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(71) Applicant: **RELIANCE INDUSTRIES LIMITED** [IN/IN]; 3rd Floor, Maker Chamber-IV, 222, Nariman Point, Mumbai 400021, Maharashtra (IN).

(72) Inventors: **SHARMA SUMEET KUMAR**; Village- Surjawali, Near to Salempur, District- Bulandshahr, Uttar Pradesh, 203001 (IN). **METTU ANILKUMAR**; Palapadu (po), Narasaraopet (MD), Guntur (dt), Andhrapradesh, 522603 (IN). **SHARMA NAGESH**; B-13, Akshardham Duplex, New Sama Road, Vadodara - 390 024, Gujarat (IN). **JASRA RAKSH VIR**; B-245, Sundervan Housing Society, New Sama Road, Vadodara - 390024 (IN).

(74) Agent: **DEWAN Mohan**; R. K. Dewan & Company, Trade Mark & Patent Attorneys, Podar Chambers, S.A.Brelvi Road, Fort, Mumbai 400 001, Maharashtra (IN).

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(54) Title: A MODIFIED FAUJASITE ZEOLITE

(57) Abstract: The present disclosure relates to a modified faujasite zeolite comprising at least one zeolite exchanged with at least two alkali metals, wherein, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:15, the amount of sodium in the modified zeolite is in the range of 3 to 10 wt%, the particle size of said modified faujasite zeolite ranges between 20 and 200 micron, and the sum total of at least two alkali metals in said modified faujasite zeolite is in the range of 0.1 to 97 wt%. The present disclosure also relates to a process for preparing the modified faujasite zeolite and the process for alkylating the alkyl group of the aromatic hydrocarbon using the modified faujasite zeolite.

A MODIFIED FAUJASITE ZEOLITE

FIELD OF THE DISCLOSURE:

The present disclosure relates to a modified faujasite zeolite. The present disclosure also relates to a process for preparing a modified faujasite zeolite.

BACKGROUND:

Zeolites are crystalline metallosilicates mainly aluminosilicates based on alkali or alkaline earth metals with an uniform pore size which may be represented by an empirical formula $M_2O.Al_2O_3.xSiO_2.yH_2O$

> wherein M represents an alkali metal cation, x is an integer from 2.5 to 6, and y is an integer from 6 to 9.

These pores of uniform size create a ring system which allows the zeolite to selectively interact with the molecules of particular dimensions and shapes. The size of the ring system in zeolites depends on the number of oxygen atoms present in the ring system. Of the various zeolites, the zeolites having intermediate to large size ring system are used for various applications in the industry. For instance, large pore zeolites such as faujasite zeolites X and ultrastable Y are widely used in petroleum refining processes such as FCC and hydrocracking.

Additionally, surface acidity and basicity of a zeolite determines its usefulness. Various attempts have been made in the past to prepare zeolites having balanced acidic and basic properties.

US20110270006 discloses a process for preparing the metal exchanged zeolite by reacting cesium hydroxide (CsOH) as a first promoter and nitrate of the second promoter selected from the group consisting of Fe, Cr, Ce, Mo, Sn, Bi, Ag, Cu, and combinations thereof with a zeolite substrate.

US5015796 discloses a process for preparing an alkaline earth metal exchanged zeolites by reacting nitrates of alkaline earth metals with zeolites.

Another US patent number US4483936 discloses a zeolite catalyst comprising at least one alkali metal selected from the group consisting of Cs, K, and Rb; at least one metal selected from the Group of metals consisting of Li, Ce, Cr, and Ag; and at least one member selected from the group consisting of boron and phosphorus. It also discloses a process for the preparation of said zeolite catalyst. In the process, hydroxide of the alkali metal is reacted with a zeolite to obtain a partially modified zeolite which is reacted with Boron phosphate and then with nitrate of metal to obtain said catalyst composition.

US4463204 discloses a zeolite selected from the group consisting of sodium form of zeolite X or Y exchanged first with potassium and then with cesium. The sodium in the zeolite is at least 90% exchanged for potassium and cesium.

The article titled “catalytic performance of X molecular sieve modified by alkali metal ions for the side-chain alkylation of toluene with methanol” published in Microporous and Mesoporous Materials 167 (2013) 213-220; discloses molecular sieve catalysts modified by several alkali metal ions and a process for preparing the same.

The processes of the prior art includes the use of metal nitrates and/or metal hydroxides for exchanging the metal present in the zeolite and imparting acidic/basic property to the zeolite.

The drawbacks associated with the processes involving the use of metal nitrates is that nitrate ions generated during the reaction cause corrosion of the equipment being used. Further, the nitrate ions impart high acidity to the zeolite which requires a large quantity of water to remove acidity and balance acidic-basic property of the zeolite. Washing the zeolite with a large quantity of water has a disadvantage of leaching alkali metal cation. Due to this concurrent existence of drawbacks the required balance of acidic and basic properties in the resulting zeolite cannot be achieved. The processes involving the use of metal hydroxides for preparing zeolites with balanced acidic and basic properties also suffer from the drawbacks similar to the process involving the use of alkali metal nitrates.

The balance of acidic and basic properties of zeolites of the prior arts is still unsatisfactory. Therefore, there is a felt need for zeolites having optimized acidic and basic properties. There is also need for a simple and efficient process for preparing zeolites having optimized acidic and basic properties.

OBJECTS:

Some of the objects of the present disclosure, which at least one embodiment herein satisfies, are as follows:

It is an object of the present disclosure to ameliorate one or more problems of the prior art or to at least provide a useful alternative.

It is another object of the present disclosure to provide a modified zeolite.

It is still another object of the present disclosure to provide a modified zeolite which possesses optimized surface acidic and basic properties.

It is still another object of the present disclosure to provide a simple and efficient process for preparing a modified zeolite.

It is yet another object of the present disclosure to provide a process for preparing a modified zeolite which possesses optimized surface acidic and basic properties.

It is further object of the present disclosure to provide a process for preparing a modified zeolite which requires optimum quantity of water.

It is still further object of the present disclosure to provide a process for preparing a modified zeolite using the metal salts which produce less corrosive anions.

Other objects and advantages of the present disclosure will be more apparent from the following description which is not intended to limit the scope of the present disclosure.

SUMMARY:

In accordance with one aspect of the present disclosure there is provided a modified faujasite zeolite comprising at least one zeolite selected from the group consisting of sodium form of faujasite zeolite X and sodium form of faujasite zeolite Y exchanged with at least two alkali metals selected from the group consisting of lithium (Li), potassium (K), rubidium (Rb), cesium (Cs) and francium (Fr),

> wherein,
>
> the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:15,
>
> the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%
>
> the particle size of said modified faujasite zeolite ranges between 20 and 200 micron, and
>
> the sum total of at least two alkali metals in said modified faujasite zeolite is in the range of 0.1 to 97 wt%

Typically, the modified faujasite zeolite is in the form selected from the group consisting of pellets, extradites, extrudates, spheres, granules and tablets, wherein the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm.

Typically, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2

The modified faujasite zeolite of the present disclosure further comprises at least one promoter selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium (Zr), niobium (Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In) in an amount ranging between 0.01 and 15 wt%.

Typically, the pore diameter of the modified faujasite zeolite ranges between 15 and 40 angstrom.

Typically, the surface area of the modified faujasite zeolite ranges between 200 and 600 m^2/g.

Typically, the weight ratio of first alkali metal to the second alkali metal ranges between 1:2.5 and 2.5:1.

In one of the preferred embodiments of the present disclosure there is provided a modified faujasite zeolite, wherein the zeolite is the sodium form of faujasite zeolite X and the alkali metals are potassium (K) in an amount ranging between 0.1 and 25 wt%, rubidium (Rb) in an amount ranging between 0.1 and 40 wt% and cesium (Cs) in an amount ranging between 0.1 and 37 wt%,

> wherein, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2; and the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%

In accordance with another preferred embodiment of the present disclosure there is provided a modified faujasite zeolite, wherein the zeolite is the sodium form of faujasite zeolite X and the alkali metals are rubidium (Rb) in an amount ranging between 0.5 and 50 wt% and cesium (Cs) in an amount ranging between 0.2 and 40 wt%,

> wherein, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2; and the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%.

In accordance with another aspect of the present disclosure there is provided a process for preparing a modified faujasite zeolite, said process comprising the following steps:

i. treating at least one faujasite zeolite, at least once, with at least one first alkali metal acetate in the presence of a liquid medium to obtain an alkali metal exchanged faujasite zeolite,
ii. retreating said alkali metal exchanged faujasite zeolite, at least once, with at least one second alkali metal acetate in the presence of a liquid medium to obtain slurry containing a modified faujasite zeolite,
iii. separating said slurry to obtain a residue containing the modified faujasite zeolite, and
iv. washing said residue by using deionized distilled water followed by drying and calcining to obtain the modified faujasite zeolite,

wherein, the particle size of said modified faujasite zeolite ranges between 20 and 200 micron; the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%; the sum total of at least two alkali metals in said modified faujasite zeolite is in the range of 0.1 wt% and 97 wt%; and the ratio of said first alkali metal to the second alkali metal ranges between 1:2.5 and 2.5:1.

Typically, the modified faujasite zeolite obtained by the process of the present disclosure is in the form selected from the group consisting of pellets, extradites, extrudates, spheres, granules and tablets, wherein the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm.

Typically, the modified faujasite zeolite obtained by the process of the present disclosure is characterized by the pore diameter in the range of 15 and 40 angstrom and the surface area in the range of 200 and 600 m^2/g.

Typically, the process of the present disclosure further comprises a method step of adding at least one promoter in an amount ranging between 0.01 and 15 wt%.

Typically, the promoter is selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium Zr, niobium (Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In).

Typically, the method steps of treating and retreating are carried out at a temperature ranging between 70 and 130 °C for a time period ranging between 2 and 10 hours.

Typically, the faujasite zeolite is selected from the group consisting of sodium form of faujasite zeolite X and sodium form of faujasite zeolite Y.

Typically, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:15.

Typically, the first alkali metal acetate and the second alkali metal acetate are selected from the group consisting of lithium acetate (CH3COOLi), potassium acetate (CH3COOK), rubidium acetate (CH3COORb), cesium acetate (CH3COOCs) and francium acetate (CH3COOFr).

Typically, the liquid medium is selected as deionized distilled water.

Typically, the method step of drying is carried out at a temperature ranging between 100 °C and 140 °C for a time period in the range of 8 to 20 hours.

Typically, the method step of calcination is carried out at a temperature ranging between 450 °C and 600 °C for a time period in the range of 2 and 8 hours in the presence of at least one gas selected from the group consisting of air and nitrogen.

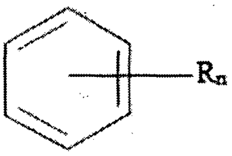
In accordance with still another aspect of the present disclosure there is provided a process for alkylating at least one alkyl group present on an aromatic compound; said process comprising the following steps:

i. mixing an aromatic compound comprising at least one alkyl group, alkanol and passing through a vaporizer at a temperature range of 125 to 250 °C to produce the vapors of reaction mixture, and
ii. heating of the vapors of the reaction mixture over the modified faujasite zeolite of the present disclosure in a catalytic amount at a temperature in the range of 250 to 600 °C and at a pressure ranging between 1 atm and 5 atm to obtain an alkylated alkyl aromatic compound,

wherein, the ratio of said aromatic compound comprising at least one alkyl group to alkanol ranges between 1:10 and 10:1; and the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%.

Typically, the process of the present disclosure comprises a pre-step of activating the modified faujasite zeolite by heating at a temperature of 400 °C and 600 °C for a time period of 0.5 and 6 hours under inert conditions.

Typically, the aromatic compound comprising at least one alkyl group is represented by a Formula I,



Formula I

wherein, R is selected from the group consisting of C1 to C5 alkyl group, and n is an integer ranging between 1 and 6.

Typically, the alkanol is represented by a Formula II,

R'-OH

Formula II

wherein, R' is selected from the group consisting of C1 to C10 straight chain or branched or cyclic alkyl group.

Preferably, the aromatic compound comprising at least one alkyl group is toluene, the alkanol is methanol and the alkylated alkyl aromatic compound is styrene.

DETAILED DESCRIPTION:

The inventors of the present disclosure developed a novel modified faujasite zeolite. The modified faujasite zeolite comprises at least one zeolite exchanged with at least two alkali metals. The zeolite is selected from the group consisting of sodium form of faujasite zeolite X and sodium form of faujasite zeolite Y whereas the alkali metal is selected from the group consisting of lithium (Li), potassium (K), rubidium (Rb), cesium (Cs), and francium (Fr). The sodium metal present in the sodium form of faujasite zeolites is at least partially exchanged with the alkali metals. The amount of sodium retained in the zeolite plays an important role in maintaining the balance between acidic and basic properties of the modified zeolite. The inventors after several experiments optimized the amount of sodium in the modified zeolite in the range of 3 to 10 wt% to obtain desired balance of acidic and basic properties. The modified zeolite of the present invention is characterized by silica to alumina molar ratio in the range of 1:1 to 1:15 and particle size in the range of 20 to 200 micron. Typically, the silica to alumina molar ratio ranges between 1:1 and 1:1.2.

The modified faujasite zeolite of the present disclosure is in the form selected from the group consisting of pellets, extradites, extrudates, spheres, granules and tablets and the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm.

The properties of the modified zeolite depend on the extent to which the balance between surface acidity and basicity is achieved. The balance between surface acidity and basicity is achieved by imparting at least two alkali metals to the sodium form of zeolite X or Y in an amount ranging between 0.1 % and 97 wt%. Further, as the properties of the modified faujasite zeolite of the present disclosure depend largely on the balance of acidic and basic properties which is the result of exchange of at least two alkali metals, it will be evident to express the molar ratio of the first alkali metal to the second alkali metal. Accordingly, the weight ratio of the first alkali to the second alkali metal ranges between 1:2.5 and 2.5:1.

Further balance of surface acidity and basicity may be imparted to the modified faujasite zeolite of the present disclosure by using at least one promoter selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium (Zr), niobium (Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In) in an amount ranging between 0.01 and 15 wt%.

The performance of the modified faujasite zeolite also depends on the pore diameter and the surface area of the modified faujasite zeolite. To obtain achieve optimum performance of the modified faujasite zeolite the pore diameter of the modified faujasite zeolite ranges between 15 and 40 angstrom and the surface area of the modified faujasite zeolite ranges between 200 and 600 m^2/g.

The modified faujasite zeolite that possesses improved properties comprises sodium form of faujasite zeolite X exchanged with potassium in an amount ranging between 0.1 and 25 wt%, rubidium in an amount ranging between 0.1 and 40 wt%, and cesium in an amount ranging between 0.1 and 37 wt%. The amount of sodium present in the zeolite exchanged with potassium, rubidium and cesium ranges between 3 and 10 wt% and the molar ratio of silica to alumina ranges between 1:1 and 1:1.2.

Another, modified faujasite zeolite that possesses improved properties comprises sodium form of faujasite zeolite X exchanged with rubidium in an amount ranging between 0.5 and 50 wt%, and cesium in an amount ranging between 0.2 and 40 wt%. The amount of sodium present in the zeolite exchanged with rubidium and cesium ranges between 3 and 10 wt% and the molar ratio of silica to alumina ranges between 1:1 and 1:1.2.

In accordance with another aspect of the present disclosure there is provided a process for preparing the modified faujasite zeolite. In the first step, sodium form of zeolite X, sodium form of zeolite Y or a combination thereof is treated with at least one; first alkali metal acetate in the presence of a liquid medium to obtain an alkali metal exchanged faujasite zeolite. The zeolite may be treated with the first alkali metal acetate only once or iteratively till the required degree of alkali metal is exchanged for sodium metal.

In the second step, the alkali metal exchanged faujasite zeolite obtained after first treatment is then again treated with at least one second alkali metal acetate in the presence of a liquid medium to obtain slurry containing a modified faujasite zeolite. The alkali metal exchanged faujasite zeolite is also treated with the second alkali metal acetate only once or iteratively till the required exchange of alkali metal is attained.

The liquid medium used in both the steps is deionized distilled water.

The alkali metal acetate used in the first and the second step obviates the shortcomings related to the use of alkali metal hydroxides and nitrates. The alkali metal acetate for the purpose of the present disclosure is selected from the group consisting of lithium acetate (CH3COOLi), potassium acetate (CH3COOK), rubidium acetate (CH3COORb), cesium acetate (CH3COOCs) and francium acetate (CH3COOFr).

The first and the second step are independently carried out a temperature ranging between 70 and 130 °C for a time period ranging between 2 and 10 hours.

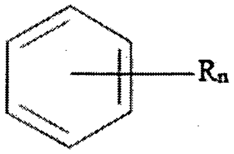
Optionally, the promoter selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium Zr, niobium (Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In) may be added before, during or after the first step and the second step. The amount in which promoter is added ranges between 0.01 and 15 wt%.

The slurry containing the modified faujasite zeolite obtained in the second step is separated in the third step to obtain a residue containing the modified faujasite zeolite. The separation may be executed by using any technique known to a person skilled in the art. Usually the residue containing the modified faujasite zeolite is obtained by filtration.

Finally, the residue containing the modified faujasite zeolite is washed with deionized distilled water and dried at a temperature ranging between 100 and 140 °C for a time period ranging between 8 and 20 hours. The dried modified faujasite zeolite is then subjected to calcination to obtain the modified faujasite zeolite. The calcination is carried out at a temperature ranging between 450 and 600 °C for a time period ranging between 2 and 8 hours in the presence of air or nitrogen or combination thereof.

The modified faujasite zeolite obtained according to the process of the present disclosure is characterized by the molar ratio of silica to alumina in the range of 1:1 to 1:15, preferably 1:1 and 1:1.2. The modified faujasite zeolite is also characterized by the presence of 3 and 10 wt% of sodium and the particle size of 20 and 200 micron. The sum total of at least two alkali metals in the modified faujasite zeolite obtained by the process of the present disclosure ranges between 0.1 wt% and 97 wt% and the ratio of first alkali metal to the second alkali metal ranges between 1:2.5 and 2.5:1. Further, the modified faujasite zeolite of the present disclosure is in the form selected from the group consisting of pellets extradites, extrudates, spheres, granules and tablets and the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm. Still further, the modified faujasite zeolite obtained by the process of the present disclosure has a pore diameter in the range of 15 to 40 angstrom and the surface area in the range of 200 and 600 m^2/g.

In accordance with the third aspect of the present disclosure there is provided a process for alkylating at least one alky group present on an aromatic compound represented by formula I using the modified faujasite zeolite as a catalyst and alkanol represented by formula II as a reactant.



Formula I

wherein, R is selected from the group consisting of C1 to C5 alkyl group, and

n is an integer ranging between 1 and 6.

R'-OH

Formula II

wherein, R' is selected from the group consisting of C1 to C10 straight chain or branched or cyclic alkyl group.

Initially, the mixture of an aromatic compound of formula I and alkanol of formula II is fed to the vaporizer at a temperature of 125 to 250 °C to produce the vapors of a reaction mixture. Then the vapors of the reaction mixture are brought in the contact with the modified faujasite zeolite of the present disclosure in a catalytic amount at a temperature in the range of 250 to 600 °C and at a pressure ranging between 1 atm and 5 atm to obtain an alkylated alkyl aromatic compound.

The progress of the reaction, selectivity for the desired product and yield of the product depends on the ratio of the reactants. Accordingly, the ratio of the aromatic compound of formula I to the alkanol of formula II ranges between 1:10 and 10:1.

The modified faujasite zeolite before mixing with aromatic compound of formula I and alkanol of formula II may be optionally, activated by heating at a temperature of 400 and 600 °C for a time period of 0.5 and 6.0 hours under inert conditions. Inert conditions during activation of the modified faujasite zeolite may be maintained by using inert gas which includes but is not limited to nitrogen.

Particularly, the modified faujasite zeolite of the present disclosure is used in a catalytic amount to selectively prepare styrene by reacting toluene and methanol. In one embodiment the process involving the use of the modified faujasite zeolite of the present disclosure exhibits high methanol conversion with selectivity for styrene in the range of 60 to 82%.

The present disclosure is further described in light of the following non-limiting examples which are set forth for illustration purpose only and are not to be construed for limiting the scope of the disclosure.

Examples

Example 1: Preparation of Cs-Rb-Na-X zeolite devoid of promoter

Synthesis of Cs-Rb-Na-X catalyst was carried out by three times ion-exchange of Na-X (Si/Al = 1.2) with 0.1 molar rubidium acetate solution in deionized distilled water (solid to liquid ratio = 20 g/L) and one time cesium acetate salt solution (solid to liquid ratio = 20 g/L) at 90-100 °C for 6 h to obtain slurry containing Cs-Rb-Na-X zeolite. The slurry was filtered and washed with warm deionized distilled water. The filter cake was dried in an oven at 120 °C for 16 h and calcined in a temperature programmed furnace at 540 °C for 6 h in air.

Example 2: Preparation of Cs-Rb-Na-X zeolite comprising boron (promoter)

The 3.96 gm of Cs-Rb-Na-X zeolite prepared as described in example 1 was mixed with 50 mL solution containing 0.071 g of boric acid in de-ionized distilled water under continuous stirring to obtain Cs-Rb-Na-X zeolite comprising 1 wt% boron oxide. The promoter loaded catalyst was dried at 120 °C for 16 h followed by calcination at 540 °C for 6 h.

Example 3: Preparation of Cs-Rb-Na-X zeolite comprising lanthanum (promoter)

The 3.96 gm of Cs-Rb-Na-X zeolite prepared as described in example 1 was mixed with 50 mL solution containing 0.0776 g of lanthanum acetate salt in de-ionized distilled water under continuous stirring to obtain Cs-Rb-Na-X zeolite comprising 1 wt% lanthanum oxide. The promoter loaded catalyst was dried at 120 °C for 16 h followed by calcination at 540 °C for 6 h.

Example 4: Preparation of Rb-Cs-Na-X zeolite devoid of promoter

Synthesis of Rb-Cs-Na-X catalyst was carried out by three times ion-exchange of Na-X (Si/Al = 1.2) with 0.1 molar cesium acetate solution in deionized distilled water (solid to liquid ratio = 20 g/L) and one time rubidium acetate salt solution (solid to liquid ratio = 20 g/L) at 90-100 °C for 6 h to obtain slurry containing Rb-Cs-Na-X zeolite. The slurry was filtered and washed with warm deionized distilled water. The filter cake was dried in an oven at 120 °C for 16 h and calcined in a temperature programmed furnace at 540 °C for 6 h in air.

Example 5: Preparation of Rb-Cs-Na-X zeolite comprising boron (promoter)

The 3.96 gm of Rb-Cs-Na-X zeolite prepared as described in example 4 was mixed with 50 mL solution containing 0.071 g of boric acid in de-ionized distilled water under continuous stirring to obtain Rb-Cs-Na-X zeolite comprising 1 wt% boron oxide. The promoter loaded catalyst was dried at 120 °C for 16 h followed by calcination at 540 °C for 6 h.

Example 6: Preparation of Rb-Cs-K-Na-X zeolite devoid of promoter

20 gm of Na-X (Si/Al = 1.2) was exchanged four times with 1000 mL of 0.1 molar potassium acetate solution in deionized distilled water, three times with 1000 mL of cesium acetate solution (0.1 molar) in deionized distilled water and one time with 1000 mL of rubidium acetate salt solution (0.1 molar) in deionized distilled water at a temperature of 90-100 °C for 6 h to obtain slurry containing Rb-Cs-K-Na-X zeolite. The slurry was filtered and washed with warm deionized distilled water. The residue was dried in an oven at 120 °C for 16 h and calcined in a temperature programmed furnace at 540 °C for 6 h in the presence of air to obtain Rb-Cs-K-Na-X zeolite.

Example 7: Preparation of Rb-Cs-K-Na-X zeolite comprising boron (promoter)

The 3.96 gm of Rb-Cs-K-Na-X zeolite prepared as described in example 6 was mixed with 50 mL solution containing 0.071 g of boric acid in de-ionized distilled water under continuous stirring to obtain Rb-Cs-K-Na-X zeolite comprising 1 wt% boron oxide. The promoter loaded catalyst was dried at 120 °C for 16 h followed by calcination at 540 °C for 6 h.

Example 8: Preparation of Rb-K-Na-X zeolite

The Rb-K-Na-X catalyst is prepared by four times ion-exchange of Na-X (Si/Al = 1.2) with 0.1 molar potassium acetate solution in deionized distilled water (solid to liquid ratio = 50 g/L) and one time rubidium acetate salt solution (solid to liquid ratio = 20 g/L) at 95-100 °C for 6 h. The resulting slurry was filtered and washed with warm deionized distilled water. The filter cake was dried in an oven at 120 °C for 16 h and calcined in a temperature programmed furnace at 540 °C for 6 h in air.

Examples 9-16: Synthesis of styrene using zeolites of examples 1 to 8

Zeolites prepared in examples 1 to 8 were used for the synthesis of styrene. The general procedure is as follows:-

Side chain alkylation of toluene with methanol were carried out in lab scale down-flow quartz reactor (OD = 32 mm; length = 16 inches). The crushed pellets of catalyst (1.0 g; 0.8-1.18 mm and 1.2-1.5mm particles size) were loaded in the reactor and activated at 500 °C for 2 h in nitrogen flowrate (40 mL/min). Then the reactor was cooled down to the reaction temperature (425 °C) under the nitrogen flow. The reaction mixture at desired methanol to toluene molar ratio was charged to the reactor via vaporizer (vaporizer temperature = 200 °C) at desired flowrate using syringe pump. After the reaction, vapors of the reaction mixture were cooled down in the condenser using chilled water flowing at 10 °C, followed by gas-liquid separation. The liquid samples were analyzed by gas chromatography (Shimadzu 17A, Japan) using innowax capillary column (60 m length and 0.32 mm diameter, 0.25 micrometer thickness) and a flame ionization detector (FID). The results are provided herein Table 1.

Comparative example 1-4: Synthesis of styrene using zeolites exchanged with one alkali metal

Side chain alkylation of toluene with methanol were carried out in lab scale down-flow quartz reactor (OD = 32 mm; length = 16 inches). The crushed pellets of catalyst (1.0 g; 0.8-1.18 mm and 1.2-1.5mm particles size) was loaded in the reactor and activated at 500 °C for 2 h in nitrogen flowrate (40 mL/min). Then the reactor was cooled down to the reaction temperature (425 °C) under the nitrogen flow. The reaction mixture at desired methanol to toluene molar ratio was charged to the reactor via vaporizer (vaporizer temperature = 200 °C) at desired flowrate using syringe pump. After the reaction, vapors of the reaction mixture were cooled down in the condenser using chilled water flowing at 10 °C, followed by gas-liquid separation. The liquid samples were analyzed by gas chromatography (Shimadzu 17A, Japan) using innowax capillary column (60 m length and 0.32 mm diameter, 0.25 micrometer thickness) and a flame ionization detector (FID). The results are provided herein Table 1.

Table 1:

Particulars	**Toluene to methanol ratio**	**% Methanol conversion**	**% Selectivity**		
			Styrene	**Ethylbenzene**	**Xylenes**
Example 9 (Cs-Rb-Na-X)	5:1	87	79	16	2
Example 10 (B-Cs-Rb-Na-X)	5:1	89	82	10	5
Example 11 (La-Cs-Rb-Na-X)	5:1	95	62	37	0
Example 12 (Rb-Cs-Na-X)	5:1	90	67	22	3
Example 13 (B-Rb-Cs-Na-X)	5:1	89	68	19	7
Example 14 (Rb-Cs-K-Na-X)	5:1	90	74	25	0
Example 15 (B-Rb-Cs-K-Na-X)	5:1	89	78	13	7
Example 16 (Rb-K-Na-X)	5:1	87	71	19	5
Comparative example 1 (Li-Na-X)	5:1	96	3	4	60
Comparative example 2 (K-Na-X)	5:1	92	60	30	6
Comparative example 3 (Rb-Na-X)	5:1	89	68	24	3
Comparative example 4 (Cs-Na-X)	5:1	91	70	25	2

From the above results, it is observed that the modified faujasite zeolite of the present disclosure i.e., a zeolite exchanged with at least two alkali metals exhibits enhanced selectivity for styrene.

Throughout this specification the word "comprise" or variations such as "comprises" or "comprising", will be understood to imply the inclusion of a stated element, integer or step, or group of elements, integers or steps, but not the exclusion of any other element, integer or step, or group of elements, integers or steps.

The use of the expression "at least" or "at least one" suggests the use of one or more elements or ingredients or quantities, as the use may be in the embodiment of the invention to achieve one or more of the desired objects or results.

Any discussion of documents, acts, materials, devices, articles or the like that has been included in this specification is solely for the purpose of providing a context for the invention. It is not to be taken as an admission that any or all of these matters form part of the prior art base or were common general knowledge in the field relevant to the invention as it existed anywhere before the priority date of this application.

The numerical values mentioned for the various physical parameters, dimensions or quantities are only approximations and it is envisaged that the values higher/lower than the numerical values assigned to the parameters, dimensions or quantities fall within the scope of the disclosure, unless there is a statement in the specification specific to the contrary.

While considerable emphasis has been placed herein on the product and the process, it will be appreciated that alterations can be made and that many modifications can be made in the preferred embodiment departing from the principles of the disclosure. These and other changes in the preferred product and the process of the disclosure will be apparent to those skilled in the art from the disclosure herein, whereby It is to be distinctly understood that the foregoing descriptive matter is to be interpreted merely as illustrative of the disclosure and not as a limitation.

Claims:

1. A modified faujasite zeolite comprising at least one zeolite selected from the group consisting of sodium form of faujasite zeolite X and sodium form of faujasite zeolite Y exchanged with at least two alkali metals selected from the group consisting of lithium (Li), potassium (K), rubidium (Rb), cesium (Cs) and francium (Fr),

   wherein,

   the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:15,

   the amount of sodium in the modified zeolite is in the range of 3 to 10 wt%,

   the particle size of said modified faujasite zeolite ranges between 20 and 200 micron, and

   the sum total of at least two alkali metals in said modified faujasite zeolite is in the range of 0.1 to 97 wt%.
2. The modified faujasite zeolite as claimed in claim 1, wherein the modified zeolite is in the form selected from the group consisting of pellets, extradites, extrudates, spheres, granules and tablets, wherein the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm.
3. The modified faujasite zeolite as claimed in claim 1, wherein the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2.
4. The modified faujasite zeolite as claimed in claim 1, further comprises at least one promoter selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium (Zr), niobium (Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In) in an amount ranging between 0.01 and 15 wt%.

5. The modified faujasite zeolite as claimed in claim 1, wherein the modified faujasite zeolite is characterized by the pore diameter in the range of 15 and 40 angstrom and the surface area in the range of 200 and 600 m^2/g.
6. The modified faujasite zeolite as claimed in claim 1, wherein the weight ratio of the first alkali metal to the second alkali metal ranges between 1:2.5 and 2.5:1.
7. The modified faujasite zeolite as claimed in claim 1, wherein the zeolite is the sodium form of faujasite zeolite X and the alkali metals are potassium (K) in an amount ranging between 0.1 and 25 wt%, rubidium (Rb) in an amount ranging between 0.1 and 40 wt% and cesium (Cs) in an amount ranging between 0.1 and 37 wt%,

   wherein, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2; and the amount of sodium in the modified zeolite catalyst is in the range of 3 to 10 wt%.
8. The modified faujasite zeolite as claimed in claim 1, wherein the zeolite is the sodium form of faujasite zeolite X and the alkali metals are rubidium (Rb) in an amount ranging between 0.5 and 50 wt% and cesium (Cs) in an amount ranging between 0.2 and 40 wt%,

   wherein, the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:1.2; and the amount of sodium in the modified zeolite catalyst is in the range of 3 to 10 wt%.
9. A process for preparing a modified faujasite zeolite, said process comprising the following steps:
   i. treating at least one faujasite zeolite, at least once, with at least one first alkali metal acetate in the presence of a liquid medium to obtain an alkali metal exchanged faujasite zeolite,

ii. retreating said alkali metal exchanged faujasite zeolite, at least once, with at least one second alkali metal acetate in the presence of a liquid medium to obtain slurry containing a modified faujasite zeolite,

iii. separating said slurry to obtain a residue containing the modified faujasite zeolite, and

iv. washing said residue by using deionized distilled water followed by drying and calcining to obtain the modified faujasite zeolite,

wherein, the particle size of said modified faujasite zeolite ranges between 20 and 200 micron; the amount of sodium in the modified zeolite catalyst is in the range of 3 to 10 wt%; the sum total of at least two alkali metals in said modified faujasite zeolite is in the range of 0.1 wt% and 97 wt%; and the ratio of said first alkali metal to the second alkali metal ranges between 1:2.5 and 2.5:1.

10. The process as claimed in claim 9, wherein the modified zeolite is in the form selected from the group consisting of pellets extradites, extrudates, spheres, granules and tablets, wherein the particle size of the modified faujasite zeolite in said form ranges between 0.05 and 10 mm.

11. The process as claimed in claim 9, wherein the modified faujasite zeolite is characterized by the pore diameter in the range of 15 and 40 angstrom and the surface area in the range of 200 and 600 m^2/g.

12. The process as claimed in claim 9, further comprises a method step of adding at least one promoter in an amount ranging between 0.01 and 15 wt%.

13. The process as claimed in claim 12, wherein the promoter is selected from the group of cations consisting of magnesium (Mg), calcium (Ca), strontium (Sr), barium (Ba), lanthanum (La), boron (B), scandium (Sc), ytterbium (Y), zirconium Zr, niobium

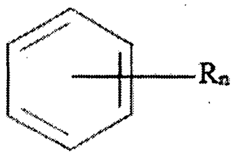
(Nb), cerium (Ce), neodymium (Nd), copper (Cu), zinc (Zn), iron (Fe), nickel (Ni), gallium (Ga) and indium (In).

14. The process as claimed in claim 9, wherein the method steps of treating and retreating are carried out at a temperature ranging between 70 and 130 °C for a time period ranging between 2 and 10 hours.
15. The process as claimed in claim 9, wherein the faujasite zeolite is selected from the group consisting of sodium form of faujasite zeolite X and sodium form of faujasite zeolite Y.

16. The process as claimed in claim 9, wherein the molar ratio of silica to alumina present in the modified faujasite zeolite ranges between 1:1 and 1:15.
17. The process as claimed in claim 9, wherein the first alkali metal acetate and the second alkali metal acetate are selected from the group consisting of lithium acetate (CH3COOLi), potassium acetate (CH3COOK), rubidium acetate (CH3COORb), cesium acetate (CH3COOCs) and francium acetate (CH3COOFr).
18. The process as claimed in claim 9, wherein the liquid medium is deionized distilled water.
19. The process as claimed in claim 9, wherein the method step of drying is carried out at a temperature ranging between 100 °C and 140 °C for a time period in the range of 8 to 20 hours.
20. The process as claimed in claim 9, wherein the method step of calcination is carried out at a temperature ranging between 450 °C and 600 °C for a time period of 2 and 8 hours in the presence of at least one gas selected from the group consisting of air and nitrogen.

21. A process for alkylating at least one alkyl group present on an aromatic compound; said process comprising the following steps:
    i. mixing an aromatic compound comprising at least one alkyl group, alkanol and passing through a vaporizer at a temperature range of 125 to 250 °C to produce the vapors of reaction mixture, and
    ii. heating of the vapors of the reaction mixture over the modified faujasite zeolite of claim 1 in a catalytic amount at a temperature in the range of 250 to 600 °C and at a pressure ranging between 1 atm and 5 atm to obtain an alkylated alkyl aromatic compound,

    wherein, the ratio of said aromatic compound comprising at least one alkyl group to alkanol ranges between 1:10 and 10:1; and the amount of sodium in the modified faujasite zeolite is in the range of 3 to 10 wt%.
22. The process as claimed in claim 21, comprises a pre-step of activating the modified faujasite zeolite by heating at a temperature of 400 °C and 600 °C for a time period of 0.5 and 6 hours under inert conditions.
23. The process as claimed in claim 21, wherein the aromatic compound comprising at least one alkyl group is represented by a Formula I,



Formula I

wherein, R is selected from the group consisting of C1 to C5 alkyl group, and n is an integer ranging between 1 and 6.

24. The process as claimed in claim 21, wherein the alkanol is represented by a Formula II,

$$R'\text{-}OH$$

Formula II

wherein, R' is selected from the group consisting of C1 to C10 straight chain or branched or cyclic alkyl group.

25. The process as claimed in claim 21, wherein the aromatic compound comprising at least one alkyl group is toluene, the alkanol is methanol and the alkylated alkyl aromatic compound is styrene.