ORIGINAL

Abstract

"Greener catalytic process for the synthesis of α-α' dibenzilidenecyclohexanone by low cost fly ash-Kota stone slurry catalyst".

This invention relates to novel route of synthesis of solid base catalyst from fly ash and Kota stone slurry waste and a single step synthetic process for α-α'- dibenzylidenecyclohexanone through condensation of benzaldehyde and cyclohexanone, the preparation of catalyst and the catalytic process comprises of, Mechanical activation of fly ash by ball milling; Thermal activation of fly ash by calcining at 900°C temperature; Mixing of Kota stone slurry in activated fly ash and refluxing the paste for 24h at 110°C under constant stirring; Separation of solid mixture from refluxing unit and drying at 110°C for 24h followed by calcinations at temperature 550°C for 2h under static conditions; Reacting benzaldehyde with cyclohexanone in presence of low cost KFC catalyst in solvent free medium, maintaining the substrate to catalyst weight ratio of 10; Maintaining the temperature of the reaction in the range of 90-120°C at atmospheric pressure for a period selected from 30-120 minutes; Separating the solid catalyst from the reaction mixture by filtration method; Washing the catalyst with acetone to remove adhering materials; Drying the catalyst at 110°C for 24h followed by calcinations at 550°C for a period of 2h under static conditions.

(MONA SAINI)
OF L.S.DAVAR & CO.,
APPLICANT'S AGENT

We Claim

1. A novel route of synthesis of solid base catalyst from fly ash and Kota stone slurry waste and a single step synthetic process for α-α' dibenzylidenecyclohexanone through condensation of benzaldehyde and cyclohexanone. The preparation of catalyst and the catalytic process comprises of:-

   i. Mechanical activation of fly ash by ball milling;
   ii. Thermal activation of fly ash by calcining at 900°C temperature;
   iii. Mixing of Kota stone slurry in activated fly ash and refluxing the paste for 24h at 110°C under constant stirring;
   iv. Separation of solid mixture from refluxing unit and drying at 110°C for 24h followed by calcinations at temperature 550°C for 2h under static conditions;
   v. Reacting benzaldehyde with cyclohexanone in presence of low cost KFC catalyst in solvent free medium, maintaining the substrate to catalyst weight ratio 10;
   vi. Maintaining the temperature of the reaction in the range of 90-120°C at atmospheric pressure for a period selected from 30-120 minutes;

vii. Separating the solid catalyst from the reaction mixture by simple filtration method;

viii. Washing the catalyst with acetone to remove adhering materials;

ix. Drying the catalyst at 110°C for 24h followed by calcinations at 550°C for a period of 2h under static conditions.

2. A catalytic process as claimed in claim 1, wherein the condensation of benzaldehyde with cyclohexanone is carried out in a single step without the use of any solvent.

3. A catalytic process as claimed in claim 1, wherein the fly ash - Kota stone slurry (KFC) catalyst having the crystallite size of 27nm.

4. A catalytic process as claimed in claim 1, wherein the condensation of benzaldehyde with cyclohexanone is effected with more than 82% conversion and 92% selectivity of desired product α-α'-dibenzylidenecyclohexanone.

5. A catalytic process as claimed in claim 1, wherein the conversion for the product is maintained between 52 to 82% and high selectivity 92%.

6. A catalytic process as claimed in claims 1-5, wherein the condensation is carried out at temperature maintained between 90-120°C at atmospheric pressure without generating any hazardous byproduct.

7. A catalytic process as claimed in claims 1-6 wherein the catalyst can be easily separated from the reaction mixture and regenerated by calcinations at 550°C for 2h was reused for three reaction cycles.

Dated this 2nd day of June, 2010

(MONA SAINI)
OF L.S.DAVAR & CO.,
APPLICANT'S AGENT.

ORIGINAL
129 JUN 10
JUN 2010

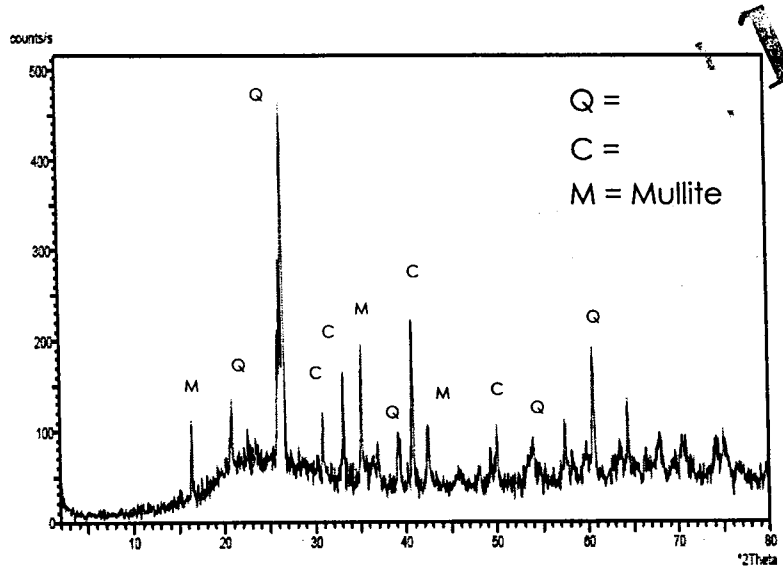

Figure 1. XRD of FA

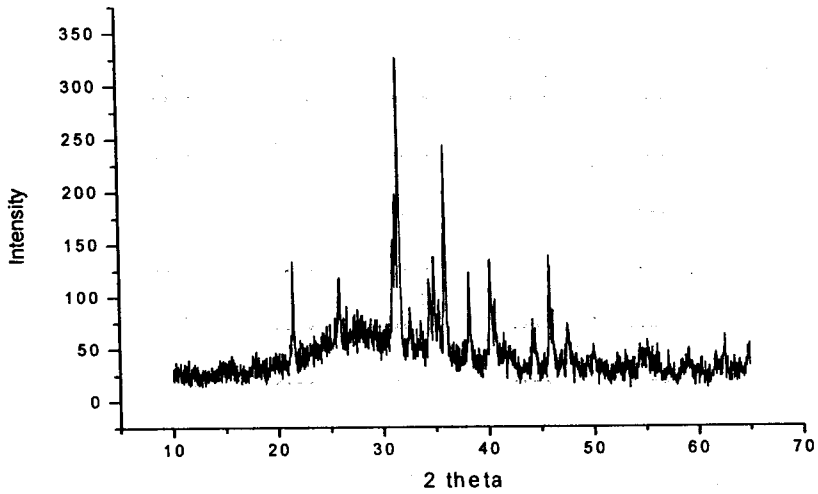

Figure 2. XRD of KFC catalyst

MONA SAINI
of L.S. Davar & Co

ORIGINAL
129 JUN 10
2010

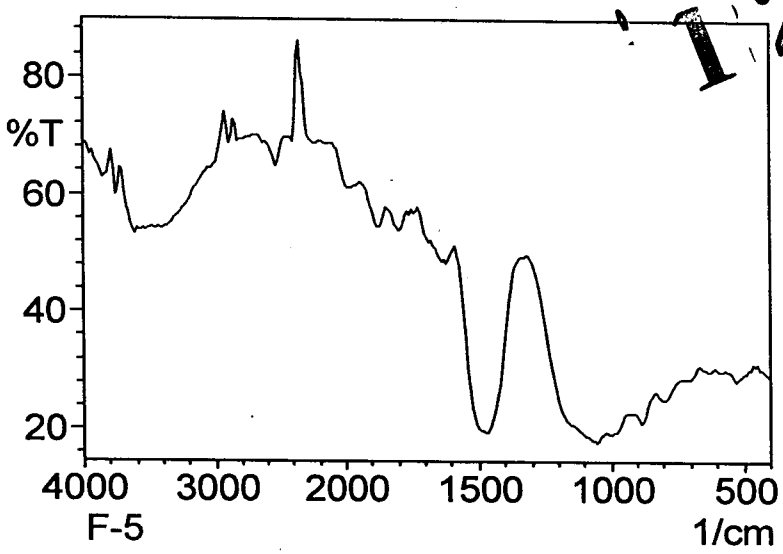

Figure 3. FT-IR of KFC catalyst

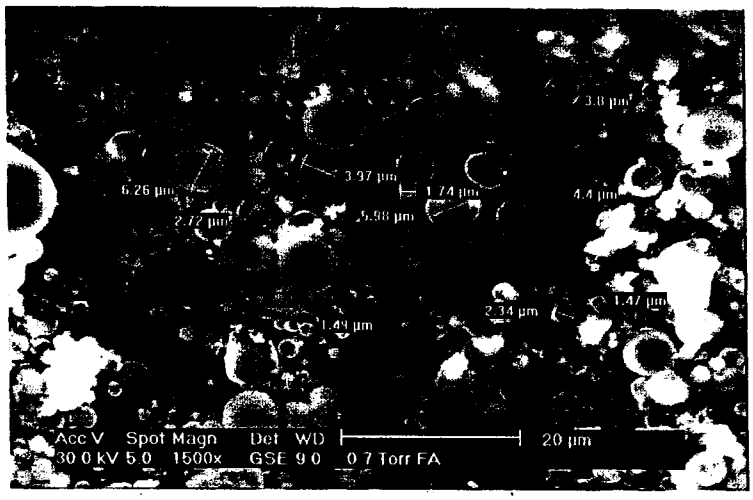

Figure 4. SEM of fly ash

MSaini
MONA SAINI
of L.S. Davar & Co.

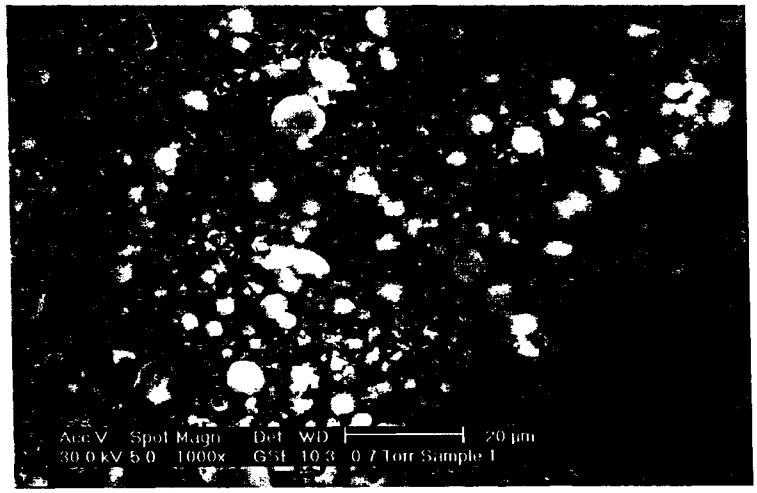

Figure 5. SEM of KFC catalyst

MSaini
MONA SAINI
of L.S. Davar & Co

"Greener catalytic process for the synthesis of α-α' dibenzilidenecyclohexanone by low cost fly ash-Kota stone slurry catalyst".

Field of Invention

This invention relates to Greener catalytic process for the synthesis of α-α' dibenzilidenecyclohexanone by low cost fly ash-Kota stone slurry catalyst.

Background of Invention

The product α-α'-dibenzylidenecyclohexanone obtained from condensation reaction of benzaldehyde and cyclohexanone is used as agrochemical and pharmaceuticals or as important intermediate in the production of agrochemical and pharmaceuticals products. As mentioned in **U.S. patent no. 6,632,959 B2** which, describes that the use of homogeneous catalysts such as NaOH, alkali alkoxide etc. are associated with environmental problems as they release large amount of harmful effluents produced after water work up. To overcome the hazards of homogeneous bases, solid base catalysts have been introduced. **U.S. patent no. 5,254,743, Holmgren et al** reported the process of use of hydrotalcite as an efficient solid base catalyst for effective aldol condensation of aldehydes and ketones and **US patent no: 20090112007 A1** describes the catalytic use of solid catalyst for biodiesel production. MgO, calcined hydrotalcites, rehydrated hydrotalcites and grafted quaternary organic ammonium hydroxides on

MCM-41, have also been used as catalysts in Knoevenagel condensation, aldolization and Michael additions. **US patent no: 6794325 B1** gives a process in which solid base catalyst is employed for producing glycol from aldehyde. So far Kota stone slurry has been reported to be used as building and construction material but not as a solid catalyst. In the present research we tried to prepare solid base catalyst by mixing fly ash and Kota stone slurry which proves to be an efficient and recyclable catalyst for condensation reaction. Method is developed for loading of basic CaO, predominantly present in Kota stone over fly ash support so that sufficient basic sites are developed over activated fly ash to provide catalytic sites for desired condensation reaction. The work is novel and yet unprecedented in the literature.

Fly ash is an ensemble of compounds (silica, alumina, ferric oxide, calcium oxide and other metal oxide such as Mn_2O_3, TiO_2 with a single reactive amorphous phase (silico-aluminous glass) and inert crystallite phases such as mullite, quartz and magnetite, having catalytic activity for catalyzing different reactions such as oxidation, chlorination, Knoevenagel condensation, Schiff base formation, Biginelli and Hantzsch reactions. Kota stone slurry, a waste from mines of Kota stone industries consists of CaO, SiO_2, Al_2O_3 and oxide of Na and Mg, which has been widely used as construction and building material. With appropriate activation of fly ash and Kota stone slurry (a solid waste) an efficient and economic solid base catalyst is prepared with similar catalytic activity as other solid base catalysts derived from silica materials.

Objects of Invention

The main object of this invention is to develop a catalytic process for the synthesis of α-α' dilbenzilidenecyclohexanene.

Other object is to utilize coal generated fly ash and Kota stone slurry (both are solid wastes) as solid base catalyst in condensation reaction through greener route.

Another object of the present invention is to synthesize nano-crystalline solid base catalyst from raw fly ash of higher silica content generated from thermal power plant.

Another object of the present invention is to synthesize solid base catalyst by mechanical activation of fly ash by ball milling.

Another object of the present invention is to synthesize solid base catalyst by thermal activation of mechanically activated fly ash by calcination at 900°C temperature.

Another object of the present invention is to synthesize nano-crystalline solid base catalytic material by mixing Kota stone slurry and activated fly ash.

Another object of the present invention is to make solid base catalyst by adding Kota stone slurry in activated fly ash (mechanically and thermally) (1:1 ratio) by refluxing at 110°C for 24h followed by filtration and drying at 110°C for 24h and calcination at 550°C.

Another object of the present invention is to synthesize α-α'-dibenzylidenecyclohexanone product by condensation of benzaldehyde and cyclohexanone using nano-crystalline fly ash-Kota stone slurry catalyst (KFC).

Yet another object of the present invention is to perform condensation of benzaldehyde and cyclohexanone with KFC in single step, solvent free environment, at atmospheric pressure and optimized reaction conditions to get high conversion and selectivity.

Statement of Invention

This invention relates to a novel route of synthesis of solid base catalyst from fly ash and Kota stone slurry waste and a single step synthetic process for ∝-∝'- dibenzylidenecyclohexanone through condensation of benzaldehyde and cyclohexanone, the preparation of catalyst and the catalytic process comprises of, Mechanical activation of fly ash by ball milling; Thermal activation of fly ash by calcining at 900°C temperature; Mixing of Kota stone slurry in activated fly ash and refluxing the paste for 24h at 110°C under constant stirring; Separation of solid mixture from refluxing unit and drying at 110°C for 24h followed by calcinations at temperature 550°C for 2h under static conditions; Reacting benzaldehyde with cyclohexanone in presence of low cost KFC catalyst in solvent free medium, maintaining the substrate to catalyst weight ratio of 10; Maintaining the temperature of the reaction in the range of 90-120°C at atmospheric pressure for a period selected from 30-120 minutes; Separating the solid catalyst from the reaction mixture by

filtration method; Washing the catalyst with acetone to remove adhering materials; Drying the catalyst at 110°C for 24h followed by calcinations at 550°C for a period of 2h under static conditions.

Brief description of accompanying drawings

Fig 1. Shows XRD of Fly ash.

Fig 2. Shows XRD of fly ash based Kota stone catalyst

Fig 3. Shows FT-IR spectrum of fly ash based Kota stone catalyst.

Fig 4. Shows SEM of fly ash.

Fig 5. Shows SEM of fly ash-Kota stone catalyst.

Detailed description of Invention

The present invention relates to a method for preparing solid base catalyst by mixing fly ash and Kota stone slurry in a specific amount (1:1 ratio) at refluxing temperature (110°C for 24h) under atmospheric pressure, thus loading slurry minerals over fly ash support. The prepared solid base catalyst (KFC) was applied for condensation of benzaldehyde with cyclohexanone for the production of pharmaceutical intermediate α-α'-dibenzylidenecyclohexanone. The present invention also relates to the development of a solvent free, eco friendly catalytic

process for condensation reaction by replacing highly corrosive and hazardous conventional homogeneous bases viz NaOH, KOH, and KF etc. with easily separable, low cost, solid base catalyst prepared from fly ash and Kota stone slurry.

A highly active solid base catalyst with improved structural and textural properties with high catalytic performance was synthesized by mixing Kota stone slurry (solid waste from Kota stone polishing and cutting industries which comprises mainly CaO, SiO_2, and 30% water) and activated fly ash (solid waste from Kota Thermal Power Plant comprising mainly SiO_2, Al_2O_3, Fe_2O_3 and other alkali metal oxides in minor quantities). The mixing of Kota stone slurry and activated fly ash was done in 1:1 ratio and the resulting mixture was refluxed for 24h at 110°C on batch reactor under atmospheric pressure. The mixture was filtered, dried and calcined at 550°C for 2h before use as solid base catalyst. The physico-chemical properties of prepared catalyst (KFC) were analyzed by X-Ray diffraction, FT-IR spectroscopy, Scanning Electron Microscopy and Flame Atomic Absorption Spectrophotometer. The catalytic activity of (Kota stone slurry-fly ash catalyst) KFC was evaluated by Claisen-Schmidt condensation of benzaldehyde and cyclohexanone as model test reaction under optimized conditions. The catalyst gave an excellent yield (82%) of the desired product α-α'-dibenzylidenecyclohexanone at 120°C for 2h with molar ratio of reactants (benzaldehyde:cyclohexanone) 2:1, indicating that the catalyst is highly active, recyclable and possess higher number of basic sites on the surface which is responsible for the higher conversion. The product α-α'

dibenzylidenecyclohexanone is an important intermediates of agrochemical, pharmaceuticals and perfumes **[1]**.

Accordingly the present invention provides introduction of a new solid base catalyst KFC for condensation reaction .The invention includes following steps.

1) Mechanical activation of fly ash by ball milling.
2) Thermal activation of mechanically activated fly ash by calcination at higher temperature 900°C.
3) Activated fly ash was added in Kota stone slurry in 1:1 ratio and stirrable aqueous mixture was prepared by adding requisite water.
4) Refluxing the mixture at 110°C for 24h under continuous stirring.
5) The solid powder was separated from the refluxing flask by simple filteration and washed with water before drying at 110°C for 24h followed by calcination at 550°C for 2h under static conditions.
6) Performing the condensation of benzaldehyde and cyclohexanone in a solvent free medium over KFC, maintaining the substrate to catalyst weight ratio 10.
7) Maintaining the temperature of the reaction in the range of 90-120°C at atmospheric pressure for a period selected from 30-120min.
8) Separating the solid base catalyst KFC from the reaction mixture by filtration and analyzing the conversion of the filtered reaction mixture.
9) Washing the catalyst with acetone to remove the adhering materials.

10) Drying the catalyst at 110°C for 2h followed by calcinations at 550°C for 2h to reuse the catalyst.

In another embodiment of the present invention fly ash-Kota stone catalyst (KFC) having high basicity and basic sites on the surface may be used for catalytic conversion at atmospheric pressure.
In another embodiment of the invention the basicity of the catalyst was permanent and not subject to leaching from the catalyst, which is evident by high conversion data of α-α'-dibenzylidenecyclohexanone upto three reaction cycles.

The process of the present invention is based on the finding that the performance of fly ash solid base catalyst (KFC) is influenced by its preparation method i.e. loading of Kota stone slurry on fly ash by refluxing at ambient temperature and time, while refluxing; CaO is loaded over silica present in the mixture and generate surface active basic sites. The present invention also provides a new greener route for synthesis of an efficient, environmental benign and cost effective solid base catalyst from solid waste fly ash and Kota stone slurry.

Fly ash collected from Kota thermal power station was of class F type, with chemical composition as given in table-1. Kota stone slurry was collected as a waste from Kota stone cutting and polishing industries and contains water content up to 32% which was confirmed by LOI at 110°C. The detail of its chemical composition is given in table-1. The fly ash and Kota stone slurry employed in the preparation of catalyst were

characterized to determine their chemical composition, activity and reactivity. Prior to the loading the fly ash was activated by mechanical and thermal activation methods. Mechanical activation of fly ash increased its surface area with lower crystallite size, while thermal activation was done at 900°C which removed TiO_2, MgO, Na_2O, and K_2O, thus increasing silica content up to 62%. When aqueous Kota stone slurry containing lime and silica was added to fly ash, it produced an aqueous slurry of hydrous calcium silicate due to reaction between lime and high silica of fly ash. After calcination at required temperature the catalyst become thermally stable with high catalytic activity. The fly ash used was of class F type with chemical composition as given in table 1.

Table 1. Chemical composition of the fly ash and Kota stone slurry.

Sample	**SiO_2 (%wt)**	**Al_2O_3 (%wt)**	**Fe_2O_3 (%wt)**	**CaO (%wt)**	**MgO (%wt)**	**TiO_2 (%wt)**	**Na_2O (%wt)**	**K_2O (%wt)**	**Other elements (%wt)**	**L.O.I. (%wt)**
FA	58	19	8	0.6	0.6	1.3	3.74	2.18	3.0	3
KSS	24	3	-	40	1	-	2	-	-	30

The fly ash was mechanically activated by ball milling to increase surface area and then thermally activated at 900°C for removing volatile compounds (mainly C and S) and other impurities and to increase the

silica and alumina content required to making fly ash a good catalyst support. Kota stone slurry having silica, high calcium content and ~ 30% water was added to the activated fly ash and converted into stirrable slurry and the resulting mixture was refluxed at 110°C for 24h. The solid material was separated from the refluxing mixture by filtration and washed with distilled water and dried at 110°C for 24h followed by calcination at 550°C for 2h under static conditions.

The KFC catalyst was characterized by FT-IR spectroscopy, X-ray powder diffraction and scanning electron microscopy techniques. The crystallite size and crystallinity was determined from XRD pattern. The results showed that the crystallite size of KFC is 27 nm (figure 2) which is lower than the crystallite size (33nm) of activated fly ash (figure 1). This shows that the KFC is nano-crystalline in nature which provides more co-ordinatively unsaturated sites for reaction. The FT-IR spectra of KFC catalyst (figure 3) shows a small peak at around 991 cm^{-1} due to the presence of Si-O-Ca bond **[20]** of calcium silicate hydrate (C-S-H) formed due to reaction between calcium of kota stone slurry with the fly ash silica. This C-S-H is responsible for the increase in basicity of fly ash catalyst, as evident from the high conversion of base catalyzed Claisen Schmidt condensation reaction. SEM photograph of fly ash in Fig 4, shows the presence of hollow cenospheres, irregularly shaped unburned carbon particles, mineral aggregates and agglomerated particles. As shown in Fig. 5, typical SEM image of the KFC catalyst shows dense particles with distribution of varying particle size confirming the loading of Kota stone particles on fly ash support.

The catalytic activity of the catalyst was tested by using condensation of benzaldehyde and cyclohexanone which was carried out in liquid phase solvent free system in a 50 ml round bottom flask equipped with magnetic stirrer at atmospheric pressure. The optimization of the reaction parameters was also carried out in detail. In this procedure benzaldehyde and cyclohexanone were heated in a 50 ml round bottom flask at temperature ranging from 90-120°C for a period of 30-120minutes. The synthesis of the desired product α-α'-dibenzylidenecyclohexanone was carried out with molar ratio of benzaldehyde to cyclohexanone 2:1 and substrate to KFC catalyst weight ratio was taken as 10. After completion of the reaction, the reaction mixture was cooled and the solid catalyst was separated by filtration. The product formed and unreacted reactants present in the reaction mixture were analyzed by gas chromatography, with a flame ionization detector using a capillary column and N_2 as carrier gas.

Conversion (wt. %) = 100 X [Initial wt.% - Final wt.%] / Initial wt.%

In order to study the stability of the prepared catalyst, the solid catalyst recovered after the reaction by filtration was washed with acetone and calcined at 550°C for 2h to be used in the next reaction cycle. The activated catalyst was then added quickly into the flask containing mixture of 0.01 mol (.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde and was kept on hot oil bath at 120°C for 2h. At the end of the reaction the spent catalyst was filtered and washed with acetone to remove physically absorbed reactant and product molecules. The product was analyzed by Gas Chromatograph. A high conversion (82%) of

α-α'-dibenzylidenecyclohexanone was obtained. The catalyst was reused for next reaction cycle and again regenerated as earlier. Thus we can say that the catalyst used in our study was considerably stable and can be reused up to a minimum of three reaction cycles.

The prepared catalyst could be recycled up to three cycles with equal efficiency. The recycling was carried out by the process comprising the following steps

a) Performing base catalyzed condensation reaction in a solvent free medium, maintaining the substrate to catalyst weight ratio 10.
b) Maintaining the temperature of the reaction in the range of 90 to 120°C at atm pressure for a period selected from 30 to 120 min.
c) Separating the solid base catalyst from the reaction mixture by filtration.
d) Washing the catalyst with acetone to remove adhering materials
e) Drying the catalyst at 110°C for 2h followed by calcination at 550°C for 2h for reuse of catalyst.

The nano-crystalline catalyst provides large fraction of active species on the surface, thus generates the catalytic activity of the catalyst.
The present invention is reported for the first time as how a solid waste fly ash can be converted into an efficient solid base catalyst by mixing with Kota stone slurry (containing Ca and silica), thus exhibiting high basicity due to increased basic sites on the surface during activation.
In the process of this invention (i) conversion of fly ash into solid base catalyst by mixing of Kota stone slurry decreases the cost of catalyst and makes the process more cost effective as both the precursors are

industrial wastes (ii) the catalytic conversion takes place in solvent free environment; (iii) the catalyst used is solid catalyst hence it can be separated from the reaction product by simple filtration; (iv) a separated catalyst can be reused in the process of this invention; (v) also the catalyst is noncorrosive hence all the serious problem associated with homogeneous catalyst used in the earlier homogeneous processes for the preparation of condensation products has overcome in the process of this invention. This dispenses the need of the use of any solvent; (vi) the catalytic conversion is achieved in benign condition; (vii) the process does not involve any post reaction overcome for the disposal of corrosive and hazardous spent base; (viii) catalyst is develop from fly ash which find an effective and novel use of fly ash.

Example-1

The coal fly ash collected from Kota Super Thermal Power Station, was mechanically activated by ball milling to increase surface area with lower crystallite size as well as for generation of surface active sites. Mechanically activated fly ash was thermally activated by calcination at 900°C for 2h. Chemical activation was done by mixing of Kota stone slurry with fly ash in (1:1 ratio) with sufficient water. The mixture was heated at 110°C and refluxed for 24 h maintaining the temperature. The condenser was then removed and slurry was cooled at room temperature. The resulting paste was then filtered and dried at 110°C for 24h and further calcined at 550°C for 2h under static conditions.

Example-2

0.01 mol (0.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde with molar ratio 1:2 were taken in a 50 ml round bottom flask immersed in oil bath maintained at 90°C for 2 h. The KFC catalyst, activated at 550°C for 2h in static condition, with substrate to catalyst weight ratio of 10, was added in benzaldehyde to catalyst weight ratio 10:1 and the mixture was heated at 90°C for 120 minutes under stirring. After completion the reaction mixture was cooled and filtered to separate the catalyst. The filtered product was analyzed by gas chromatograph. A conversion of 52% of α-α'-dibenzylidenecyclohexanone was obtained with high selectivity 92%.

Example-3

0.01 mol (0.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde with molar ratio 1:2 were taken in a 50 ml round bottom flask immersed in oil bath maintained at 110°C for 2 h. The KFC catalyst, activated at 550°C for 2h in static condition, with benzaldehyde to catalyst weight ratio 10, was added and the mixture was heated at 110°C for 120 minutes. After the completion the reaction mixture was cooled and filtered to separate the catalyst. The filtered product was analyzed by gas chromatograph. A conversion of 67% of α-α'-dibenzylidenecyclohexanone was obtained with high selectivity 92%.

Example-4

0.01 mol (0.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde with molar ratio 1:2 were taken in a 50 ml round bottom flask immersed in oil bath maintained at 120°C for 2 h. The KFC catalyst, activated at 550°C for 2h in static condition with benzaldehyde

to catalyst weight ratio 10, was added and the mixture was heated at 120°C for 120 minutes. After the completion the reaction mixture was cooled and filtered to separate the catalyst. The filtered product was analyzed by gas chromatograph. A conversion of 82% of α-α'-dibenzylidenecyclohexanone was obtained with high selectivity 92%.

Example-5

0.01 mol (0.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde with molar ratio 1:2 were taken in a 50 ml round bottom flask immersed in oil bath maintained at 120°C for 2 h. The KFC catalyst, activated at 550°C for 2h in static condition with benzaldehyde to catalyst weight ratio 10, was added and the mixture was heated at 120°C for 30, 60 and 120 minutes. After the completion the reaction mixture was cooled and filtered to separate the catalyst. The filtered product was analyzed by gas chromatograph. A conversion of α-α'-dibenzylidenecyclohexanone obtained at 30, 60 and 120 minutes are 62, 79 and 82% respectively with high selectivity 92%.

Example-6

The spent catalyst, after filtration from the reaction was washed thoroughly with acetone and dried in oven at 110°C for 12h. Thus dried catalyst was activated at 550°C for 2h in flow of air. The regenerated catalyst was used for further reaction cycle under the similar reaction conditions. The regenerated catalyst was used for further reaction cycles, under similar reaction conditions i.e. 0.01 mol (0.86gm) of cyclohexanone and 0.02 mol (1.88gm) of benzaldehyde and was kept on hot oil bath at

120°C for 2h. Regenerated catalyst was added in reaction mixture with reactant to catalyst weight ratio 10 and the mixture was heated at 120°C for 2h. The reaction mixture was cooled and filtered to separate the catalyst. The catalyst was found equally efficient upto three reaction cycles, for giving similar conversion of the desired product which is confirmed by gas chromatograph.

The main Advantages of the process over conventional process include

1. The present process employs use of coal generated fly ash and Kota stone slurry (solid wastes) in the synthesis of solid base catalyst KFC.
2. The KFC is eco-friendly and cost effective, safe in handling and do not generate any byproduct.
3. An introduction of new class of solid base catalyst from waste is a novel use of fly ash thus minimizing the solid waste which is of great environmental concern as well as reduces product cost.
4. Catalyst being solid in nature can be easily separated from the liquid reaction mixture by means of filtration.
5. The catalyst of this invention shows very high catalytic activity towards condensation products.
6. The reaction process is a single step process without use of any solvent.
7. The activity of the catalyst used in the process of this invention is much higher in smaller quantity at moderate conditions of pressure and temperature.

8. The catalyst of this invention being nano-crystalline and thermally stable can be reused repeatedly in the process and the reused catalyst shows very high activity even after its repeated use in the process.
9. Fly ash based solid base catalyst is easy in transforming, handling etc. in comparision to the conventional catalyst like NaOH, KOH, KF, and $Ba(OH)_2$. This catalyst is equally effective like other solid bases such as hydrotalcite, $NaOH/Al_2O_3$, KF/Al_2O_3 etc.