**Dear Reviewers**

Thanks for your kindly attention. We have tried to answer your comments, meticulously. The sentences in the paper which address your precious comments are highlighted by using yellow pattern.

We hope that our answers can be enough clear, complete and acceptable. We are looking forward to receiving positive answers from you. Thanks for your kindly attention.

Truly Yours,

Mehdi Rashidzadeh

**Reviewer A:**   
Is the manuscript clearly and concisely written?   
Ans. Based on the reviewer comments, the manuscript was revised.

Are the conclusions adequately supported by the data?  
Ans. Based on the reviewer comments, the manuscript was revised.

Is the quality of the figures (including legends and axes labelling) satisfactory?   
Ans. The quality of all figures is improved.  
  
Are the English grammar and syntax satisfactory?  
Ans. The English grammar and syntax of manuscript was totally improved.

ADDITIONAL COMMENTS

- Lines 87 and 88: “sodium aluminate via post-synthetic alumination method was used” → “sodium aluminate was used for post-synthetic alumination”

Ans. It is done (Page 3)

- Lines 94-95: The sentence “It should be mentioned…, respectively” should be removed, it is repetition of the previous sentence.

Ans. It is removed (Page 3).

- Line 98: “physical” → “physical properties”

Ans. It is done (Page 4)

- Lines 111 and 112: the temperature and duration of the samples pretreatment are very high/long; the temperature is higher than of two calcination temperatures (350 and 450 ⁰C); therefore, it can be expected the changes of the structure of these samples during the pretreatment and the results of the acidity determination are not reliable for these samples.

Ans. The procedure written for determination of acidic properties of proposed samples by AutoChem 2900 (Micromeritics) automatic analyzer is a routine procedure that has be mentioned in many references.

In this study, the samples were synthesized and calcined at different temperatures at first. Then, Al-grafting was done for all samples in different Si/Al mole ratios. Afterward, fresh portions of each samples were used for SAXS, XRD, FTIR, N2 adsorption-desorption and NH3-TPD analyses for determination of structural, textural and acidic properties. It should be mentioned that used samples in each above mentioned analyses is not used for the second times.

Although, it is expected that the structure of these samples is modified after NH3-TPD analysis, but NH3-TPD results of samples are reliable because the amount of acid sites that created on different calcined samples is not changed during NH3-TPD analysis.

- Use K or ⁰C, not both.

Ans. “⁰C” is used entire the manuscript.

- Why the results of SAXS analysis are given just for samples SBA-450-y? For example, it is useful to give SAXS results for SBA-x-10Al, to discuss the influence of calcination temperature on the structure of the aluminated samples.

Ans. Due to higher number of acid sites resulted due to higher amount of aluminum loading (NH3-TPD results), The SAXS analysis was performed only for SBA-450-(y) samples for investigating the effect of Al-loading on the structure of samples. Results showed that the order of structure is maintained even at high Al-loading. Therefore, by considering results of SAXS analysis of SBA-450-(y) samples and N2 adsorption-desorption isotherms (type V with hysteresis loop H1), it was expected that the same results were obtained for samples SBA-350-(y) and SBA-550-(y). Therefore, the SAXS analyses were not performed for these samples.

- It is stated that for Al-samples “reflections have higher intensity indicating the degree of crystallinity increases vs. the concentration of aluminum”. But, intensity of the reflections for the sample SBA-450-30Al is higher than for sample SBA-450-10Al, which has higher content of Al. The same situation is for the positions of the reflections. Please, explain!

1. The sample SBA-450-10Al has higher content of Aluminum and then the sentence “reflections have higher intensity indicating the degree of crystallinity increases vs. the concentration of aluminum” is modified in the manuscript for clarification:

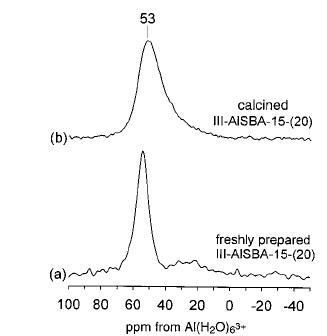
"In the diffraction patterns of SBA-450-(y) samples, reflections of SBA-450-10Al and SBA-450-30Al have higher intensity compared to the pure one. Therefore, the degree of crystallinity enhances by grafting aluminum." (Page 4)

1. The sentence “values for all three reflections (100, 110 and 200) shift to lower 2θ value in case of SBA-450-10Al and SBA-450-30Al samples” seems to be correct and results of SAXS patterns confirmed this conclusion.

- The shifting of diffraction peak to lover 2θ angles is not a proof for tetrahedral coordination of aluminum! Anyway, it should be explained.

Ans. As noted in the manuscript, this conclusion is drawn from the results of Mouli et al. and Luan et al. works (references 13 and 10, respectively). They concluded that:

1. The important observation from the XRD analysis is that in case of Al-SBA-15 samples prepared using the post synthesis modification methodology, 2Ѳ values for all three reflections (100, 110 and 200) shift to lower 2Ѳ value (higher d-spacing). The shifting of 2Ѳ to lower value is well in line with the presumption of tetrahedral co-ordination of Al in the network of Si-SBA-15." Such behavior may be linked with larger Al–O bond length than that of Si–O bond.
2. The 27Al MAS NMR spectra of AlSBA- 15 produced by reacting siliceous SBA-15 with an aqueous solution of sodium aluminate illustrated in below Figure, exhibit only one resolved line at 53 ppm, corresponding to tetrahedral aluminum. This result indicates that all aluminum has been incorporated into the siliceous framework of SBA-15.



- The XRD pattern in Figure 2 is very noise.

Ans. By using smoother tool in Minitab software, a smooth XRD pattern is created and use in Fig. 2 (Page 5).

- Lines 142-145: Explanation for curves offset and shift should be given in bracket at the end of the title of Figure 3.

Ans. It is done. (Page 7)

- It is stated that “SBA-450-y and SBA-550-y samples have a narrow mono-modal pore size distribution centered on 10 nm.” But, the value of the maximum on the pore size distribution (dmax) should be given in Table 1 and compare with the value of the average pore diameter (dav). In that way, pore size distributions can be assessed: the distribution is very narrow if dmax is very close to dav and vice versa!

Ans. The pore diameter distribution curves of SBA-450-(y) and SBA-550-(y) samples indicate the presence of pore diameters from about 2 to 18 nm. But, as shown in Fig. 3, the most of the pores are in the range of about 10–11 nm, which is evident in the corresponding bell diagrams. Therefore, as it is common in literatures, only the average diameter of pores is reported in Table 1, and the wide or narrow pore size distribution is concluded from Fig. 3.

- Line 151: What it means: their pore distribution curves have less uniformity?

Ans. As shown in Figs. 3(e) and 3(f), after grafting the aluminum, the pore size distribution curves for samples that were calcined at 450 and 550 °C were approximately as same as pure ones. However, for samples that were calcined at 350 °C, it was observed that the uniformity of the curve was less than pure SBA-350 sample, and the corresponding curves were not bell-type. Therefore, for clarifying the issue, the related sentence in the manuscript was modified as below:

"Additionally, the pore size distribution curves of SBA-350-10Al and SBA-350-30Al samples have less uniformity than that of the pure one." (Page 6)

- Lines 157 and 158: “due to pore filling of AlO2 species after grafting the aluminum”??? It needs better explanation!

Ans. As the aluminum atoms are grafted on the pore walls of prepared samples, the capillary condensation of these samples decreases and shifts to lower P/P0 values. Therefore, for better explanation, the corresponding sentence was modified in the manuscript as below:

"Besides, N2 adsorption–desorption isotherm of SBA-x-(y) samples exhibits the reduction in the capillary condensation step of Al-containing samples due to filling their pores by AlO2 species." (Page 6)

- Line 166: “increases in consequence with decreasing BET surface and micropore areas”??? The English language should be highly improved!

Ans. This sentence was rewritten as follow:

"Moreover, Table 1 shows that the BET surface areas and total pore volumes of SBA-x-(y) samples decrease due to the probability of forming Al2O3 islands and blocking some pores after incorporating of aluminum into the pure SBA-x samples." (Page 7)

- Line 177: “exhibited a different trend, and its diversity was negligible after Al-grafting” – A different trend in comparison to what? It is not clear what diversity is.

Ans. In order to prevent any misunderstanding, the sentence was modified as follows:

"The BJH pore diameters of SBA-x-(y) samples exhibited a different trend at the same Si/Al mole ratio and did not substantially vary after Al-grafting." (Page 7)

- Generally, the results of nitrogen adsorption-desorption are not discussed well. The English language should be highly improved. The results should be discussed mainly according to the values from the Table 1. The influence of calcination temperature on the textural properties should be also discussed.

1. The N2 adsorption-desorption section is modified based on above mentioned comments (Pages 6 and 7)
2. Since the main purpose of calcination step is the removal of surfactant and change of surface silanol/siloxane groups, it is observed that calcination at 450 and 550 °C does not have any significant effect on the physical properties of the samples. But, calcination temperature has a significant effect on the acidity of the samples which is shown by NH3-TPD results. For explanation the influence of calcination temperature on the textural properties, this sentence is added to manuscript:

"The pure SBA-450 and SBA-550 samples have an equal specific surface area and total pore volume whereas pure SBA-350 sample has minimum surface area due to remaining some part of P123 surfactant in its framework even after calcination at 350 °C." (Page 7)

1. It is our pleasure if the referee has additional points of view on this section, announces it to apply to the article.

- There is no proof in the results of FTIR analysis (just for two samples) for the conclusion “the incorporation of tetrahedrally coordination of aluminium into SBA-15 framework”.

Ans. FTIR analysis is done also for SBA-450-10Al sample and results are added to Fig. 4. (Page 8)

The positions of the main bands should be labelled at the Figure 4 by wavenumbers. The band between 400 and 500 cm-1 is not mentioned.

Ans. The following sentence related to the band between 400 and 500 cm-1 is added to manuscript:

"Moreover, the band at 400-500 cm-1 is assigned to the Si-O bending vibration.” (Page 8)

It is stated that the intensity of the T-O symmetric stretching mode (at 800 and 960 cm-1) is manipulated (?) by the aluminium incorporation. What it means?

Ans. it means that by incorporation aluminum species to SBA-450-10Al and SBA-450-30Al samples, the intensity of FTIR peaks at 800 and 960 cm−1 is varied and shifted to lower Wavenumber. This sentence is rewritten as follows:

"The bands around 800 cm−1 and 960 cm−1 are related to T–O (T= Si and Al) symmetric stretching mode, and their intensities decrease after incorporating aluminum into pure SBA-450 framework." (Page 8)

It is stated: “These bands are due to TO4 vibrations assigned to bending Al–O–Si indicating the incorporation of tetrahedrally coordination of aluminum into SBA-15 framework.” But, the same bands (of higher intensity) are present in the FTIR spectra of SBA without Al and it is not clear why the decreasing of the intensity of the bands indicates tetrahedral incorporation of Al in SBA-15 framework. Therefore, the results of FTIR analysis should be highly improved, including the incorporation of spectra for other samples.

1. It is accepted that the shift in the bands 800 and 960 cm-1 proves clearly the incorporation of Aluminum atoms in the Si-OH framework. Therefore, based on the SAXS and FTIR results, it was concluded that aluminum was tetrahedrally incorporated in SBA-15 framework. However, due to the inability to directly deduce these results from FTIR analysis, this phrase is omitted from the abstract (Page 1) and FTIR results (Page 8) sections.
2. FTIR analysis is done for SBA-450-10Al, and results are added to Fig. 4.

- Line 195: “It is obvious” → “It is well-known”

Ans. This correction was performed in page 8.

- Lines 200-203: It is stated that “the peak at 102-2014 ºc corresponds the weak acidity (mainly contributed to pentahedral aluminum species and surface hydroxyl group attached with Al)”, but this peak is present also in pure SBA, which does not contain Al-species.

Ans. This sentence is corrected and rewrite as below:

"The first peak from 102 °C to 214 °C corresponds to the weak acidity (mainly contributed by pentahedral aluminum species and surface hydroxyl group attached with Si or Al)," (Page 9)

- Lines 204-205: “the temperature attributed to the week acidity is shifted to the higher temperature” → “the position of the peak attributed to the weak acidity is shifted to higher temperatures”

Ans. It is done in page 9.

- The discussion from lines 207 to 231 is difficult to follow and understand. It needs to improve highly both the discussion and the English language.

Ans. The discussion and the English language of this section were totally improved (Pages 9-11)

Why the first sentence in (iii) is incorporated in the discussion? In which connection is this sentence with the next: Coversely,…

Ans. This sentence is incorporated in the discussion to say that although zeolites with a high number of sites are widely used in the hydrocracking process, by altering the synthesis conditions and use of Al-grafting, the SBA-15 with high acidity can be synthesized. However, the sentence in order to maintain the integrity of the text is deleted.

What is optimum acidity (line 223)?

Ans. The optimum acidity is determined by the type of process. For hydrocracking processes requiring high number of acid sites, calcination at 450 °C with Si/Al mole ratio of 30 is suitable, while to obtain a fewer number of acid sites the calcination at the temperature of 550 °C with Al/Si molar ratio of 0.033 is suggested (like hydrotreating processes). However, by improving discussion and the English language of this section, “optimum acidity” is omitted.

What is medium acidity (line 226)? How this acidity was determined?

Ans. Due to the highest and lowest number of acidic sites, the results were divided into three intervals: low, medium and high. Therefore, samples that calcined at 450 and 550 °C with Si/Al mole ratio of 30 are in the range of high and medium, respectively. Therefore, for clarifying the issue, the related paragraph in the manuscript was modified as below:

"Furthermore, Fig. 6 shows that calcination at 450 °C with Al/Si molar ratio of 0.033 is recommended for catalysts requiring high number of acid sites." (Page 11)

What it means: “Varying acidity at even high aluminum loading without any significant variation in physical and structural properties of prepared samples may be rationalized by the easy diffusion of aluminum precursor to the mesochannels of parent SBA-x particles during Al-grafting.” Is it better to have strong or weak acidity?

Ans. In previous works, it is observed that physical properties (especially specific surface area and total pore volume) of samples are decreased dramatically by increasing Al-loading (reference 10). But in this work due to suitable size of pores (in range of 10-11 nm), there is no significant change in the physical properties of synthesized samples with increasing aluminum content. Therefore, for clarifying the issue, the related paragraph in the manuscript was modified as below:

"The acidity of Al-grafted samples is varied even at high aluminum content, but there is no significant variation in physical and structural properties of these samples. The main reason for this phenomenon is the facilitated diffusion of aluminum precursor to mesochannels of pure SBA-x particles during Al-grafting." (Page 10)

As mentioned above, type of process determines number of required acid site (low, medium and high)

Additional comment:  
- there is no analysis of the samples to prove Al incorporation (chemical analysis) and type of Al incorporation (for example 27Al MAS NMR)

Ans. Solid 27Al MAS NMR analysis is one of the most reliable methods for detecting aluminum phases in synthesized samples. Unfortunately, performing this test is not possible due to the limited analysis facilities. However, attempts have been done by authors to interpret and justify the results using N2, FTIR, SAXS, XRD and NH3-TPD analyses.

**Reviewer B:**

Is the quality of the figures (including legends and axes labelling) satisfactory?

Ans. The quality of all figures is improved.

ADDITIONAL COMMENTS  
Section: Experimental  
Line 77: space group P6mm (P capital letter italic, m italic) instead p6mm.

Ans. This correction was performed in page 3.

Line 101: CuKα radiation (K italic) instead CuKα.

Ans. This correction was performed in page 4.

Section: Results and Discussion  
Line 123: space group P6mm (P capital letter italic, m italic) instead of p6mm.

Ans. This correction was performed in page 4.

Line 139: Fig. 2 Only powder XRD pattern of the SBA-450-10Al sample is presented in Fig. 2. It is necessary to add XRD pattern of SBA-450-30Al sample to confirm that both samples containing the amorphous silica. The diffractogram of SBA-450-30Al sample should be added to the same diagram. Only after this revision, plural nouns "…diffractograms…" (Line 136) and "…patterns…" (Line 140) should be correct, as it stated in submitted paper.    
Ans. Since no peak was observed at the highest aluminum loading sample (SBA-450-10Al), it can be concluded that the sample with the lower aluminum content (SBA-450-30Al) also did not have any peak. Therefore, it is not necessary to perform the relevant analysis for this sample. If this explanation is acceptable, we do not change RESULTS AND DISCUSSION section. Otherwise, we should perform XRD analysis for SBA-450-30Al sample and justified its results based on your comment.