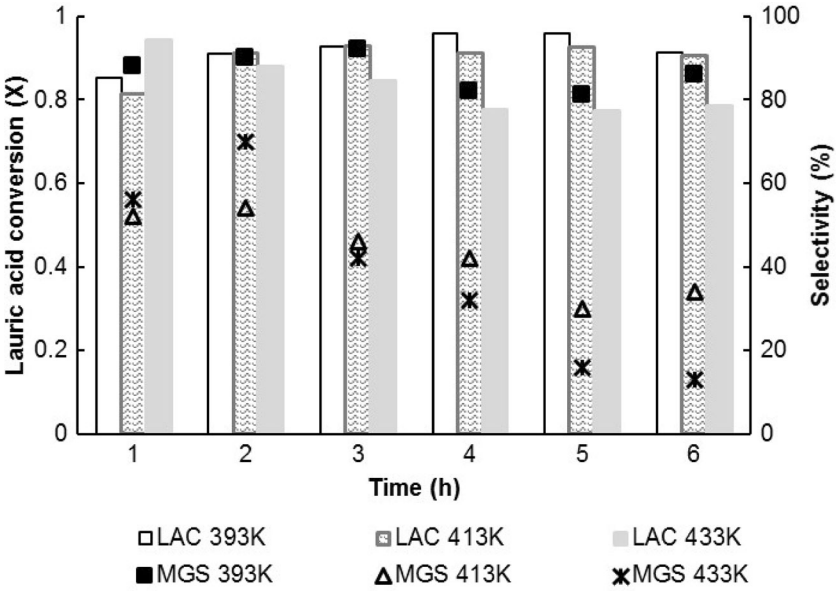

Fig. 2 Effect of temperature on conversion and monoglyceride (MGS) selectivity in the presence of STA/SBA-15 catalyst (Catalyst loading: 1 g)

reaction, which resulted in the decrease of conversion in the case of 6/3/1 feed ratio (Fig. 3). On the other hand, an increase in DGS was observed with 3/3/1 feed ratio. The increase in DGS could be explained by the affinity of the catalyst towards glycerol. In the case of low glycerol amount, the adsorbed amount of lauric acid was higher, which enabled progress of the consecutive reaction leading to diglyceride formation. Triglyceride formation could be considered as negligible as triglyceride formation was observed only in the case of 6/3/1 feed ratio with an average selectivity of 5 % (Fig. 3).

The reusability of the catalysts was investigated under identical experimental conditions, in the presence of catalysts recovered from reaction mixture. Recovered catalysts were washed with water and dried at the temperature of reaction. The conversion and selectivity obtained at 393 K are given in Fig. 4. In order to explain the decrease in activity, the reaction temperature and washing conditions need to be taken into consideration. The decrease in LAC was thought to be due to adsorbed lauric acid in the catalyst, which could not have been fully removed from the catalyst even after intensive washing. On the other hand, almost 100 % MGS has been obtained in the presence of reused catalyst (Fig. 4). Reaction experiments conducted in the presence of fresh STA/SBA-15 at 413 K led to formation of triglyceride. However, it was interesting to observe that triglyceride was not formed in the presence of recovered catalyst. Instead, an increase in MGS in the presence of recovered catalyst was observed. Recovered catalysts contained lauric acid, which could not be removed with the applied

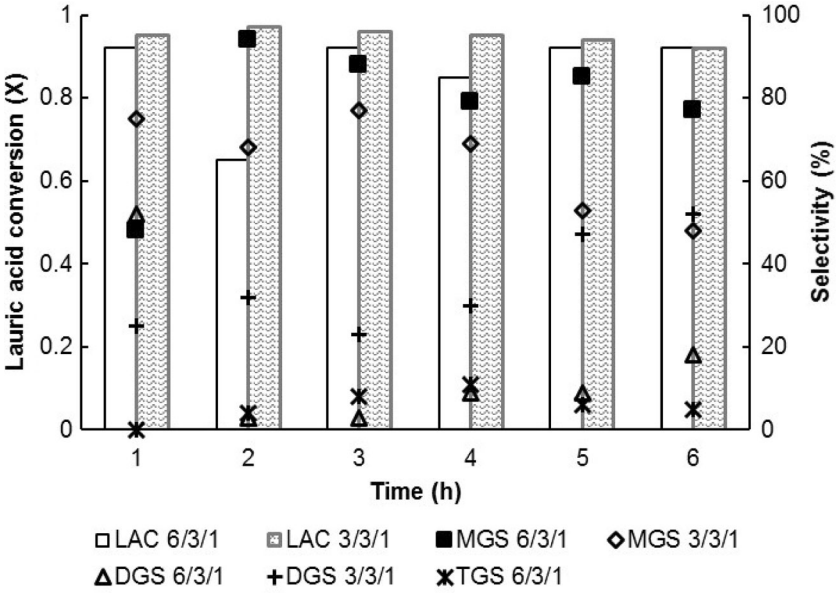

Fig. 3 Effect of feed ratio on lauric acid conversion (LAC) and mono-di-tri glyceride (MGS, DGS,TGS) selectivity (Catalyst loading: 0.5 g; T = 433 K)

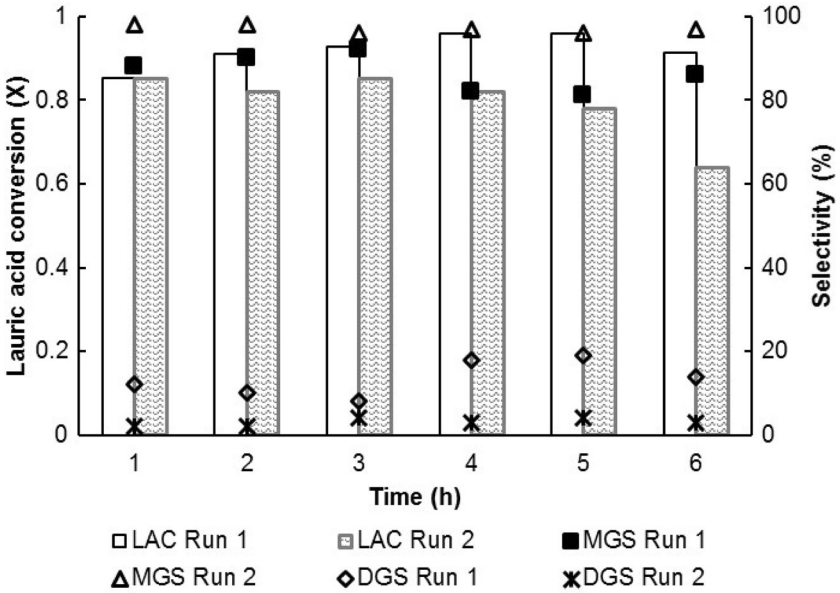

Fig. 4 Reusability studies conducted in the presence of STA/SBA-15 catalysts at 393 K (Catalyst loading: 1 g)

washing procedure. The presence of lauric acid in the catalyst structure, in our opinion, enhanced monoglyceride formation. Reaction experiments conducted in the presence of recovered catalysts at 433 K resulted in an increase in lauric acid conversion. The kinetic energy of glycerol and lauric acid was higher at higher temperatures which led enhancement of chemisorption on the active sites and effective interaction between reactant molecules [21]. MGS was higher and triglyceride formation was not observed in the presence of recovered catalyst. The results obtained with reaction experiments in the presence of recovered catalysts was one of the highlights of present study as it was shown that selectivity could have been altered towards monoglyceride by simply applying a washing procedure to the catalyst.

Experiments in the presence of STA/SBA-15 catalyst were conducted at 433 K for 1 week to determine catalyst stability. The results showed a LAC of 84 % at the end of 7 days. However, diglyceride and triglyceride formation was also observed as the experiment has proceeded. This result was due to the coverage of the catalyst surface and pores by glycerol, which hindered the adsorption of lauric acid and led to the formation of diglyceride and triglyceride through further esterification steps. The coverage of the pores and the surface with glycerol were reasonable due to the high affinity of the catalysts towards alcohols during esterification reaction [24] (Fig. 5). An increase in di- and triglyceride yield due to coverage of active sites was also observed in the work of Hoo and Abdullah [15], in the presence of HPA/SBA-15 catalyst with 30 and 40 % HPA loading.

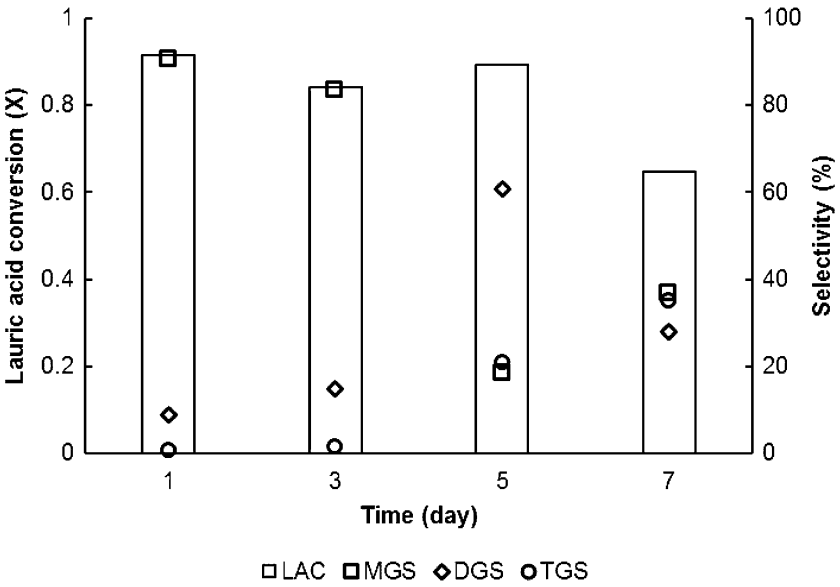

Fig. 5 Catalyst stability at the end of 7 days (Catalyst loading: 0.5 g; T = 433 K)

Characterization studies conducted in the presence of recovered catalysts

The XRD patterns of STA/SBA-15 catalysts recovered after 2nd run of reaction experiments at 393 and 433 K and STA/SBA-15 catalyst recovered after 7 days of experiment at 433 K are shown in Fig. 6. Compared to fresh STA/SBA-15 [32], identical patterns were obtained with spent catalysts. XRD patterns of spent catalysts were also identical regardless of feed conditions and reaction time, which indicated the negligible effect of reaction temperature, time and reactant interaction on the catalyst structure. The same conclusion could be reached by the evaluation of the nitrogen adsorption–desorption isotherms of the catalysts (Fig. 7).

A type IV isotherm was observed for fresh [32] and spent catalysts with a decrease in hysteresis in the case of catalyst recovered after 7 days of reaction. Isotherms indicated that catalysts preserved their mesoporous structure after reaction. On the other hand, the decrease of the hysteresis effect with spent catalysts showed that some of the reactants adsorbed inside the pores during reaction could not have been fully removed from the catalyst even after the applied washing procedure. In our opinion, the majority of the reactants adsorbed inside the pores were lauric acid and combined with its elevated kinetic energy due to reaction temperature (433 K), their presence inside the pores resulted in elevated LAC and MGS (Fig. 7). The pore size distribution of recovered catalysts indicated a mesoporous structure regardless of reaction time and amount of glycerol used in feed mixture. This result implied that most of the glycerol, although used in excess, was successfully recovered from the catalyst structure by the applied washing procedure. It was previously stated that lauric acid, remaining inside the catalyst

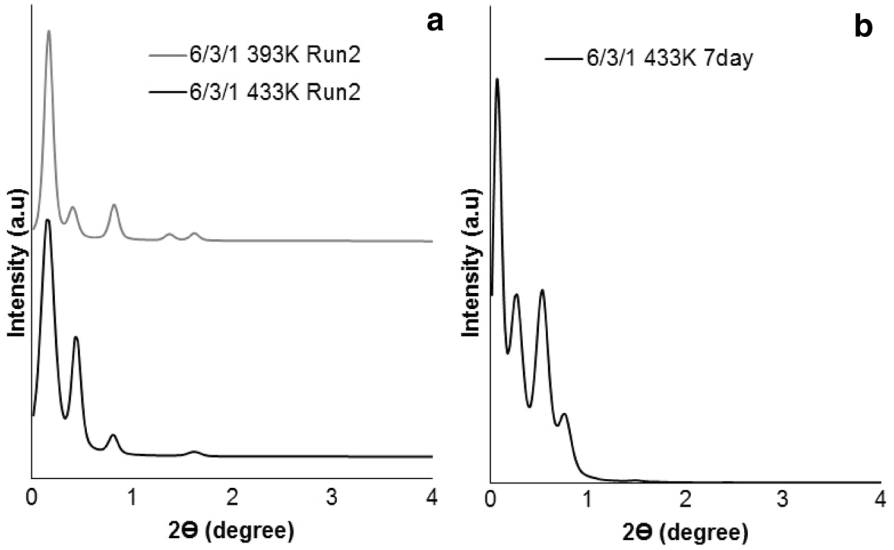

Fig. 6 XRD patterns of **a** STA/SBA-15 catalysts recovered after 2nd run of reaction experiments (T = 393, 433 K; Catalyst loading: 1 g) **b** STA/SBA-15 catalyst recovered after 7 days of reaction experiment (T = 433 K; Catalyst loading: 0.5 g)

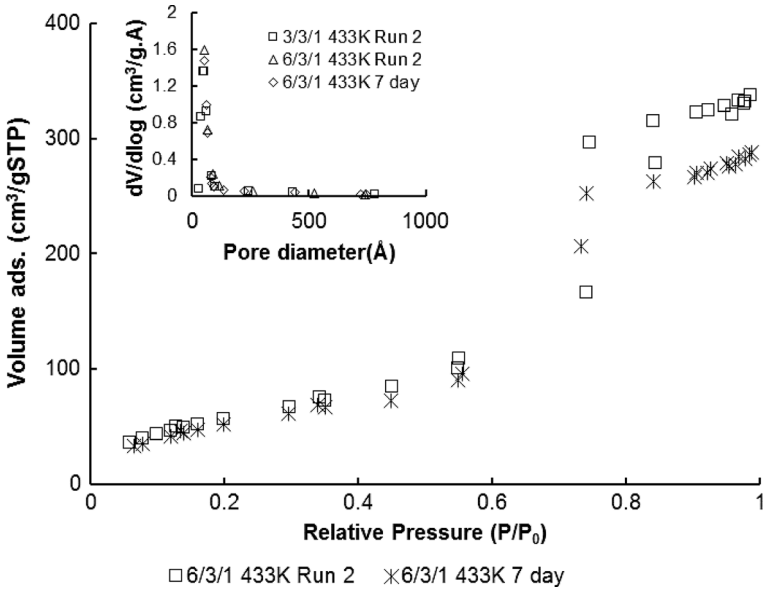

Fig. 7 Nitrogen adsorption–desorption isotherms of recovered catalysts (Catalyst loading: 1 g; T = 433 K)

after washing, was responsible of the increase of MGS in the presence of recovered catalysts. This conclusion was further validated by comparing the results of FT-IR analyses conducted to STA/SBA-15 and recovered catalysts at the end of 2nd run and 7 day. The results obtained from fresh and recovered catalysts were compared to FT-IR spectrums of glycerol, lauric acid and monoglyceride. Compared to fresh STA/SBA-15, the FT-IR spectrum of glycerol and lauric acid revealed distinctive peaks at 2879 and 2885 cm^{-1}, which might indicate CH_2 groups of glycerol or symmetric stretching peak of -CH_2 and stretching vibration of -CH_2 belonging to lauric acid (Fig. 8) [33, 34]. In the case of recovered catalyst, a distinctive peak at 1695 cm^{-1}, which was not present in FT-IR spectrum of glycerol and fresh catalyst, was observed. This peak might correspond to the carbonyl group (C=O) of lauric acid. The interpretation of the results so far implied the presence of lauric acid in the structure of the recovered catalysts. Although the FT-IR spectrum of mono laurin was almost identical to that of lauric acid, a deduction on the absence of mono laurin could be made by evaluating the results of FT-IR analysis and reaction experiments in accordance. Reaction experiments in the presence of recovered catalysts indicated negligible formation of diglyceride and tri glyceride, which could also be interpreted as the lack of reaction between monoglyceride and lauric acid to form diglyceride (Fig. 4). This, in our opinion, could only be possible in the case of monoglyceride absence in the structure of recovered catalyst (Fig. 8). The evaluation of FT-IR results as stated also explain the increase observed in MGS selectivity in the presence of recovered catalysts.

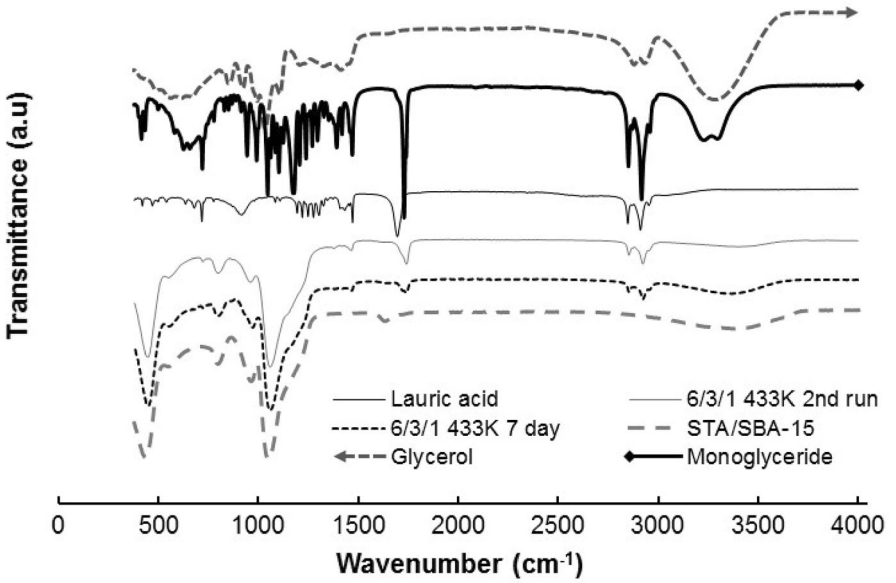

Fig. 8 FT-IR spectra of lauric acid, SBA-15 and recovered catalysts

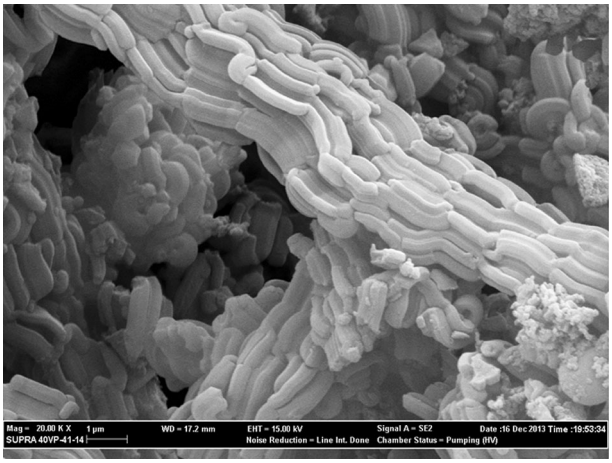

Fig. 9 SEM images obtained with STA/SBA-15 catalysts recovered after the 2nd run of reaction (Catalyst loading: 1 g; T = 433 K)

The wheat-like morphology of SBA-15 [18] could be seen from the SEM image of recovered catalyst (Fig. 9). This result clearly showed that the structure of the catalyst was well preserved after reaction. EDX results and textural and physical properties of STA/SBA-15 catalysts recovered after 2nd run of reaction at 433 K and recovered after 7 days of reaction at 433 K are given Table 3. Compared to

Table 3 EDX results and textural and physical properties of recovered STA/SBA-15 at 433 K (Feed molar ratio: 6/3/1 glycerol/ethanol/lauric acid)

Catalyst	EDX (W/Si %)	Surface area (m^2/g)	Pore volume (cm^3/g)
STA/SBA-15 [32]	5.00	663	1.10
STA/SBA-15 2nd run	4.48	218	0.53
STA/SBA-15 7 days	4.06	201	0.45

BET surface area and pore volume of STA/SBA-15, results indicated a decrease of BET surface areas and pore volumes due to the presence of adsorbed reactants inside the pores of recovered catalysts. On the other hand, EDX analysis of recovered catalysts showed that STA, leaching to reactant mixture during reaction, was negligible. This was also one of the highlights of the present study. The evaluation of characterization results revealed that a simple washing procedure could have greatly enhanced MGS and the catalysts could have been used repeatedly without changes in their structure.

Conclusions

Reaction experiments conducted with STA/SBA-15 and recovered catalysts showed that the synthesized STA/SBA-15 catalyst was more convenient than Amberlyst IR-120 by means of obtaining the desired product. A comparison in their activity revealed similar conversions which implied that the synthesized catalyst be used as an alternative of Amberlyst IR-120 in esterification reactions. In the case of experiments conducted in the presence of recovered catalysts, the decrease of LAC observed at 393 and 413 K was due to higher affinity of the catalyst towards glycerol. However, further increase of reaction temperature resulted in the increase of lauric acid conversion and MGS which was due to the increase of the kinetic energy of lauric acid. Decreasing the amount of glycerol in the feed mixture greatly influenced the type of product in favor of the desired monoglyceride. It was concluded that most of the excess glycerol could have been recovered by the washing procedure, yet lauric acid still remained inside the catalyst. The recovery of the glycerol was interpreted by the pore size distributions of recovered catalysts showing a mesoporous structure. The presence of lauric acid, responsible of the increase of MGS, inside the catalyst was validated by FT-IR analyses of recovered catalysts. Consequently, the effect of washing was shown to have an increasing effect on MGS and it was concluded that the catalyst can be regenerated by simply washing with water.

Characterization studies mainly showed that the catalyst was been influenced by temperature, reaction time and the reactants used in reaction experiments. The fact that STA amount was nearly stable after reaction clearly showed that sustainable activity with desired selectivity could have been obtained in long term with the synthesized catalyst.

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