**Journal of Zankoy Sulaimani**
Part-A- (Pure and Applied Sciences)
http://jzs.univsul.edu.iq

# Synthesis and characterizations of zeolite catalyst from natural quartz in Kurdistan Region-Iraq

Heman A.Smail [1*]

[1]*Department of Chemistry, College of Science, University of Salahaddin, Erbil, Kurdistan Region, Iraq*
**Corresponding author's e-mail: heman.smail@su.edu.krd*

**Article info**

Original: 10 August 2020
Revised: 28 October 2020
Accepted: 29 November 2020
Published online: 20 June 2021

***Key Words:***
Natural quartz, NaK-LSX, zeolite, hydrothermal synthesis

**Abstract**

In this work a cheap and an affordable natural quartz powder was used as a silica source, converted to a sodium silicate solution, for synthesising the low silica kind X (LSX) with process of hydrothermal the zeolite LSX with the formula $Na_{73}K_{22}[Si_{97}Al_{95}O_{384}]$ . $212H_2O$. The synthesized zeolite with the types cations of K and Na, considered as NaK-LSX, the best time to complete crystallization was 3.0 h, was features of the agent performing Fourier transforms infrared spectroscopy (FT-IR) and X-ray diffraction (XRD), that affirmed the synthesis effectiveness. Images of field emission scanning electron microscope (FE-SEM) in the (NaK-LSX) illustrated particles of multi-faceted spherulite composing polycrystal particles that have various measures as well as particles of small amorphous. Measuring of the average pore size and, volume of total pore for sample utilizing the Barrett-Joyner-Halenda (BJH) technique, the particle magnitude spread over an area of NaK-LSX about 67nm, and nitrogen adsorption is indicating an area of surface about 732.29 $m^2/g$ applying Brunauer-Emmett-Teller (BET) technique. The synthesizes zeolite (LSX) may have different kind of usages including, industrial applications as a gas or vapour adsorption, separation and as a catalyst.In addition they may also be used as a selective adsorbent in purifying polluted air.

## 1.Introduction

Quartz is the naturally occurring inorganic substance kind of $SiO_2$ stable in pressures and temperatures of low. Its English form a specified derived from the querkluftertz (cross-vein ore), a Saxon word [13]. It occurs in hydrothermal mineral, metamorphic, sedimentary, and igneous environments, average within belonging to regions of continental. Nevertheless in oceanic rocks it is not frequent. Based on its acentric structure, it happens in both kinds of right and left -handed and is pyroelectric and piezoelectric. It is often approximately pure and obtains just less measures of other substitutions. Keatite, stishovite, coesite, cristobalite, tridymite, and β-quartz are polymorphs.

Zeolites are aluminosilicates of crystalline made of tetrahedral $TO_4$ units (T= Al or Si) with accepted characteristics of hydrothermal firmness, high thermal, strong Brønsted acidity, high ion-exchange capacity, shape selectivity, precise microporosity and uniform and high surface area ([14, 30]). Thus, it has been applied in different environmental as well as industrial situations including exchange of ion ([11, 31]) catalysts ([8, 26, 28]), membrane separations ([3, 37]) and adsorbents ([2, 5, 12]). Based on Auerbach *et al.* [4] zeolite LSX is inside family of silicate minerals faujasite (FAU) household in the company of a skeleton hold dual six circles joined all the time sodalite enclosure that caused super cages escorted by inlet 7.4 Å diameters. The model Si/Al proportion of LSX is from 1 to 1.5. It is considered as group $F_{3m}$ of space. The main raw materials applied in the synthesizing the zeolites are various alumina and silica sources that are usually gathered from aluminium salts, sodium aluminate ($NaAlO_2$), and silicates of sodium. Nevertheless, old techniques of synthesis of zeolites usually include reagents of chemical as crystallization or begining materials from a clear solution or gel in conditions of hydrothermal that have the benefits of environmental

unfriendly nature, excessive waste, and high cost. Thus, more endeavors are performing for zeolites economical synthesizing. Generally, solid wastes of industry and minerals of silicate and natural aluminosilicate have been examined as source of alumina and/or silica since they are precursors of less costy and could cause reducing the expense of synthesizing. Several researches have conducted about the zeolites synthesis from natural minerals including kaolinite [1, 16], bentonite [16], feldspar [38] and other parents [7, 24, 27, 34]. Though synthesizing zeolites from the solid wastes, including fly ash [32, 40], rice husk ash [29] and coal gangue minerals [34], the unreliability in their contributes and, the contamination in their constituents may increase maximum their a reasonable purpose. Thus, direct synthesizing zeolites from silicate minerals and natural aluminosilicate with various reserves in the earth has been pursued due to its major potential for probably changing the characteristics of the obtaining zeolites, energy saving, and decreasing the dangerous wastes production [25]. Although, majority silicate alumino minerals are inoperative, limited their usage in practice for the synthesizing zeolite. In addition, following the thermal activating, just a section of the bonds of aluminum-oxygen could be broken that is a section of the Aluminum oxide ($Al_2O_3$) and a less measure of Silicon dioxide ($SiO_2$) could take part in the synthesizing zeolite. LSX synthesizing using a natural quartz as a silica source has not been reported.

## 2.Materials and methods

### 2.1. Chemicals

Quartz was collected from Iraqi-Kurdistan region in Erbil-Warte(latitude: 36º30'1.62"N and longitude: 44º45'15.67"E) used as a silica source for synthesis. Sodium aluminate (93% of $NaAlO_2$, Fluka), sodium hydroxide (99% NaOH, Aldrich), potassium hydroxide (90% KOH: Flake lab. grade).

### 2.2. Hydrothermal Synthesis

The LSX, according to Kühl [20], was produced by the hydrothermal method reported beyond. The solution of sodium silicate was created with regular addition of qusrtz to 150 mL of 16% wt NaOH solution in stirring until achieving a solution of homogeneous. In the process of LSX synthetic, sodium aluminate was dissolved in D.W and regularly enhanced in a compound of NaOH and KOH. The recent settlement was assorted by a liquid silicate-sodium suspension. The resulting solution was moved into the polypropylene bottle, cover on and secured sealed by film of paraffin. Crystallization and aging were conducted at 75 °C for 4 h without stirring, later, adjusted to 110 °C (for 0.5, 1.0, 2.0 and 3.0 h) for completing crystallization; the sample was cooled down to 20 °C and cleaned with D.W and 0.02 normality NaOH solution, and dried at 115 - 135 °C for 24 h. The result was zeolite LSX in the appearance of Na and K cations and was designated as LSX-NaK all around this object .

## 3. Characterization NaK- LSX

### 3.1. X-Ray Diffraction

The homogeneous solid substance building of cooking zeolite catalyst was identified at 20 °C by XRD (D5000 by the diffraction of X-rays, Germany) utilizing Bragg-Brentano the shape, λ = 0.154 nm CuKα moving subatomic particles at a current of 35 mA, and 45 kV. The powder catalyst was added to the sample holder and scanned at 2θ angle between 5º to 50º and a 0.05º angular step size [17]. The amount of crystallinity was calculated [39] through Equation (1).

$$\text{Crystallinity}\% = \frac{\sum_{Intensity\ of\ peaks\ of\ catalyst}^{12}}{\sum_{\text{Intensity of peaks of standard}}^{12}} * 100 \qquad (1)$$

### 3.2. Fourier Transform Infrared Spectroscopy

Groups of functional in the NaK-LSX catalysts of zeolite were identified with spectrometer of FT-IR (USA, MA, Waltham, PerkinElmer, Perkin Elmer model Spectrum One) taken as approximately 20 °C. At least 36 scans were carried out by an average infrared signal by a resolution of 5 $cm^{-1}$ between 400 to 4000 $cm^{-1}$ and the final outcome was in transmittance%. The trial was prepared with blend 6 mg of prepared catalyst in 250 mg of KBr pressed movement a disk of FT-IR calculation.

### 3.3. Nitrogen adsorption-desorption

The specific outside areas (BET), pore dimension of NaK-LSX, and inlet-volume was founded in $N_2$ adsorption-desorption isotherm at -195.75 °C by a zeolite catalyst was obtained according to the (BET) procedure, engage micromeritics (3FLEX, GA, USA). The catalyst was taken pre-treated at 350 °C for 4 h in a vacuum for dehydrating the catalysts before analysis. Tidy measurement of area of BET surface, the $N_2$ adsorption isotherms ($\Delta G_{ads.} = RT(\ln P_{ads.} - \ln P_o)$) detail above the correlative pressure scope of 0.06–0.33 was applied in sample of prepared catalyst. Nevertheless, desorption isotherm ($\Delta G_{des.} = RT\ (\ln P_{des.} - \ln P_o)$) was applied for measuring the mean pore size as well as, total pore amount for sample by the technique of Barrett-Joyner-Halenda (BJH). Since $P_{des.} < P_{ads}$ and $\Delta G_{des.} < \Delta G_{ads.}$ Therefore, the desorption amount of relative pressure corresponds to the more stable adsorbate condition and that is why desorption isotherms must be applied in analysis of pore size [15].

### 3.4. Morphology

The morphological and dimension attribute in prepared zeolite catalysts was evaluated using a (FE-SEM, ZEISS SIGMA-Belgium). A few particles of NaK-LSX catalyst was distinctly dispersed in acetone, later, one drop of the suspended catalyst was put on a sample holder and coated with Platinum (Pt) before analysis for removing the charging impact in the electron beam.

## 4. Results and discussions

The development of the NaK-LSX framework was established beside the XRD differentiate accompanied by standard NaX (Figure 1). The prepared catalyst sample obtained peaks at locations like the NaX quality, resulting in the faujasite formula development, and showing high crystallinity in the sharp peaks. The comparative crystallinity to the standard NaX was examined from the XRD image in equation (1) and, the achieved amount was about 100%. The particular of comparable strength, d-spacing and peak locations of NaK-LSX (standard NaX are showen in Table 1).

Although the XRD peaks of NaK-LSX was identical to the standard NaX, the sequence of intensities was not the same. It was predicable since our prepared sample of catalyst makes together Na and K cations, there is only the Na cation in the interval standard. Based on the previous report the sequence of peak intensities depended greatly on cations kind [10, 19] and the existence of $K^+$ attenuated them. Moreover, according to Kühl [20], a less proportion of Si/Al increased the intensities of line.

The synthesized NaK-LSX was distinguished for identifying functional groups of the structure with FT-IR. Figure 2 illustrates a great peak at 950 $cm^{-1}$ it was assigned to an asymmetric T-O stretching. Lee *et al*. [23] stated that usually, T-O is a tetrahedral atom alluded to the framework of Al, Si composition. It might transfer to a less frequent by expanding some tetrahedral Al atoms. Bands in the region 573 - 460 $cm^{-1}$, 692 $cm^{-1}$ and 774 $cm^{-1}$ were distributed mainly to T-O bending vibrations, double ring and symmetric stretching, respectively. It was mentioned that a band at 602 - 500 $cm^{-1}$ was relating to the topological arrangement of secondary units of structure in zeolites, which include the double 4 and 6 rings external linkage peak associated with the FAU structure, in addition, observed in zeolite structures ([23, 36]). According to [9] the band with a peak at 3,490 $cm^{-1}$ was assigned to OH stretching, and the shake at 1,640 $cm^{-1}$ was mentioned to bending vibration of an adsorbed water molecule.

The nitrogen adsorption/desorption isotherm of (NaK-LSX) zeolite catalyst is illustrated in Figure 3(a). It was type (I) according to International Union of Pure and Applied Chemistry (IUPAC)'s categorization with a large nitrogen uptake at low pressure and the desorption almost overlapping with the adsorption. The step enhanced in $N_2$ adsorption by an enhanced relative pressure, $P/P_0$ and $N_2$ adsorbed measure reaching 7.0 $cm^3/g$ at $P/P_0 = 0.047$ recommending the pressure of an appreciable measure of micropore on the NaK-LSX surface. As explained by Langmi *et al*. [21], this outcome was typical for the zeolite structure, and zeolite X has an greatly open framework and an abundant cation does not limit the entries to inner pores. Furthermore, the impact of a cation on the pore structure and surface area illustrate that the processes of ion-exchange can cause various modifications in surface and pore structure and these ions inhibited the movement of nitrogen

molecule into pores or the so-called pore blocking impact, leading to the decline in surface area ([18]; [22] and [35]).

Figure 3(b) illustrates the increasing amount of pore and spreads of pore size of LSX-NaK from $N_2$ adsorption. The pore diameter of NaK-LSX, illustrated in Figure 3(b), was measured with the Horváth-Kawazoe (HK) technique was developed for determining the pore size distribution with a pore volume of 0.065 $cm^3/g$. However, the curves of pore size distribution examined from desorption data with using the model of BJH illustrated in Figure 3(b), exhibiting a narrow pore size distribution with the greatest pore size at 67 nm. Nevertheless, a shoulder peak distribution is dominant and it was probably from interconnected surface pores of LSX [6, 23].

Table 2 illustrates outcomes of a $N_2$ adsorption investigation such as mean pore diameters of LSX-NaK, pore volumes, and the BET surface areas. Langmi *et al*. [21] noted that the pore amount of NaK-LSX and BET outside area form natural quartz was gripping when carrying material for preparing catalysis or other usages.

FESEM studied size and shape of NaK-LSX particles synthesized from natural images and quartz which are illustrated in Figure 4 by the amplification of 5.00 KX. In the smaller magnification of the image, the solid production included a mixture of multi-faceted spherulite crystals with a particle diameter about 6.5 - 10 μm as well as round amorphous particles with a particle diameter about 5.4 μm. The multi-faceted spherulite particles as well as small particles was equabled of polycrystal particles that had various sizes. Since some particles clearly related to other particles [33].

## Conclusions

The chemical composition, morphology of crystal, distribution and size of crystal, as well as rate of crystallization relates to the mole fraction of K in the Na-K system while synthesizing LSX. An affordable and a cheap natural quartz was utilized as original silica for the synthesizing of catalyst (LSX-NaK). The zeolite structure forming was affirmed with FTIR and XRD. LSX-NaK crystal morphology was feature spherulite particles illustrates with micrograph of FE-SEM. The area of BET surface in the NaK-LSX was about 732.29 $m^2/g$. The chemical composition, morphology of crystal, distribution and size of crystal, as well as rate of crystallization relates to the mole fraction of K in the system of Na-K while synthesizing LSX. The lofty mole fraction of K in the beginning gels developed in lower crystallization rate, and smaller framework Si/Al quotient in the LSX got. The synthesizes zeolite (LSX) may have a wide range of applications including, industrial applications as a gas or vapour adsorption, separation and as a catalyst.In addition they may also be used as a selective adsorbent in purifying polluted air.

**Acknowledgements:** The Author acknowledges to Ibnu sina institute for fundamental science studies University of UTM/Malaysia.

## References

[1] Alaba, P.A., Sani, Y.M., Mohammed, I.Y., Abakr, Y.A. and Daud, W. *"Synthesis and characterization of sulfated hierarchical nanoporous faujasite zeolite for efficient transesterification of shea butter*". Journal Cleaner Production. Vol. 142, pp. 1987-1993. (2017).

[2] Alver, E. and Metin, A.Ü. *"Anionic dye removal from aqueous solutions using modified zeolite:Adsorption kinetics and isotherm studies*". Chemical Enginering Journal. Vol. 200-202, pp. 59-67. (2012).

[3] Arefi Pour, A., Sharifnia, S., Neishabori Salehi, R. and Ghodrati, M. *"Adsorption separation of CO /CH on the synthesized NaA zeolite shaped with montmorillonite clay in natural gas purification process*". Journal of Natural Gas Science and Engineering. Vol. 36, pp. 630-643. (2016).

[4] Auerbach, S.M., Carrado, K.A. and Dutta, K.P. *"Handbook of Zeolite Science and Technology"*. 1st ed. Marcel Dekker Inc: New York,1p. (2003).

[5] Benaliouche, F., Hidous, N., Guerza, M., Zouad, Y. and Boucheffa, Y. *"Characterization and water adsorption properties of Ag-and Zn-exchanged A zeolites*". Microporous Mesoporous Materials. Vol. 209, pp. 184-188. (2015).

[6] Chang, F.W., Kuo, W.Y. and Yang, H.C. *"Preparation of $Cr_2O_3$-promoted copper catalysts on rice husk ash by incipient wetness impregnation*". Applied Catalysis A-General. Vol. 288, pp. 53-61. (2005).

[7] Chen, D., Hu, X., Shi, L., Cui, Q., Wang, H.and Yao, H. *"Synthesis and characterization of zeolite X from lithium slag*". Applied clay science.Vol. 59-60, pp. 148-151. (2012).

[8] Doyle, A.M., Alismaeel, Z.T., Albayati, T.M. and Abbas, A.S. *"High purity FAU-type zeolite catalysts from shale rock for biodiesel production*". Fuel. Vol. 199, pp. 394-402. (2017).

[9] Dyer, A., Tangkawanit, S. and Rangsriwatananon, K. *"Exchange diffusion of $Cu^{2+}$, $Ni^{2+}$, $Pb^{2+}$, and $Zn^{2+}$ into analcimesynthesized from perlite"*. Micropore Mesopore Materials. Vol. 75, pp. 273-279. (2004).

[10] Esposito, S., Ferone, C., Pansini, M., Bonaccorsi, L. and Proverbio, E. *"A comparative study of the thermal transformations of Ba-exchanged zeolites A, X and LSX*". Journal of the European Ceramic Society. Vol. 24, No. 2, pp. 689-697. (2004).

[11] Fan, M., Sun, J., Bai, S. and Panezai, H. *"Size effects of extraframework monovalent cations on the thermal stability and nitrogen adsorption of LSX zeolite*". Microporous Mesoporous Materials. Vol. 202, pp. 44-49. (2015).

[12] Feng, P., Zhang, G., Zang, K., Li, X. and Xu, L., Chen X. *"A theoretical study on the selective adsorption behavior of dimethyl ether and carbon monoxide on H-FER zeolites"*. Chemical physics letters. Vol. 684, pp. 279-284. (2017).

[13] Gaines, R.V., Skinner, H.C.W., Foord, E.E., Mason, B. A., Rosenzweig, V.T., King and Dowty, E. "*Dana's New Mineralogy"*. 8th ed. New York, John Wiley and Sons, 1819 pp. (1997).

[14] Garcia, G., Cardenas, E., Cabrera, S., Hedlund, J.and Mouzon, J. *"Synthesis of zeolite Y from diatomite as silica source*". Microporous Mesoporous Materials. Vol. 219, pp. 29-37. (2016).

[15] Gardy, J. *"Biodiesel Production from Used Cooking Oil Using Novel Solid Acid Catalysts"*. Ph.D. Thesis University of Leeds, Leeds, UK. (2017).

[16] Garshasbi, V., Jahangiri, M. and Anbia, M. *"Equilibrium CO adsorption on zeolite 13X prepared from natural clays*". Applied Surface Science. Vol. 393, pp. 225-233. (2016).

[17] Harding, G. *"X-ray diffraction imaging-A multi-generational perspective*". Applied Radiation and Isotopes. Vol. 67, pp. 287-295. (2009).

[18] Huang, F.-C., Lee, J.-F., Lee, C.-K. and Chao, H.-P. *"Effects of cation exchange on the pore and surface structure and adsorption characteristics of montmorillonite*". Colloids and surfaces A. Vol. 239, pp. 41-47. (2004).

[19] Joshi, U.D., Joshi, P.N., Tamhankar, S.S., Joshi, V.P., Idagec, B.B., Joshid, V.V. and Shiralkar, P.V. *"Influence of the size of extra framework monovalent cations in X-type zeolite on their thermal behavior"*. Thermochimica Acta. Vol. 387, pp. 121-130. (2002).

[20] Kühl, G. *"Low silica type X. In: Verified Syntheses of Zeolitic Materials"*. 2nd ed.Robson, H. (ed). Elsevier Science B.V. and Amterdam. pp. 153-154. (2001).

[21] Langmi, H.W., Book, D., Walton, A., Johnson, S.R., Al-Mamouri, M.M., Speight, J.D., Edwards, P.P., Harris, I.R. and Anderson, P.A. *"Hydrogen storage in ionexchanged zeolites*". Journal of Alloys and Compounds. Vol. 404-406, pp. 637-342. (2005).

[22] Langmi, H.W., Walton, A., Al-Mamouri, M.M., Johnson, S.R., Book, D., Speight, J.D., Edwards, P.P., Gameson, I., Anderson, P.A., and Harris, I.R. *"Hydrogen adsorption in zeolites A, X, Y and RHO*". Journal of Alloys and Compounds. Vol. 356-357, pp. 710-715. (2003).

[23] Lee, H.J., Kim, Y.M., Kweon, O.S. and Kim, I.J. *"Structural and morphological transformation of NaX zeolite crystals at high temperature*". Journal of European Ceramic Society. Vol. 27, No. 2-3, pp. 561-564. (2006).

[24] Li. C., Zhong, H., Wang, S., Xue, J. and Zhang, Z. *"Removal of basic dye (methylene blue) from aqueous solution using zeolite synthesized from electrolytic manganese residue".* journal of industrial and engineering chemistry. Vol. 23, pp. 344-352. (2015).

[25] Li, T., Liu, H., Fan, Y., Yuan, P., Shi, G., Bi, X.T.and Bao, X. *"Synthesis of zeolite Y from natural aluminosilicate minerals for fluid catalytic cracking application*". Green Chemistry. Vol. 14, pp. 3255-3259. (2012).

[26] Liu, H., Lu, G., Guo, Y., Guo, Y. and Wang J. *"Deactivation and regeneration of TS-1/diatomite catalyst for hydroxylation of phenol in fixed-bed reactor*". Chemical Enginnering Journal. Vol. 108, pp. 187-192. (2005).

[27] Ma, D., Wang, Z., Guo, M., Zhang, M. and Liu, J. *"Feasible conversion of solid waste bauxite tailings into highly crystalline 4A zeolite with valuable application*". Journal of Waste Management. Vol. 34, pp. 2365-2372. (2014).

[28] Maihom, T., Wannakao, S., Boekfa, B. and Limtrakul, J. *"Density functional study of the activity of goldsupported ZSM-5 zeolites for nitrous oxide decomposition*". Chemical physics letters. Vol. 556, pp. 217-224. (2013).

[29] Manadee, S., Sophiphun, O., Osakoo, N., Supamathanon, N., Kidkhunthod, P., Chanlek, N.,Wittayakun, J. and Prayoonpokarach, S. *"Identification of potassium phase in catalysts supported on zeolite NaX and performance in transesterification of Jatropha seed oil".* Fuel processing technology. Vol. 156, pp. 62-67. (2017).

[30] Marcos, A., Mohuli., Sudipta, D., Andrea, N., Nattan, R., Paulo, R. and Carlos, A. *"Comparative study using different external sources of aluminum on the zeolites synthesis from rice husk ash*". Materials Research Express. Vol. 7, pp. 1-19. (2020).

[31] Murad, S., Jia, W. and Krishnamurthy, M. *"Molecular simulations of ion exchange in NaA zeolite membranes".* Chemical physics letters. Vol. 369, pp. 402-408. (2003).

[32] Musyoka, N.M., Ren, J., Langmi, H.W., North, B.C. and Mathe, M. *"A comparison of hydrogen storage capacity of commercial and fly ash-derived zeolite X together with their respective templated carbon derivatives*". International journal of hydrogen energy. Vol. 40, pp. 12705-12712. (2015).

[33] Pongtanawat, K., Sanchai, P. and Jatuporn, W. *"Synthesis and characterization of zeolite LSX from rice husk silica*". Suranaree Journal of Science and Technology. Vol. 14, No. 4, pp. 367-379. (2007).

[34] Qian, T. and Li, J. *"Synthesis of Na-A zeolite from coal gangue with the in-situ crystallization technique*". Advanced Powder Technology. Vol. 26, pp. 98-104. (2015).

[35] Rakoczy, R.A. and Traa, Y. *"Nanocrystalline zeolite A: synthesis, ion exchange and dealumination*". Microporous and Mesoporous materials. Vol. 60, pp. 69-78. (2003).

[36] Sang, S., Liu, Z., Tian, P., Liu, Z., Qu, L. and Zhang, Y. *"Synthesis of small crystals zeolite NaY*". Materials Letters. Vol. 60, pp. 1131-1133. (2006).

[37] Selishchev, D.S., Kolinko, P.A. and Kozlov, D.V. *"Adsorbent as an essential participant in photocatalytic processes of water and air purification: Computer simulation study*". Applied Catalysis A: General. Vol. 377, pp. 140-149. (2010).

[38] Su, S., Ma, H. and Chuan, X. *"Hydrothermal synthesis of zeolite A from K-feldspar and its crystallization mechanism".* Advanced Powder Technology. Vol. 27, pp. 139-144. (2016).

[39] Tangkawanit, S. and Rangsriwatananon, K. *"Synthesis and kinetic study of zeolite from Lopburi perlite".* Suranaree Journal of Science and Technology. Vol. 12, pp. 61-68. (2004).

[40] Volli, V. and Purkait, M.K. *"Selective preparation of zeolite X and A from flyash and its use as catalyst for biodiesel production".* Journal of Hazardous Materials. Vol. 297, pp. 101-111. (2015).

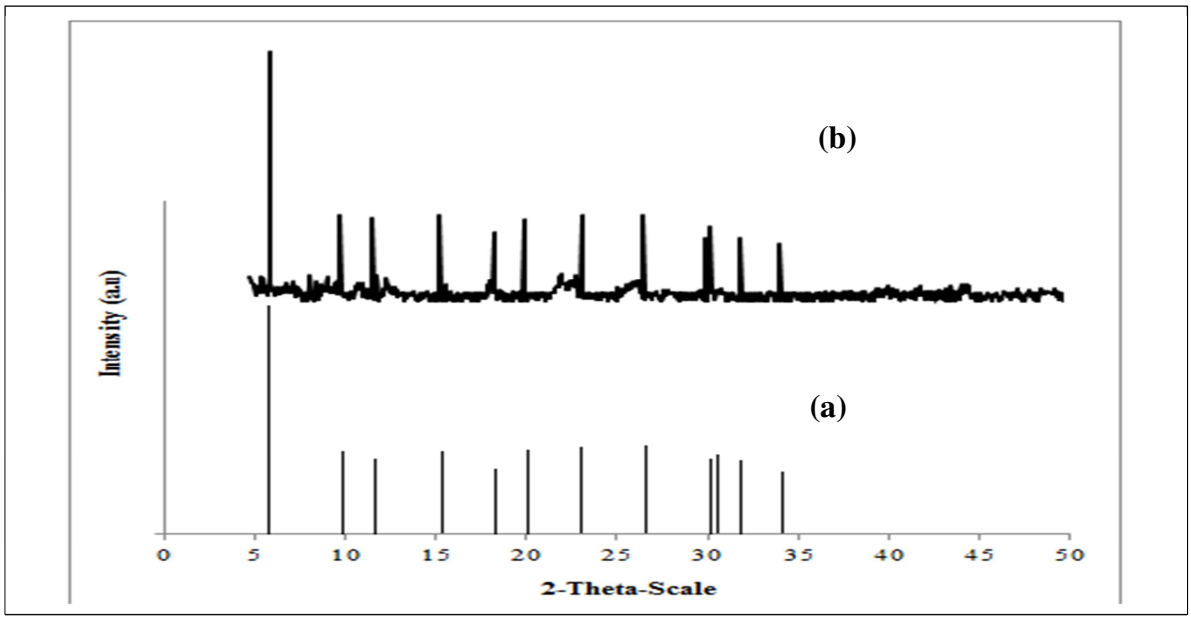


Figure (1): XRD patterns of (a) reference sample and(b) synthesized NaK-LSX catalyst zeolite using natural quartz as a silica source.

Table 1.Peak locations, d-spacing and respective strength of NaK-LSX catalyst zeolite and then standard NaX.

| 2θ | d | Relative intensities | |
|---|---|---|---|
| | | Standard NaX | Synthesized LSX |
| 6.10 | 14.47 | 100.00 | 100.00 |
| 9.99 | 8.85 | 22.4 | 17.28 |
| 11.73 | 7.54 | 16.54 | 17.78 |
| 15.43 | 5.74 | 22.7 | 13.11 |
| 18.43 | 4.81 | 7.18 | 4.21 |
| 20.09 | 4.42 | 12.60 | 6.91 |
| 23.32 | 3.81 | 21.00 | 20.00 |
| 26.70 | 3.34 | 22.10 | 29.41 |
| 30.10 | 2.88 | 20.40 | 26.20 |
| 30.36 | 2.94 | 11.21 | 13.11 |
| 32.03 | 2.79 | 8.62 | 17.28 |
| 34.24 | 2.62 | 4.89 | 6.18 |

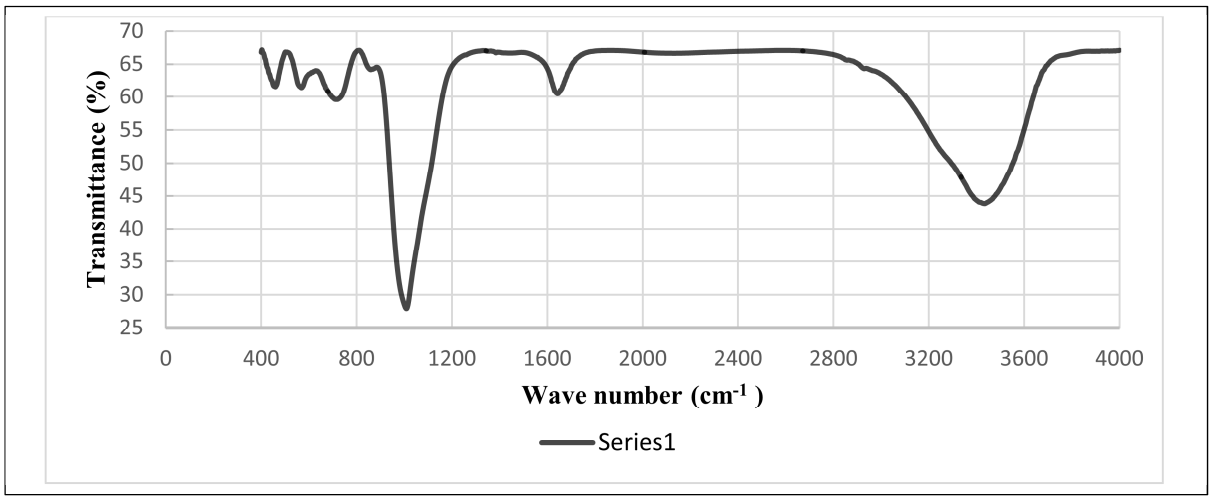


Figure 2.FTIR spectrum of zeolite NaK-LSX synthesized from natural quartz.

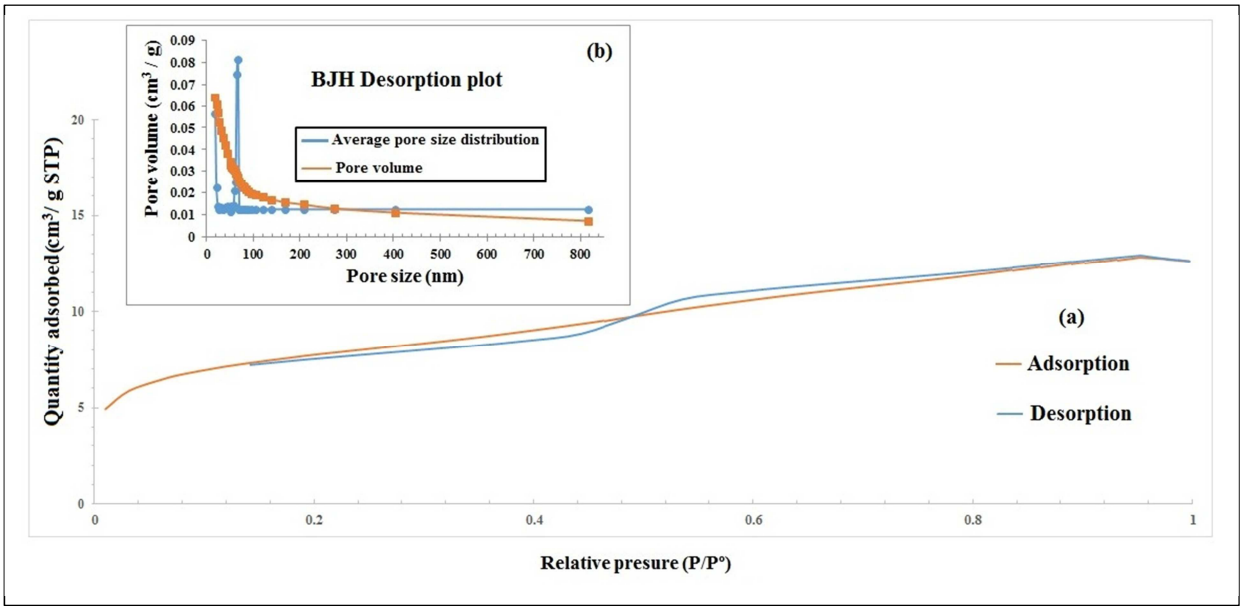


Figure 3(a)Adsorption-Desorption isotherm of zeolite catalyst NaK-LSX (b)Average pore size distribution.

Table 2. The surface properties of synthesized NaK-LSX zeolite catalyst.

| **Textural properties** | **Value** ($m^2/g$) |
|---|---|
| BET Surface area [a] | 732.29 |
| External surface area [b] | 376.11 |
| Micro pore surface area [b] | 365.18 |
| Langmuir surface area [c] | 599.30 |

*[a] Multipoint BET analysis, [b] BJH analysis (t-method),[c] t-method.*

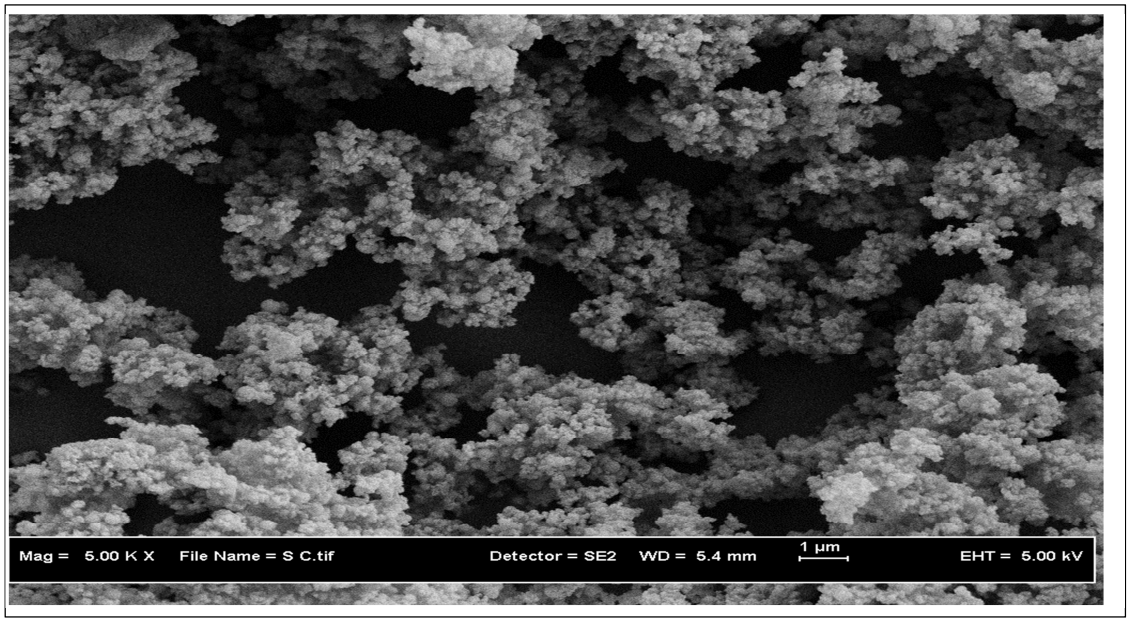


Figure 4. FESEM image of NaK-LSX zeolite catalyst.