Manoharbhai Shikshan Prasarak Mandal Armori's

# Rashtrapita Mahatma Gandhi Arts & Science College, Nagbhid, Dist-Chandrapur 441205

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# Self Study Report SSR: CYCLE-II

(2017-2018 To 2021-2022)

### Research, Innovations and Extension

3.2.1 Institution has created an ecosystem for innovations, Indian Knowledge System (IKS), including awareness about IPR, establishment of IPR cell, Incubation centre and other initiatives for the creation and transfer of knowledge/technology and the outcomes of the same are evident



### Manoharbhai Shikshan Prasarak Mandal, Armori's RASHTRAPITA MAHATMA GANDHI ART'S & SCIENCE COLLEGE, NAGBHID, DIST. CHANDRAPUR

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# Declaration

This is to declare that the information, reports, true copies etc. given in this file as a supporting documents is verified by IQAC and found correct.



Dr. G. D. Deshmukh

Principal



(Established by Government of Maharashtra Not-fication No MiSC-2007/(322/07) UNI-4 Dated 27° Sept. 2011 & Presently a State University governed by Maharashtra Public University Act, 2016 (Maharashtra Act No VI of 2017)

### (Office of the Registrar)

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E-Mail: registrar.gondwanauniversity.a/gmail.com Phone Fas: 07132-223104(o) Dr. Ishwar S. Mohurley Registrar

No./ GUG/Reg./9790/2020

Date: 29 / 09 / 2020

To
The Principal
R. M. G. Arts and Science College
Nagbhid, Distt – Chandrapur

Sub: Regarding Incubation Center.

Ref: Your Letter No. RMGC/513/2019 dtd. 11.09.2019.

Dear Sir,

With reference to the subject cited above, we are to inform you that as per the provisions specified in Maharashtra Public Universities Act'2016 under section 54(a) and (b), Hon'ble Vice Chancellor has permitted to establish and operate Ethno-medicine & herbal product based Incubation Center at your college with effect from 27<sup>th</sup> Sep'2019.

Herein we would like to mention that the given center shall undertake incubation of relevant and useful ideas such as product, process, service and innovation, relating to treatment of varied illness/diseases and health care in general, into a scalable mode so and such that shall lead to generation of enterprises by the incubates.

We do advise you for seeking all kinds of assistance including the financial one for achieving the specific objectives of the center through means of Central & State Govt. departments, Corporates, Private Funding agencies etc.

We wish you all the success for future innovations and endeavours.

Regards,

By the order of Hon'ble Vice-Chancellor

Dr. Ishwar S. Mohurley)



### (12) United States Patent Chilukuri et al.

#### (10) Patent No.: US 11,000,831 B2 (45) Date of Patent: May 11, 2021

### (54) TRANSITION METAL(S) CATALYST SUPPORTED ON NITROGEN-DOPED MESOPOROUS CARBON AND ITS USE IN CATALYTIC TRANSFER HYDROGENATION REACTIONS

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#### (73) Assignee: Council of Scientific & Industrial Research, New Delhi (IN)

Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 474 days.

| (21) | Appl. No.: | 15/758,804    |
|------|------------|---------------|
| (22) | PCT Filed: | Sep. 12, 2016 |

(86) PCT No.: // PCT/IN2016/050304 § 371 (c)(1), Mar. 9, 2018 (2) Date:

(87) PCT Pub. No.: WO2017/042838 PCT Pub. Date: Mar. 16, 2017

(65)**Prior Publication Data** US 2020/0230578 A1 Jul. 23, 2020

#### (30)Foreign Application Priority Data

| (2006.01) |
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(52) U.S. Cl. B01J 23/462 (2013.01); B01J 23/002 (2013.01); B01J 35/006 (2013.01); B01J 35/023 (2013.01); B01J 35/1014 (2013.01); B01J 35/1019 (2013.01); B01J 35/1023 (2013.01); B01J 35/1028 (2013.01); B01J 35/1038 (2013.01); B01J 35/1042 (2013.01); B01J 35/1047 (2013.01); B01J 37/04 (2013.01); B01J 37/06 (2013.01); B01J

37/343 (2013.01); C07D 307/36 (2013.01); B01J 21/18 (2013.01)

### (58) Field of Classification Search

CPC ... B01J 21/18; B01J 27/24; B01J 23/46; B01J 23/462; B01J 35/006; C07D 307/36 See application file for complete search history. SORIFICATION OF THE PARTY OF TH

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Primary Examiner - Noble E Jarrell (74) Attorney, Agent, or Firm - Dinsmore & Shohl, LLP

#### (57)ABSTRACT

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The present invention discloses a novel transition metal(s) catalyst supported on nitrogen-doped mesoporous carbon and a process for the preparation of the same. Further, the present invention discloses use of transition metal(s) supported on nitrogen-doped mesoporous carbon catalyst in catalytic transfer hydrogenation reaction. The invention also discloses an improved process for the synthesis of 2,5-Dimethylfuran (DMF) and 2-Methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural respectively, using alcohols as hydrogen donor over a transition metal supported on nitrogen-doped mesoporous carbon, especially ruthenium supported on nitrogen-doped mesoporous carbon without using any co-catalysts.

15 Claims, 6 Drawing Sheets

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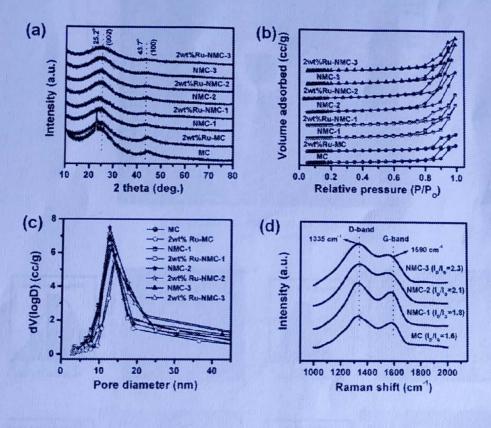


Fig. 1



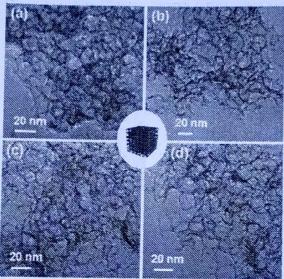


Fig. 2

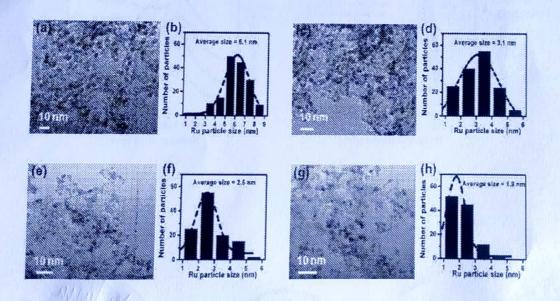
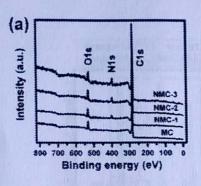
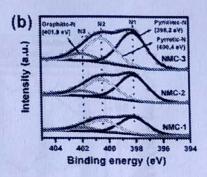


Fig. 3



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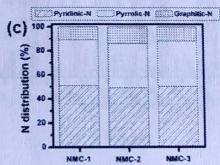


Fig. 4

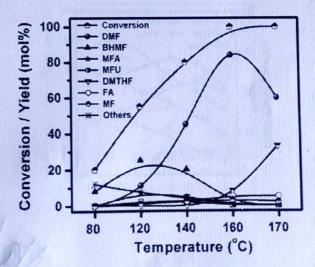
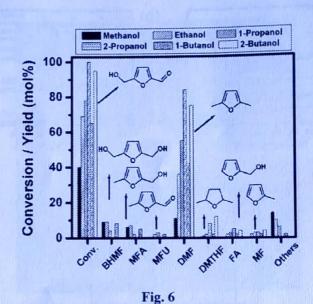


Fig. 5



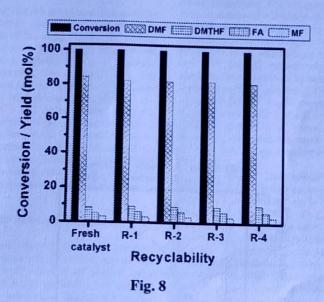
May 11, 2021



DMF Yield (mal%) 10 12 Time (h)

Fig. 7





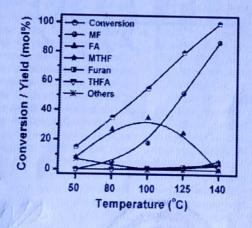


Fig. 9



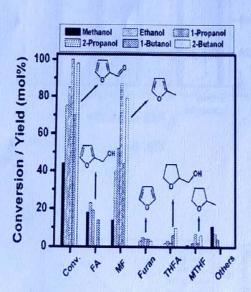


Fig. 10



#### TRANSITION METAL(S) CATALYST SUPPORTED ON NITROGEN-DOPED MESOPOROUS CARBON AND ITS USE IN CATALYTIC TRANSFER HYDROGENATION REACTIONS

#### FIELD OF THE INVENTION

The present invention relates to a novel transition metal(s) supported on nitrogen-doped mesoporous carbon and a 10 process for the preparation of the same. Further, the present invention discloses a use of transition metal(s) supported on nitrogen-doped mesoporous carbon catalyst in catalytic transfer hydrogenation reaction. The invention also discloses an improved process for the synthesis of 2,5-Dim- 15 ethylfuran (DMF) and 2-Methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural using transition metal(s) supported on nitrogen-doped mesoporous carbon catalyst. More particularly, the present invention also relates to a process for the preparation of 2,5-dimethylfuran (DMF) 20 and 2-methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural, respectively catalyzed by ruthenium (Ru) nanoparticles supported on nitrogen doped mesoporous carbon

#### BACKGROUND AND PRIOR ART

In recent years, 2,5-dimethylfuran (DMF), which is produced by the selective hydrogenolysis of biomass-derived 5-hydroxymethylfurfural (HMF) and is considered as a new-fashioned liquid biofuel for transportation, has received much more attention from many researchers in the world. Compared to the current market-leading bioethanol, DMF possesses a higher energy density, a higher boiling point and a higher octane number and is immiscible with water. In 35 view of the diminishing oil reserves and the clear visible signs of climate change attributed to greenhouse gas emissions to the atmosphere, utilization of renewable biomass sources for the production of fuels and chemicals has become imperative. Worldwide, there is a remarkable 40 research activity in this area for the development of processes and catalysts for transforming abundant lignocellulosic biomass into liquid fuels for the transportation sector as well as to prepare platform chemicals. For these efforts to become a commercial reality the developed process has to 45 be economically feasible and the developed catalyst should be cheap and easy to scale-up. Hydrogenolysis reaction holds great promise for the proposed bio-refinery concept, as biomass-derived substrates possess very high oxygen content, which need to be reduced to get useful chemicals and 50 fuels. The selective hydrogenolysis of furfural and 5-hydroxymethylfurfural (HMF) to 2-methylfuran (MF) and 2,5-dimethylfuran (DMF), respectively, holds great potential for the production of liquid fuel substitutes/additives from renewable compounds. Furfural and HMF can be obtained from the acid-catalyzed dehydration of hemicellulose and cellulose-derived carbohydrates, respectively. Both DMF and MF have desirable chemical and physical properties to be used as transportation fuels; with nearly 40% more energy density (35 MJkg<sup>-1</sup>, 31.2 MJkg<sup>-1</sup>) than etha- 60 nol. They also have high research octane numbers (RON=119, RON=131), very little solubility in water (2.3 gL-1, 7.0 gL-1) and ideal boiling points (92-94° C., 64.7° C.). Furthermore, DMF consumes only one-third of the energy for the separation by distillation, compared to that 65 required for the separation of ethanol from fermented broth The DMF and MF have been successfully tested as biofuels

in a single cylinder spray guided direct-injection sparkignition engine. Their performance was satisfactory against gasoline in terms of ignition, emission and combustion properties. These excellent characteristics make DMF and MF as promising renewable fuels for transportation.

One of the vital challenges for the upgradation of HMF and furfural to value added chemicals and fuels is the product selectivity. For instance, hydrogenation of HMF and furfural results in a mixture of ring- and side chain-hydrogenated products along with ring-opening products. Decoupling these processes is highly important for using furans as renewable fuels. The hydrogenolysis of HMF and furfural into DMF and MF, respectively, were studied over various supported metal catalysts by using molecular hydrogen. The break-through of preparing DMF from biomass-derived compound (HMF) was first reported by Roman-Leshkov et al. using a bimetallic CuRu/C catalyst with 71% DMF yield at 220° C. and 6.8 bar H2 pressure. Chatterjee et al. reported the use of Pd/C catalyst in supercritical CO2 to get high yield of DMF. But, this process involves the use of high pressure CO, and H,

Article titled "Chemoselective Hydrogenation of Biomass-Derived 5-Hydroxymethylfurfural into the Liquid Biofuel 2,5-Dimethylfuran" by L Hu et al. published in Ind. 25 Eng. Chem. Res., 2014, 53 (24), pp 9969-9978 reports catalytic systems and the latest research achievements for the selective hydrogenation of HMF into DMF in light of the diversity of hydrogen donors such as molecular hydrogen, formic acid, alcohols, and water are systematically summarized and discussed. The study shows the maximum yield of DMF up to 94.6% and 100% conversion of HMF by using Ru/C as catalyst in presence of THF solvent and molecular hydrogen.

Article titled "Selective Transformation of 5-Hydroxymethylfurfural into the Liquid Fuel 2,5-Dimethylfuran over Carbon-Supported Ruthenium' by L Hu et al. published in Ind. Eng. Chem. Res., 2014, 53 (8), pp 3056-3064 reports a simple and efficient process for the selective hydrogenation of 5-hydroxymethylfurfural (HMF) into the high-quality liquid fuel 2,5-dimethylfuran (DMF) in the presence of tetrahydrofuran (THF) using carbon-supported ruthenium (Ru/C) which led to 94.7% DMF yield with 100% HMF conversion at a relatively mild reaction temperature of 200° C. for only 2 h.

Article titled "Effect of hydrogen donor on liquid phase catalytic transfer hydrogenation of furfural over a Ru/RuO<sub>2</sub>/C catalyst" by P Panagiotopoulou et al. published in Applied Catalysis A: General, 2014, 480, pp 17-24 reports catalytic transfer hydrogenation of furfural toward methyl furan over a Ru/C catalyst in the temperature range of 120-200° C. using 2-propanol as a solvent. An optimum methyl furan yield of 61% has been achieved using said process as reported.

Article titled "Production of Dimethylfuran from Hydroxymethyl-furfural through Catalytic Transfer Hydrogenation with Ruthenium Supported on Carbon" by J Jae et al. published in ChemSusChem, 2013, 6(7), pp 1158-62 reports transfer hydrogenation using alcohols as hydrogen donors and supported ruthenium catalysts results in the selective conversion of hydroxymethylfurfural to dimethylfuran (>80% yield).

Article titled "Novel microwave synthesis of ruthenium nanoparticles supported on carbon nanotubes active in the selective hydrogenation of p-chloronitrobenzene to p-chloroniline" by C Antonetti et al. published in Applied Catalysis A: General, 2012, 421-422, pp 99-107 reports selective hydrogenation of p-chloronitrobenzene (p-CNB) to p-chloroniline

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Rashtrapita Mahatma Gandhi Art's & Science College, Nagbhid, Dist. Chandrapur roaniline (p-CAN) using carbon nanotubes (CNTs) supported ruthenium nanoparticles with almost complete substrate conversion with total selectivity to the target product.

Article titled "Ruthenium nanoparticles supported on nitrogen-doped carbon nanofibers for the catalytic wet air oxidation of phenol" by AB Ayusheev et al. published in Applied Catalysis B: Environmental, 2014, Volume 146, pp 177-185 reports effect of nitrogen content in N-doped carbon nanofibers (N-CNFs) on the catalytic activity of Ru/N-CNFs in the wet air oxidation of phenol. In the case of 10 Ru-containing catalysts, nitrogen in N-CNFs was found to be responsible for both the increased activity and stability of the catalysts toward deactivation. The Ru/N-CNFs catalyst is prepared by using wetness impregnation technique.

Article titled "Ruthenium Nanoparticles Supported on 15 Carbon Nanotubes for Selective Hydrogenolysis of Glycerol to Glycols" by J Wang et al. published in Chemistry Letters, 2009, 38(6), pp 572-573 reports Ru nanoparticles supported on carbon nanotubes showed efficient reactivity for hydrogenolysis of aqueous glycerol solution to produce glycols of 20 1,2-propanediol and ethylene glycol. The glycerol conversion along with the product selectivity depended essentially

on the mean size of Ru nanoparticles.

Most of the reported processes for the hydrogenolysis of HMF to DMF and furfural to MF were conducted under 25 high-pressure H2 and at elevated reaction temperatures. Such type of processes are energy intensive, thus difficult to commercialize. It is desirable to minimize the use of external H<sub>2</sub> required for biofuel production, especially if this H<sub>2</sub> has to be prepared from fossil fuels. The catalytic transfer 30 hydrogenation (CTH) process for reducing the substrates has advantages compared to processes involving molecular H2. Several investigators employed CTH process for the conversion of HMF to DMF and furfural to MF over various metal catalysts. Instead of molecular H2, secondary alco- 35 hols, supercritical methanol, cyclohexane or 1,4-butanediol were used as a hydrogen source as well as reaction media. Rauchfuss et al. reported DMF yield up to 95% using Pd/C as catalyst and formic acid as a hydrogen donor. However, this process needs simultaneous use of formic acid and 40 H<sub>2</sub>SO<sub>4</sub> to get high yield of DMF. Use of acids is not environment friendly and also requires corrosion-resistant equipment for the process that will add cost to the process. Moreover, the reported CTH processes include the use of mineral acids as co-catalysts to improve the hydrogenation 45 activity, which are difficult to separate from the reaction mixture and are responsible for undesirable side reactions.

Therefore, there is need to develop an environment friendly alternative process for the hydrogenolysis of HMF to DMF and furfural to MF by CTH. Accordingly, the 50 present inventors developed an improved process for hydrogenolysis of HMF to DMF and furfural to MF using alcohols as hydrogen donor instead of molecular H<sub>2</sub> over a Ru supported on nitrogen-doped mesoporous carbon without using any co-catalysts.

#### **OBJECT OF THE INVENTION**

The main object of the present invention is to develop a novel transition metal(s) catalyst supported on nitrogendoped mesoporous carbon. The another object of the present invention to provide a process for the preparation of the same. Further, object the present invention is use of transition metal(s) catalyst supported on nitrogen-doped mesoporous carbon in catalytic transfer hydrogenation reaction. 65 Yet another object of the invention is to provide an improved process for the synthesis of 2,5-Dimethylfuran (DMF) and

2-Methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural using transition metal(s) catalyst supported on nitrogen-doped mesoporous carbon.

### SUMMARY OF THE INVENTION

Accordingly present invention provides a catalyst composition comprising;

a transition metal supported on nitrogen doped mesoporous carbon; wherein the transition metal(s) in the range of 0.5 to 10 weight % of the catalyst.

In another embodiment the transition metal(s) of the catalyst is selected from the group consisting of Ru, Pt, Pd, Rh, Au, Ag, Os, Ir, Cu, Ni, Re, Cr, Mn, Fe, Zn, Co; either alone or in combination of any two or more metals

In another embodiment the BET surface area of the transition metal(s) catalyst in the range of 30 to 1200 m<sup>2</sup>/g. In another embodiment the BET surface area of the transition metal(s) catalyst in the range of 36 to 1000 m<sup>2</sup>/g. In another embodiment the total pore volume of the

transition metal(s) catalyst in the range of 0.07 to 1.2 cc/g.

In another embodiment the average particle/crystal size of

the transition metal(s) in the range of 1 to 10 nm.

In another embodiment the present invention provides a

process for synthesis of metal(s)-nitrogen doped mesoporous carbon catalyst comprising the steps of;

 a. dispersing nitrogen doped mesoporous carbon in deionized water under ultrasonication to obtain high dispersion,

b. charging transition metal chloride to above dispersion obtained in step (a) under ultra sonication and stirring the mixture at 70-90° C. for 6 to 12 hours.

c. cooling the mixture of step (b) and charging aqueous solution of NaBH<sub>4</sub> slowly under ultrasonication and stirring for 30-60 minutes.

d. filtering the solution and washing with deionized water and drying at 80-100° C. for 10-12 hours to obtain the catalyst.

In another embodiment the transition metal chloride selected from the chlorides of Ru, Pt, Pd, Rh, Au, Ag, Os, Ir, Cu, Ni, Re, Cr, Mn, Fe, Zn, Co; either alone or in combination of any two or more metal chlorides.

In another embodiment, the present invention provides a process for the preparation of 2,5-Dimethylfuran (DMF) and 2-Methylfuran (MF) from 5-Hydroxymethylfurfural (HMF) and furfural using alcohols as hydrogen donor instead of molecular  $\rm H_2$  over transition metal supported on nitrogendoped mesoporous carbon, especially Ru supported on nitrogen-doped mesoporous carbon without using any co-catalysts.

In another embodiment the transition metal supported on nitrogen-doped mesoporous carbon used is Ru supported on nitrogen-doped mesoporous carbon for the preparation of 2,5-Dimethylfuran (DMF) and 2-Methylfuran (MF) from 5-Hydroxymethylfurfural (HMF) and furfural

In another embodiment the catalytic transfer hydrogenation (CTH) agents are selected from alcohols, organic acids or organic acid salts or mixture thereof.

In another embodiment the alcohol used as catalytic transfer hydrogenation (CTH) agents is selected from the group consisting of 2-propanol, 2-butanol, methanol, ethanol, 1-propanol.

In another embodiment the organic acid used as catalytic in. 65 transfer hydrogenation (CTH) agents is selected from the group consisting formic acid, acetic acid, propionic acid and advantage acid etc.

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In another embodiment the catalytic transfer hydrogenation reaction is carried out in the presence of molecular hydrogen alone or along with catalytic transfer hydrogenation agent.

In another embodiment the temperature of the catalytic transfer hydrogenation reaction is in the range of  $80-170^{\circ}$  C.

In preferred embodiment, the nitrogen content in said transition metal catalyst support is 3-15 wt %.

In preferred embodiment, the metal content in said ruthenium catalyst in the range of 0.5 to 10 weight % of the 10 catalyst

In preferred embodiment, the nitrogen content in said ruthenium catalyst support is 3-15 wt %.

In another preferred embodiment, the yield of 2,5-Dimethylfuran (DMF) is greater than 80% and conversion of 15 5-hydroxymethylfurfural (HMF) is 100% in catalytic transfer hydrogenation reaction.

In another preferred embodiment, the yield of 2,5-Dimethylfuran (DMF) is greater than 80% and conversion of 5-hydroxymethylfurfural (HMF) is 100% when ruthenium 20 metal nitrogen doped mesoporous carbon used as catalyst.

metal nitrogen doped mesoporous carbon used as catalyst. In still another preferred embodiment, the yield of 2-methylfuran (MF) is >87% and conversion of furfural is 100% in catalytic transfer hydrogenation reaction.

In still another preferred embodiment, the yield of 252-methylfuran (MF) is >87% and conversion of furfural is 100% when ruthenium metal nitrogen doped mesoporous carbon used as catalyst.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1: (a) XRD, (b) N<sub>2</sub> adsorption-desorption isotherm, (c) BJH pore size distribution and (d) Raman spectra of the various samples.

FIG. 2: TEM image of the (a) MC, (b) NMC-1, (c) 35 NMC-2 and (d) NMC-3.

FIG. 3: TEM images and the Ru nanoparticles size distribution for 2 wt % Ru-MC (3a and 3b), 2 wt % Ru-NMC-1 (3c and 3d), 2 wt % Ru-NMC-2 (3e and 3f) and 2 wt % Ru-NMC-3 (3g and 3h), respectively.

FIG. 4: (a) XPS survey and (b) high-resolution N is spectra of the NMCs. (c) The distribution of N species in the NMCs from resolving peaks of the N 1s spectra.

FIG. 5: Effect of reaction temperature on HMF conversion and product yields. Reaction conditions: HMF (1 mmol); 45 catalyst (2 wt % Ru-NMC-3, 25 mg); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (2-propanol, 25 mL); time (8 h). Other products include ethers.

FIG. 6: Effect of hydrogen donor on HMF conversion and product yields. Reaction conditions: HMF (1 mmol); catalyst (2 wt % Ru-NMC-3, 25 mg); temperature (160° C.); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (25 mL); time (8 h). Other products include ethers.

FIG. 7: Effect of H<sub>2</sub> pressure on DMF yields over 2 wt % Ru-NMC-3 as a function of reaction time. Reaction conditions: HMF (1 mmol); catalyst (25 mg); temperature (160° C.); solvent (THF, 25 mL).

FIG. 8: Recyclability study in CTH of HMF to DMF over 2 wt % Ru-NMC-3. Reaction conditions: molar ratio of HMF to Ru (200); temperature (160° C.); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (2-propanol, 25 mL); time (8 h).

FIG. 9: Effect of reaction temperature on furfural conversion and product yields. Reaction conditions: furfural (5 mmol); catalyst (2 wt % Ru-NMC-3, 25 mg); N<sub>2</sub> pressure 65 (20 bar); solvent and hydrogen donor (2-propanol.) The time (10 h). Others include ethers.

FIG. 10: Effect of hydrogen donor on furfural conversion and product yields. Reaction conditions: furfural (5 mmol); catalyst (2 wt % Ru-NMC-3, 25 mg); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (25 mL); time (10 h). Others include ethers.

### DETAILED DESCRIPTION OF THE INVENTION

The invention will now be described in detail in connection with certain preferred and optional embodiments, so that various aspects thereof may be more fully understood and appreciated.

In view of the above, the present invention provides a process for the preparation of 2,5-dimethylfuran (DMF) and 2-methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural, respectively using alcohols as hydrogen donor instead of molecular H<sub>2</sub> over Ru supported on nitrogendoped mesoporous carbon without using any co-catalysts.

In an embodiment, the present invention provides an improved process for the synthesis of 2,5-dimethylfuran (DMF) and 2-methylfuran (MF) from 5-hydroxymethylfurfural (HMF) and furfural, respectively catalyzed by transition metal nanoparticles supported on nitrogen doped mesoporous carbon in the presence of 2-propanol as hydrogen donor in the temperature range of 80-170° C., in that nitrogen content in said catalyst is in the range of 3-15 wt %.

In preferred embodiment, the yield of 2,5-Dimethylfuran (DMF) is greater than 80% and conversion of 5-hydroxym-30 ethylfurfural (HMF) is 100%.

In another preferred embodiment, the yield of 2-methylfuran (MF) is >87% and conversion of furfural is 100%.

In still another preferred embodiment, said reaction is carried out at temperature range in the range of 80-170° C.

The ruthenium content in said catalyst is in the range of 1-10 wt %. Preferably the ruthenium content in said catalyst is 2 wt %.

In one embodiment, the transition metals are selected from Ru, Pt, Pd, Rh, Au, Ag, Os, Ir, Cu, Ni, Re, Cr, Mn, Fe,

Zn, Co; either alone or in combination of any two or more metals in the range of 0.5-10 wt % of the catalyst.

The NMCs are prepared by a colloidal silica nanocasting route, which involved the mixing of melamine-phenolformaldehyde polymer sol with colloidal silica nanoparticles to obtain composite hydrogel. Its subsequent carbonization in N2 atmosphere at 800° C. followed by silica dissolution by treatment with NaOH, gave NMCs with disordered mesopores (Table 1). The X-ray diffraction (XRD) patterns of nitrogen-free carbon (MC), nitrogen-doped carbon samples (NMC-1, NMC-2 and NMC-3) and as-synthesized 2 wt % Ru deposited on various supports are shown in FIG. 1a. The formation of highly dispersed Ru nanoparticles is proven by XRD (FIG. 1a), as no diffraction peaks pertaining to metallic Ru are observed. Moreover, no change in diffraction peaks of the supports on Ru loading is observed, indicating that the Ru loading has no effect on the structure of the supports

As shown in FIG. 1b, support materials and Ru containing samples shows similar type IV adsorption-desorption isotherms with  $\rm H_2$  hysteresis loop, corresponding to the typical mesoporous structure of the materials. These results suggest that variation in nitrogen content does not strongly affect the mesoporous structure of the samples. Their BJH pore size distributions are similar in the range of 10-20 nm (FIG. 1c).

The formation of graphitic ordered carbon is further proved by Raman spectroscopy (FIG. 1d). Raman spectra show characteristic D- and G-bands of disordered graphitic

carbon at 1335 and 1590 cm $^{-1}$  respectively. The intensity ratios of the D-band to the G-band ( $I_D/I_G$ , calculated from integral area of the peaks) are 1.6, 1.8, 2.1 and 2.3 for MC, NMC-1, NMC-2 and NMC-3, respectively. These results show that the defects in graphite-like layers enhanced with increasing nitrogen content.

The mesoporous structure of the MC and nitrogen-doped carbon samples is further confirmed by transmission electron microscopy (TEM) observations (FIG. 2). From TEM images, nitrogen containing samples (FIGS. 2b, 2c and 2d) have no differences when compared with their nitrogen-free counterpart (FIG. 2a), consisting of spherical mesopores with a disordered amorphous carbon structure. The Ru nanoparticles sizes as well as their distribution for various Ru containing catalysts are investigated by TEM (FIG. 3). The TEM results suggested that the dominant contributor for stabilizing Ru nanoparticles is the nitrogen doped in the carbon structure and increasing nitrogen content in the sample led to decrease in the size of Ru nanoparticles.

The X-ray photoelectron spectroscopy (XPS) is employed to investigate the nitrogen-carbon bonds formed in the carbon framework. In XPS only oxygen, carbon and nitrogen are detected in the overall region scanned (FIG. 4a). XPS of C is shows very similar peak at 284.6 eV, which is associated with the graphitic carbon, suggesting that most of the C atoms are assembled in conjugated honeycomb lattices.

The catalyst evaluation for hydrogenolysis of HMF to get DMF is performed using 2-propanol as a hydrogen donor in 3 a batch reactor. FIG. 5 shows HMF conversion and product distributions as a function of reaction temperature over 2 wt % Ru-NMC-3 catalyst. The catalytic transfer hydrogenation (CTH) reaction occurs at a lower reaction temperature (80° C.) and DMF yield increased with increasing temperature. 35 At lower temperature, the primary product is 2,5-bis(hydroxymethyl)furan (BHMF) and upon increasing the temperature to 160° C., BHMF is completely converted to DMF with a yield up to 84 mol % at 100 mol % HMF conversion after 8 h of reaction. Other products observed at 120 and 140° C. are hydrogenated furans: 5-methyl furfural (MFU), 5-methyl furfuryl alcohol (MFA), 2-methylfuran (MF) and furfuryl alcohol (FA). The formation of FA indicates the occurrence of decarbonylation reaction, however, the yield of FA is low (<4 mol %). The formation of MFU suggested 45 that hydrogenolysis of -OH group in HMF occur in parallel to the hydrogenation of the -CHO group. In addition, ethers are also observed at lower temperature (80 and 120° C.), which are formed via etherification of BHMF or MFA and 2-propanol. They are not observed upon increasing the 50 reaction temperature (160 and 170° C.)

The effect of hydrogen donor on HMF conversion and product yields is investigated at 160° C. over 2 wt % Ru-NMC-3 catalyst. The results (FIG. 6) clearly indicate that the activity of catalyst heavily depends on the type of 55 alcohol used. The HMF conversion increased from 40 to 100 mol % and the following reactivity order is observed: methanol<l-butanol<ethanol<l-propanol<2-butanol<2-propanol. Among the hydrogen donors tested, 2-propanol showed highest activity giving 84 mol % DMF yield. The formation of ethers is negligible in 2-propanol and 2-butanol but significant in other alcohols (2-14 mol %). This shows that etherification reaction favored with lower chain alcohols (methanol, ethanol and 1-propanol) indicative of steric effect.

In preferred embodiment, 2-propanol is used as hydrogen donor.

In order to investigate the effect of nitrogen content of the catalyst on CTH activity, the Ru catalyst supported on various nitrogen containing mesoporous carbons and nitrogen-free carbon are tested for HMF hydrogenolysis (Table 3). The HMF conversions are 41, 61, 79 and 100 mol % with DMF yield of 11, 30, 55 and 84 mol % on 2 wt % Ru-MC, 2 wt % Ru-NMC-1, 2 wt % Ru-NMC-2 and 2 wt % Ru-NMC-3 catalysts, respectively, which shows that the catalytic activity is improved with increasing nitrogen content in the catalyst. The high nitrogen content in NMC-3 not only led to very stable and homogeneous dispersion of Ru nanoparticles but also enriches the electron density of the Ru nanoparticles, which accelerates the hydrogenolysis reaction, as compared to nitrogen-free catalyst (2 wt % Ru-MC). 5 The results also shows that the nitrogen plays a key role in the dehydrogenation of 2-propanol leading to higher H2 pressure

In preferred embodiment, nitrogen content in said catalyst is 3-15 wt %.

It is possible that adsorbed hydrogen species formed by the dehydrogenation of 2-propanol during CTH process can directly hydrogenate HMF molecule adsorbed on Ru nanoparticles instead of being released as H<sub>2</sub>. To understand this aspect, HMF hydrogenation is performed at H<sub>2</sub> pressure of 7 bar (total pressure developed including H<sub>2</sub> partial pressure during CTH reaction at 160° C. over 2 wt % Ru-NMC-3 with 2-propanol) using tetrahydrofuran (THF) as solvent. The results shows HMF conversion and DMF yield were 86 and 59 mol %, respectively, which are inferior to those using 2-propanol as hydrogen donor (compare entry 5 and 7 in Table 3). This result strongly suggests that HMF hydrogenation using hydrogen donors can be more efficient than that using external H<sub>2</sub> gas.

The 2 wt % Ru-NMC-3 catalyst is compared with 2 wt % Ru-AC under the same reaction conditions as well as with other oxide-supported Ru nanoparticles, including 2 wt % Ru-CeO<sub>2</sub>, 2 wt % Ru-MgO, 2 wt % Ru-TiO<sub>2</sub>, 2 wt % Ru-Mg(Al)O and 2 wt % Ru-y-Al2O3. Catalyst 2 wt % Ru-NMC-3 showed remarkably high activity in CTH, giving 100 mol % HMF conversion and 84 mol % DMF yield (Table 4). Oxide and AC supported Ru nanoparticles were not effective for this reaction, affording lower yields of DMF than 2 wt % Ru-NMC-3. In case of 2 wt % Ru-γ-Al<sub>2</sub>O<sub>3</sub> and 2 wt % Ru-TiO2, the main product was BHMF (entry 6 and 7, Table 4). These results show that these two catalysts have ability towards carbonyl group hydrogenation to hydroxyl group, but have less ability for C-O bond hydrogenolysis. From these results, it is clear that the nature of the support plays an important role in the activity and selectivity of Ru based catalysts in CTH reactions.

The CTH of HMF is investigated using NMC-3 supported Ru, Pt, Pd, Rh, Ni, Cu and Au catalysts. The HMF conversion varied in the range of 21 to 100 mol % and followed the reactivity order of Cu<Ni<Au<Rh<Pt<Pd=Ru (Table 5). Gratifyingly, an excellent DMF yield of 84 mol % with 100 mol % HMF conversion was obtained over Ru-based catalyst (Table 5, entry 1). Interestingly, when the reaction was conducted with Pd catalyst, significant amount of decarbonylation product FA was obtained (Table 5, entry 3), indicating Pd is more suitable for HMF decarbonylation.

The effect of H<sub>2</sub> pressure on the DMF yields is studied by varying the pressure from 2-15 bar at 160° C. in THF solvent over 2 wt % Ru-NMC-3 catalyst (FIG. 7). When the reaction was carried out at low pressure (2 and 5 bar), the intermediate products such as BHMF and MFA formed in significant quantities, which may be converted to DMF on prolonging allowed by the converted to DMF on prolonging the pressure (7 bar);

the DMF yield reaches the maximum (68 mol %) and the yield decreased on continuation of the reaction for further duration. This decrease in DMF yield is due to its ring hydrogenation, leading to the formation of 2,5-dimethyltetrahydrofuran (DMTHF). Increasing H<sub>2</sub> pressure further to 5 10 and 15 bar, has an adverse effect on the DMF yield. At higher H<sub>2</sub> pressure, DMF yield suppressed mostly due to the increased rate of consecutive ring hydrogenation of DMF, led to the formation of DMTHF in significant amounts. Moreover, the concentration of other undesirable by-products such as 5-methyl tetrahydrofurfuryl alcohol (MTHFA), 2,5-bis(hydroxymethyl)tetrahydrofuran (BHMTHF) and hexanediol increased at higher H<sub>2</sub> pressure.

The recyclability of the 2 wt % Ru-NMC-3 catalyst is evaluated by repeating the reaction with the same catalyst 15 (FIG. 8). The results show that the catalytic performance remains same even after being reused for four times. These results indicate good stability of the catalyst. ICP-OES analysis showed no leaching of Ru after each recycle. Moreover, the amount of Ru in the catalyst after four cycles 20 is found to be similar to that of starting catalyst.

The 2 wt % Ru-NMC-3 catalyst was also investigated for hydrogenolysis of furfural to get 2-methylfuran (MF). The effect of temperature on the furfural conversion and product yields over 2 wt % Ru-NMC-3 catalyst is investigated by 25 varying the reaction temperature in the range of 50-140° C. (FIG. 9). The primary product of the transfer hydrogenation of furfural is FA, which is prominent at lower temperature (80, 100 and 125° C.). In a consecutive step, hydrogenolysis of the —OH group in FA yields MF up to 87 mol % at 140° 30 C. In parallel to hydrogenation, furfural also undergoes decarbonylation reaction to furan. Furthermore, furan-ring hydrogenated products like 2-methyl tetrahydrofuran (MTHF) and tetrahydrofurfuryl alcohol (THFA) have also been observed.

The effect of hydrogen donor on furfural conversion and product yields is investigated at 140° C. over 2 wt % Ru-NMC-3 catalyst (FIG. 10). The results clearly suggested that the catalyst activity is greatly depends on the nature of hydrogen donor. The furfural conversion increased from 44 to 100 mol % and follows the reactivity order: methanol<1-butanol<ethanol<1-propanol<2-butanol<2-propanol.

Among the hydrogen donors tested. 2-propanol displayed

Among the hydrogen donors tested, 2-propanol displayed the highest activity leading to 87 mol % MF yield. These results are in good agreement with the reported results, 45 which normally reports that in CTH secondary alcohols have a better tendency to release hydrogen than primary alcohols.

The following examples, which include preferred embodiments, will serve to illustrate the practice of this invention, it being understood that the particulars shown are 50 by way of example and for purpose of illustrative discussion of preferred embodiments of the invention.

#### **EXAMPLES**

#### Example 1: Synthesis of Nitrogen-Doped Mesoporous Carbons (NMC)

The NMCs were prepared by colloidal silica assisted sol-gel process, using melamine as a nitrogen source. In a typical synthetic procedure, 3.67 g of phenol (39 mmol) and 6.33 g of formaldehyde (78 mmol) were added drop-wise to 50 mL of NaOH solution (0.2 M, 10 mmol) under stirring. This mixture was stirred at room temperature for 20 min and then heated in an oil bath at 70° C. while stirring, for 40 to but a minutes. About 4.92 g of melamine (39 mmol) and another part of formaldehyde (9.5 g, 107 mmol) were then added to off nit still the s

the above solution with continuous stirring for 30 min. This was followed by the addition of 50 g of Ludox SM-30 sol (30 wt % SiO<sub>2</sub>) with stirring for 1 h. The suspension was then transferred to sealed bottle and heated at 80° C. for 3 days. The obtained gels were dried at 80° C. and powdered using mortar-pestle. The material obtained was carbonized in nitrogen flow at 800° C. for 3 h while raising the temperature at a heating rate of 5° C./min. The NMC was obtained after dissolution of the silica in 2M NaOH solution at 80° C. for 12 h. The solid obtained was washed with distilled water until pH is close to neutral and dried at 100° C. for 10 h. The NMCs with different nitrogen contents were prepared by changing the mole ratio of melamine to phenol. The nitrogen free mesoporous carbon (MC) was also prepared by using above process without adding any melamine content.

#### Example 2: Synthesis of Ru-NMC

The catalyst 2 wt % Ru-NMC was synthesized by modified ultrasonic-assisted method. Typically, 0.1 g of NMC was dispersed in 50 mL of deionized water in a 100 mL round bottom flask by ultrasonication (20 min). To it, 0.5 mL of aqueous solution of RuCl<sub>3</sub> (Ru content 4 mg/mL) was added under agitation in ultrasonicator. This mixture was stirred at 80° C. for 6 h and cooled to room temperature. Then, aqueous solution of NaBH<sub>4</sub> (Ru/NaBH<sub>4</sub>=1:4 mol mol-1) was added to it slowly under ultrasonication (30 min). The solution was filtered and washed with deionized water. The resulting 2 wt % Ru-NMC catalyst was dried at 80° C. for 10 h and used as catalyst for hydrogenolysis reactions. Catalyst samples with different Ru loadings (0.5, 1 and 5 wt %) were prepared using a similar procedure by taking appropriate amounts of RuCl3. Catalysts 2 wt % Ru-MC, 2 wt % Ru-AC (AC-activated carbon), 2 wt % Pd-NMC, 2 wt % Pt-NMC, 2 wt % Rh-NMC, 5 wt % Ni-NMC, 5 wt % Cu-NMC and 2 wt % Au-NMC were also prepared by adopting above procedure.

#### Example 3: Hydrogenolysis of HMF or Furfural

All the reactions were carried out using 100 mL Parr autoclave (SS316). In a typical experiment, the reactor was charged with 1 mmol HMF (or 5 mmol furfural), hydrogen donor (25 mL), n-decane (0.2 g, internal standard) and required amount of freshly prepared catalyst. The reactor contents were mixed thoroughly and the reactor was sealed, purged 2-3 times with N<sub>2</sub> and pressurized to 20 bar N<sub>2</sub> pressure. Subsequently, the reaction vessel was heated under stirring at required temperature for a desired duration. Liquid samples were withdrawn periodically during the reaction and analyzed by GC (Agilent 7890A) equipped with a flame ionization detector (FID) having CP Sil 8CB capillary column (30 m length, 0.25 mm diameter). Product identification was done using authentic standards and GC-MS (Varian, Saturn 2200) analysis.

#### Example 4: Characterization of Catalysts

The NMCs were prepared by a colloidal silica nanocasting route, which involved the mixing of melamine-phenol-formaldehyde polymer sol with colloidal silica nanoparticles to obtain composite hydrogel. Its subsequent carbonization in N<sub>2</sub> atmosphere at 800° C. followed by silica dissolution by treatment with NaOH, gave NMCs with disordered mesopores (Table 1). The X-ray diffraction (XRD) patterns of nitrogen-free carbon (MC), attrogen-doped carbon

TABLE 2-continued Physico-chemical properties of Ru-containing catalysts BET Ru partic pore volume[b] surface content[a] (nm)  $(cm^3/g)$ (wt %)  $(m^2/g)$ Catalyst 0.22 130 1.79 2 wt % Ru-Mg(Al)O 0.07 1.86 2 wt % Ru-TiO<sub>2</sub> 10 2 wt % Ru-y-Al<sub>2</sub>O<sub>3</sub> [a]Estimated by [CP-OES.

2 wt % Ru deposited on various supports are shown in FIG. 1a. These samples exhibited very similar diffraction features. Broader diffraction peaks are observed at around 25.2 (20) and 43.7° (20) that correspond to the (002) and (100) 5 planes of graphite, respectively. The (002)-diffraction peak gradually broadens with the increasing nitrogen content in the sample. The intensity of (100)-diffraction peak related to interlayer organization gradually decreases with increasing nitrogen content and even disappears when nitrogen content reaches 11.6 wt % (NMC-3). However, the positions of two peaks (25.2 and 43.7°) do not change with the nitrogen content. The formation of highly dispersed Ru nanoparticles was proven by XRD (FIG. 1a), as no diffraction peaks pertaining to metallic Ru were observed. Moreover, no change in diffraction peaks of the supports on Ru loading was observed, indicating that the Ru loading has no effect on the structure of the supports. As shown in FIG. 1b, support materials and Ru containing samples shows similar type IV 20 adsorption-desorption isotherms with an H2 hysteresis loop, corresponding to the typical mesoporous structure of the materials. These results suggest that variation in nitrogen content does not strongly affect the mesopore structure of the samples. Their BJH pore size distributions are similar in the range of 10-20 nm (FIG. 1c). The physico-chemical properties of various samples are summarized in Table 1 and Table 2. The micropores created by the release of volatiles during the pyrolysis under inert gas flow are dependent on the melamine to phenol ratio. Increasing nitrogen incorporation in carbon framework causes inhibiting effect on the formation of micropores during pyrolysis, resulting in

TABLE 1

|        | BET<br>surface<br>area | Total<br>pore<br>volume <sup>[a]</sup> | pore Micropore | Elemental<br>analysis (wt %) |      |     |         |  |
|--------|------------------------|--|----------------|------------------------------|------|-----|---------|--|
| Sample | (m <sup>2</sup> /g)    | (cc/g)                                 | (cc/g)         | С                            | N    | Н   | O (cal. |  |
| МС     | 750                    | 0.74                                   | 0.15           | 86.3                         | 0    | 1.3 | 12.4    |  |
| NMC-1  | 836                    | 1.08                                   | 0.12           | 80.4                         | 5.1  | 1.0 | 13.5    |  |
| NMC-2  | 878                    | 1.13                                   | 0.09           | 77.1                         | 8.2  | 0.8 | 13.9    |  |
| NMC-3  | 844                    | 1.09                                   | 0.07           | 73.0                         | 11.6 | 0.7 | 14.7    |  |

[a]Total pore volume at P/P<sub>0</sub> = 0.9.

[b]Calculated by T-plot method.

decreased micropore volume.

TABLE 2

| Catalyst                   | Ru<br>content <sup>[a]</sup><br>(wt %) | BET<br>surface<br>area<br>(m²/g) | Total<br>pore<br>volume <sup>[b]</sup><br>(cm <sup>3</sup> /g) | Average<br>Ru particle<br>size <sup>[c]</sup><br>(nm) | 60 |
|----------------------------|--|----------------------------------|--|---|----|
| wt % Ru-MC                 | 1.92                                   | 722                              | 0.71   | 6.1   |    |
| 2 wt % Ru-NMC-1            | 1.89                                   | 792                              | 1.04   | 3.1   |    |
| 2 wt % Ru-NMC-2            | 1.91                                   | 849                              | 1.10   | 2.5   |    |
| 2 wt % Ru-NMC-3            | 1.95                                   | 805                              | 1.05   | 1.9   |    |
| 2 wt % Ru-AC               | 1.81                                   | 991                              | 0.51   |   |    |
| 2 wt % Ru-CeO <sub>2</sub> | 1.76                                   | 105                              | 0.17   |   | 65 |
| 2 wt % Ru-MgO              | 1.82                                   | 91                               | 0.13   | MG.A  | -  |

The formation of graphitic ordered carbon was further proved by Raman spectroscopy (FIG. 1d). Raman spectra show characteristic D- and G-bands of disordered graphitic carbon at 1335 and 1590 cm-1 respectively. The D-band is a defects-induced Raman feature reflecting the non-perfect crystalline structure of the carbon, while G-band indicates the in-plane vibration of sp<sup>2</sup> carbon atoms. There is no dependence of the position of the two bands on the nitrogen content. The D-band appears to be stronger than G-band, suggesting amorphization of the graphitic carbon. The intensity ratios of the D-band to the G-band  $(I_D/I_G)$ , calculated from integral area of the peaks) are 1.6, 1.8, 2.1 and 2.3 for MC, NMC-1, NMC-2 and NMC-3, respectively. These results show that the defects in graphite-like layers enhanced with increasing nitrogen content. The mesoporous structure of the MC and nitrogen-doped carbon samples can be further confirmed by transmission electron microscopy (TEM) observations (FIG. 2). From TEM images, nitrogen containing samples (FIGS. 2b,2c and 2d) have no differences when 35 compared with their nitrogen-free counterpart (FIG. 2a), consisting of spherical mesopores with a disordered amorphous carbon structure. The Ru nanoparticle sizes as well as their distribution of various Ru containing catalysts were investigated by TEM (FIG. 3). It was found that Ru nan-40 oparticles were dispersed unevenly and large agglomerated Ru nanoparticles were detected on the surface of MC with an average particle size of 6.1 nm (FIG. 3a,b). On the other hand Ru nanoparticles were dispersed homogeneously over nitrogen-doped carbons with an average particle size of 3.1, 45 2.5 and 1.9 nm for 2 wt % Ru-NMC-1, 2 wt % Ru-NMC-2 and 2 wt % Ru-NMC-3 catalyst, respectively (FIG. 3c,3d, 3e,3f,3g and 3h). Thus, it can be concluded that the dominant contributor for stabilizing Ru nanoparticles is the nitrogen doped in the carbon structure and increasing nitrogen con-50 tent in the sample led to decrease in the size of Ru nanoparticles. X-ray photoelectron spectroscopy (XPS) was employed to investigate the nitrogen-carbon bonds formed in the carbon framework. In XPS only oxygen, carbon and nitrogen are detected in the overall region scanned (FIG. 4a). XPS of C 1s shows very similar peak at 284.6 eV, which is associated with the graphitic carbon, suggesting that most of the C atoms are assembled in conjugated honeycomb lattices. The N Is spectra (FIG. 4b) are curve-fitted into three peaks with the binding energies at 398.2, 400.4 and 401.8 eV that correspond to pyridinic N (N1), pyrrolic N (N2) and graphitic N (N3), respectively. The peaks related to nitrogen bonded to oxygen (404-408 eV) are absent. It is important to note that the distribution of these nitrogen species is very similar at same pyrolysis temperature (800° C.), being 65 independent of nitrogen content in the sample (FIG. 4c). As own in FIG. 4c, regardless of nitrogen content, a majority

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trogen atoms are localized at the edges of graphene

sheets (N1 and N2) whereas small amount of nitrogen atoms are situated at the central region of graphene sheet (N3).

# Example 5: Catalytic Activity in the Conversion of HMF to DMF

a) Effect of Reaction Temperature

Catalyst evaluation for hydrogenolysis of HMF to get DMF was performed using 2-propanol as a hydrogen donor in a batch reactor. FIG. 5 shows HMF conversion and product distributions as a function of reaction temperature over 2 wt % Ru-NMC-3 catalyst. The catalytic transfer hydrogenation (CTH) reaction occurs at a lower reaction temperature (80° C.) and DMF yield increased with increasing temperature. At lower temperature, the primary product was 2,5-bis(hydroxymethyl)furan (BHMF) and upon

increasing the temperature to 160° C., BHMF is completely converted to DMF with a yield up to 84 mol % at 100 mol % HMF conversion after 8 h of reaction (Scheme 2). Other products observed at 120 and 140° C. are hydrogenated furans: 5-methyl furfural (MFU), 5-methyl furfuryl alcohol (MFA), 2-methylfuran (MF) and furfuryl alcohol (FA). The formation of FA indicates the occurrence of decarbonylation reaction, however, the yield of FA was low (<4 mol %). The formation of MFU suggested that hydrogenolysis of —OH group in HMF occur in parallel to the hydrogenation of the —CHO group. In addition, ethers are also observed at lower temperature (80 and 120° C.), which are formed via etherification of BHMF or MFA and 2-propanol. They are not observed upon increasing the reaction temperature (160 and 170° C.).

Scheme 1 General scheme for the catalytic transfer hydrogenation of (CTH) reaction for preparation of DMF and MF

Scheme 2 Reaction network of the hydrogenolysis of HMF to DMF using 2-propanol and Ru --- NMC catalyst.

Compounds: 5-Hydroxymethylfurfural (HMF); 2,5-bis (hydroxymethyl)furan (BHMF); 5-methyl furfural (MFU); 5-methyl furfuryl alcohol (MFA); furfuryl alcohol (FA); 2,5-bis(hydroxymethyl)tetrahydrofuran 5-methyl tetrahydrofurfuryl alcohol (MTHFA); 2,5-dimethylfuran (DMF); 2,5-dimethyltetrahydrofuran (DMTHF); 2-methylfuran (MF).

b) Effect of Hydrogen Donors

The effect of hydrogen donor on HMF conversion and product yields was investigated at 160° C. over 2 wt % Ru-NMC-3. The results (FIG. 6) clearly indicate that the activity of catalyst heavily depends on the type of alcohol used. The HMF conversion increased from 40 to 100 mol % and the following reactivity order was observed: methanol < 1 - but a nol < e than ol < 1 - propanol < 2 - but a nol < 2 - propanol.Among the hydrogen donors tested, 2-propanol showed highest activity giving 84 mol % DMF yield. The formation of ethers was negligible in 2-propanol and 2-butanol but significant in other alcohols (2-14 mol %). This shows that etherification reaction favoured with lower chain alcohols 20 (methanol, ethanol and 1-propanol) indicative of steric effect. These findings are in good agreement with the reported results, which show that for CTH, aliphatic secondary alcohols show better activity than aliphatic primary alcohols. The higher tendency to release hydrogen from 25 secondary alcohols has been attributed to the highest reduction potential of corresponding dialkyl ketones

c) Effect of Nitrogen Content in the Catalyst on CTH In order to investigate the effect of nitrogen content of the catalyst on CTH activity, the Ru catalyst supported on 30 various nitrogen containing mesoporous carbons and nitrogen-free carbon were tested for HMF hydrogenolysis (Table

Ru nanoparticles (1.9 nm) that are surrounded by basic centers (nitrogen) provided by the support, which may favor ionic hydrogenation pathways and this is expected to be more active in hydrogenation reaction compared to nonionic hydrogenation pathways. Sanchez-Delgado et al. reported heterolytic splitting of H2 into H+ and H- (ionic hydrogenation pathway) over Ru nanoparticles, assisted by the basic pyridinic groups of the support (poly(4-vinylpyridine)). During CTH reaction H2 was produced and the H2 partial pressure in the closed reactor has increased with the increasing N content of the catalyst. These results show that the nitrogen plays a key role in the dehydrogenation of 2-propanol leading to higher H2 pressure. It is possible that adsorbed hydrogen species formed by the dehydrogenation of 2-propanol during CTH process can directly hydrogenate HMF molecule adsorbed on Ru nanoparticles instead of being released as H2. To understand this prospect, HMF hydrogenation was performed at H2 pressure of 7 bar (total pressure developed including H2 partial pressure during CTH reaction at 160° C. over 2 wt % Ru-NMC-3 with 2-propanol) using tetrahydrofuran (THF) as solvent. The HMF conversion and DMF yields were 86 and 59 mol %, respectively, which were inferior to those using 2-propanol as hydrogen donor (compare entry 5 and 7 in Table 3). This result strongly suggests that HMF hydrogenation using hydrogen donors can be more efficient than that using external H2 molecule. It is well known that there are differences between heterogeneous catalytic hydrogenation using hydrogen donor molecules as the source of hydrogen and hydrogenation using molecular H2. The CTH reaction could occur through direct hydride transfer from 2-propanol to

TABLE 3

|       |                | HMF           | Mark III | TOF[*] |     |       |           |                    |
|-------|----------------|---------------|----------|--------|-----|-------|-----------|--------------------|
| Entry | Catalyst       | conv. (mol %) | BHMF     | MFA    | DMF | DMTHF | Others[d] | (h-1)              |
| 1     |                | 8             | 0        | 0      | 0   | 0     | 8         |                    |
| 2[6]  | NMC-3          | 15            | 3        | 4      | 1   | 0     | 7         | THE REAL PROPERTY. |
| 3     | 2 wt %Ru-NMC-1 | 61            | 7        | 22     | 30  | 0     | 2         | 7.5                |
| 4     | 2 wt %Ru-NMC-2 | 79            | 5        | 14     | 55  | 2     | 3         | 13.7               |
| 5     | 2 wt %Ru-NMC-3 | 100           | 0        | 0      | 84  | 8     | 8         | 21.0               |
| 6     | 2 wt %Ru-MC    | 41            | 7        | 6      | 11  | 0     | 17        | 2.7                |
| 7[0]  | 2 wt %Ru-NMC-3 | 86            | 1        | 3      | 59  | 13    | 10        | 14.7               |

 $<sup>^{[\</sup>sigma]}Reaction$  conditions: molar ratio of HMF to Ru (200); temperature (160  $^{\circ}$  C.);

Piyaber

3). The HMF conversions were 41, 61, 79 and 100 mol % with DMF yield of 11, 30, 55 and 84 mol % on 2 wt % Ru-MC, 2 wt % Ru-NMC-1, 2 wt % Ru-NMC-2 and 2 wt % Ru-NMC-3 catalysts, respectively. Therefore, it can be concluded that the catalytic activity is improved with increasing nitrogen content in the catalyst. The high catalytic performance of 2 wt % Ru-NMC-3 in HMF hydrogenolysis has been attributed to the uniqueness of the support-Ru 60 interaction, i.e., N-doped mesoporous carbon-Ru heterojunction. The high nitrogen content in NMC-3 not only led to very stable and homogeneous dispersion of Ru nanoparticles but also enriches the electron density of the Ru nanoparticles, which accelerates the hydrogenolysis reac tion, as compared to nitrogen-free catalyst (2 wt % Ru-M Moreover, 2 wt % Ru-NMC-3 catalyst is composed of man RM have

d) CTH of HMF Over Different Catalysts

The catalyst 2 wt % Ru-NMC-3 was compared with 2 wt % Ru-AC under the same reaction conditions as well as with other oxide-supported Ru nanoparticles, including 2 wt % Ru-CeO<sub>2</sub>, 2 wt % Ru-MgO, 2 wt % Ru-TiO<sub>2</sub>, 2 wt % Ru-Mg(Al)O and 2 wt % Ru-γ-Al2O3. Catalyst 2 wt % Ru-NMC-3 showed remarkably high activity in CTH, giving 100 mol % HMF conversion and 84 mol % DMF yield (Table 4). Oxide and AC supported Ru nanoparticles were not effective for this reaction, affording DMF in lower yields than 2 wt % Ru-NMC-3 catalyst. In case of 2 wt % Ru-γ-Al<sub>2</sub>O<sub>3</sub> and 2 wt % Ru-TiO<sub>2</sub>, the main product was BHMF (entry 6 and 7, Table 4). These results show that wo catalysts have ability towards carbonyl group tion to hydroxyl group out have less ability for

N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (2-propanol, 25 mL); time (8 h). [6]25 mg catalyst.

<sup>[</sup>c]7 bar H<sub>2</sub> press

<sup>[6]</sup> To Br Hz pressure was used instead of 2-popanol and THF (25 mL) as solvent.

[6] It includes furfuryl alcohol (FA), 2-methylfuran (MF), ethers and some condens

[6] TOF = turnover frequency (moles of DMF produced per mole of Ru per hour).

C—O bond hydrogenolysis. From these results, one could conclude that the nature of the support plays an important role in the activity and selectivity of Ru based catalysts in CTH reactions.

intermediate products such as BHMF and MFA formed in significant quantities, which are converted to DMF with prolonging the reaction time. On further increasing H2 pressure (7 bar); the DMF yield reaches the maximum (68

TABLE 4

|       |                  | HMF           |      | TOF[e] |     |       |           |       |
|-------|------------------|---------------|------|--------|-----|-------|-----------|-------|
| Entry | Catalyst         | conv. (mol %) | BHMF | MFA    | DMF | DMTHF | Others[b] | (h-1) |
| 1     | 2 wt %Ru-NMC-3   | 100           | 0    | 0      | 84  | 8     | 8         | 21.0  |
| 2     | 2 wt %Ru-AC      | 56            | 7    | 7      | 15  | 0     | 27        | 3.7   |
| 3     | 2 wt %Ru-CeO2    | 39            | 10   | 9      | 6   | 0     | 14        | 1.5   |
| 4     | 2 wt %Ru-MgO     | 37            | 10   | 4      | 6   | Ö     | 17        | 1.5   |
| 5     | 2 wt %Ru-Mg(Al)O | 48            | 15   | 6      | 5   | 1     | 21        | 1.2   |
| 6     | 2 wt %Ru-TiO2    | 45            | 26   | 1      | 0   | 0     | 18        | 0     |
| 7     | 2 wt %Ru-y-Al2O1 | 60            | 35   | 2      | 0   | 0     | 23        | 0     |

[6]Reaction conditions: molar ratio of HMF to Ru (200); temperature (160 ° C.); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (2-propanol, 25 mL); time (8 h). [6]It includes ethers, FA, MF and some condensation compounds.

IctTOF = turnover frequency (moles of DMF produced per mole of Ru per hour).

e) CTH of HMF Over Different Metal Catalysts

The CTH of HMF was investigated using NMC-3 supported Ru, Pt, Pd, Rh, Ni, Cu and Au catalysts. The HMF 25 conversion varied in the range of 21 to 100 mol % and followed the reactivity order of Cu<Ni<Au<Rh<Pt<Pd=Ru (Table 5). Gratifyingly, an excellent DMF yield of 84 mol % with 100 mol % HMF conversion was obtained over Rubased catalyst (Table 5, entry 1). Interestingly, when the  $^{30}$  reaction was conducted with Pd catalyst, significant amount of decarbonylation product, FA was obtained (Table 5, entry 3), indicating Pd is more suitable for HMF decarbonylation.

mol %) and the yield decreased on continuation of the reaction for further duration. This decreased in DMF yield is due to its ring hydrogenation, leading to the formation of DMTHF. On rising H<sub>2</sub> pressure further to 10 and 15 bar, has an adverse effect on the DMF yield. At higher H2 pressure, DMF yield suppressed mostly due to the increased rate of consecutive ring hydrogenation of DMF, led to the formation of DMTHF in significant amounts. Moreover, the concentration of other undesirable by-products such as 5-methyl tetrahydrofurfuryl alcohol (MTHFA), 2,5-bis(hy-

| _     | and the second second | and the              | TATA MINING | ADLL   | 2           |               |          |                   |                    |
|-------|-----------------------|----------------------|-------------|--------|-------------|---------------|----------|-------------------|--------------------|
| 14    | Product di            | istributions         | s for CTH   | of HMF | over differ | rent metal ca | talysts. | [a]               | 1/4,00             |
| но    | HMF Cata              | lyst                 |             |        |             |               |          |                   |                    |
|       | но о                  | ∠OH -                | O<br>MFA    | ✓OH    | O FA        | OH A          | OMF      | DMTH              | Others             |
|       |                       | HMF<br>conv.<br>(mol |             |        | Product y   | ields (mol %  | )        |                   | TOF <sup>[e]</sup> |
| Entry | Catalyst              | %)                   | BHMF        | MFA    | DMF         | DMTHF         | FA       | Others[b]         | (h <sup>-1</sup> ) |
| 1     | 2 wt % Ru-NMC-3       | 100                  | 0           | 0      | 84          |               | 1000     | The second second |                    |
| 2     | 2 wt % Pt-NMC-3       | 75                   | 5           | 8      | 31          | 8 2           | 5        | 3                 | 21.0               |
| 3     | 2 wt % Pd-NMC-3       | 100                  | 0           | 5      | 15          | 0             | 62       | 28                | 7.7                |
| 4     | 2 wt % Rh-NMC-3       | 66                   | 25          | 15     | 11          | 4             | 62       | 18                | 3.7                |
| 5     | 2 wt % Au-NMC-3       | 40                   | 25          | 3      | 2           | 0             | 2        | 10                | 2.7                |
| 6     | 5 wt % Ni-NMC-3       | 25                   | 6           | 1      | 3           | 0             | 1        | 8                 | 0.5                |
| 7     | 5 wt % Cu-NMC-3       | 21                   | 3           | 3      | 2           | 0             | 2        | 11                | 0.7                |

[e]Reaction conditions: molar ratio of HMF to metal (200); temperature (160° C.); N<sub>2</sub> pressure (20 bar); solvent and hydrogen donor (2-propanol, 25 mL); time (8 h).
[6] It includes ethers, MF, 5-methyltetrahydrofurfuryl alcohol (MTHFA) and some condensation products.

[c)TOF = turnover frequency (moles of DMF produced per mole of metal per hour).

f) Catalytic Activity for the Conversion of HMF to DMF

The effect of H<sub>2</sub> pressure on the DMF yields was studied by varying the pressure from 2-15 bar at 160° C. in THF 65 solvent over 2 wt % Ru-NMC-3 catalyst (FIG. 7). When the

droxymethyl)tetrahydrofuran (BHMTHF) and hexanediol increased at higher H2 pressure.

g) Recyclability Study
The recyclability of the 2 wt % Ru-NMC-3 catalyst was reaction was carried out at low pressure (2 and 5 bar), the R (FIG. 8). The results show that the catalyst performance

remains same even after being reused for four times. These results indicate good stability of the catalyst. ICP-OES analysis showed no leaching of Ru after each recycle. Moreover, the amount of Ru in the catalyst after four cycles was found to be similar to that of starting catalyst.

#### Example 6: Catalytic Activity for the Conversion of Furfural to MF

a) Effect of Reaction Temperature

The effect of temperature on the furfural conversion and product yields over 2 wt % Ru-NMC-3 catalyst was investigated by varying the reaction temperature in the range of 50-140° C. (FIG. 9). The primary product of the transfer hydrogenation of furfural was FA, which is prominent at 15 lower temperature (80, 100 and 125° C.). In a consecutive step, hydrogenolysis of the -OH group in FA yields MF up to 87 mol % with 100 mol % furfural conversion at 140° C In parallel to hydrogenation, furfural also undergoes decarbonylation reaction to furan. Furthermore, furan-ring hydro- 20 genated products like 2-methyl tetrahydrofuran (MTHF) and tetrahydrofurfuryl alcohol (THFA) have also been observed.

b) Effect of Hydrogen Donors

The effect of hydrogen donor on furfural conversion and product yields was investigated at 140° C. over 2 wt % Ru-NMC-3 catalyst (FIG. 10). The results clearly suggested that the catalyst activity is greatly depends on the nature of hydrogen donor. The furfural conversion increased from 44 to 100 mol % and follows the reactivity order: methanol<1butanol<ethanol<1-propanol<2-butanol<2-propanol.

Among the hydrogen donors tested, 2-propanol displayed the highest activity leading to 87 mol % MF yield with 100 mol % furfural conversion. These results are in good agreement with the reported results, which normally reports that in CTH secondary alcohols have a better tendency to release 35 hydrogen than primary alcohols.

Advantages of invention:

Simple and environment friendly process

- 2. The novel transition metal catalyst, especially Ru based
- 3. This catalyst also exhibited good recyclability and can be reused many times without loss in activity.
- 4. The CTH process using the catalyst does not require molecular hydrogen as hydrogen source. It uses alcohol 45 organic acid, organic acid salts as hydrogen source.
- 5. The process does not require use of any co-catalysts.

We claim:

1. A catalyst comprising:

a transition metal supported on nitrogen doped mesoporous carbon,

wherein the transition metal(s) either alone or in combination is from 0.5 weight % to 5 weight % of the catalyst; and wherein average particle/crystal size of the transition metal 55 (s) is from 1 nm to 5 nm; and wherein the nitrogen content is from 8.2 weight % to 15 weight % of the catalyst.

2. The catalyst of claim 1, wherein the transition metal is selected from the group consisting of Ru, Pt, Pd, Rh, Au, Ag,

3. The catalyst of claim 1, wherein a BET surface area of the transition metal(s) catalyst is from 30 m<sup>2</sup>/g to 1200 m<sup>2</sup>/g.

4. The catalyst of claim 3, wherein the BET surface area of the transition metal(s) catalyst is from 36 m<sup>2</sup>/g to 1200

m<sup>2</sup>/g.

5. The catalyst of claim 1, wherein total pore volume of the catalyst of claim 1, wherein total pore volume of the catalyst is from 0.07 cm<sup>3</sup>/g to 1.2 the transition metal(s) catalyst is from 0.07 cm<sup>3</sup>/g to 1.2 cm3/g

6. A process for synthesis of the catalyst according to claim 1, the process comprising:

(a) dispersing nitrogen doped mesoporous carbon in deionized water under ultrasonication to obtain a dis-

(b) charging transition metal chloride to the dispersion obtained in (a) under ultrasonication and stirring the dispersion at 70° C. to 90° C. for 6 hours to 12 hours to obtain a mixture;

(c) cooling the mixture of (b) and charging aqueous solution of NaBH4 under ultrasonication and stirring for 30 minutes to 60 minutes to obtain a catalyst mixture: and

(d) filtering the catalyst mixture obtained in (c) and washed with deionized water and dried at 80° C. to 100° C. for 10 hours to 12 hours to obtain the catalyst.

7. The process of claim 6, wherein the transition metal chloride is selected from the group consisting of chlorides of Ru, chlorides of Pt, chlorides of Pd, chlorides of Rh, chlorides of Au, chlorides of Ag, chlorides of Os, chlorides of Ir, chlorides of Cu, chlorides of Ni, chlorides of Re, chlorides of Cr, chlorides of Mn, chlorides of Fe, chlorides of Zn, chlorides of Co, and combinations of any two or more of the transition metal chlorides.

8. A process for the synthesis of 2,5-Dimethylfuran (DMF) or 2-Methylfuran (MF) using the catalyst according

to claim 1, the process comprising:

stirring 5-hydroxymethylfurfural (HMF) and the catalyst, or furfural and the catalyst, in the presence of a catalytic transfer hydrogenation (CTH) agent at a temperature from 80° C. to 170° C. for a period 0.5 hours to 10 hours, to convert the HMF to DMF or to convert the furfural to MF.

catalyst exhibits excellent activity for the CTH of bio- 40 selected from the group consisting of alcohols, organic acids, and mixtures thereof.

10. The process of claim 9, wherein the alcohol is selected from the group consisting of 2-propanol, 2-butanol, methanol, ethanol, and 1-propanol.

11. The process of claim 9, wherein organic acid is selected from the group consisting formic acid, acetic acid, propionic acid, and butyric acid.

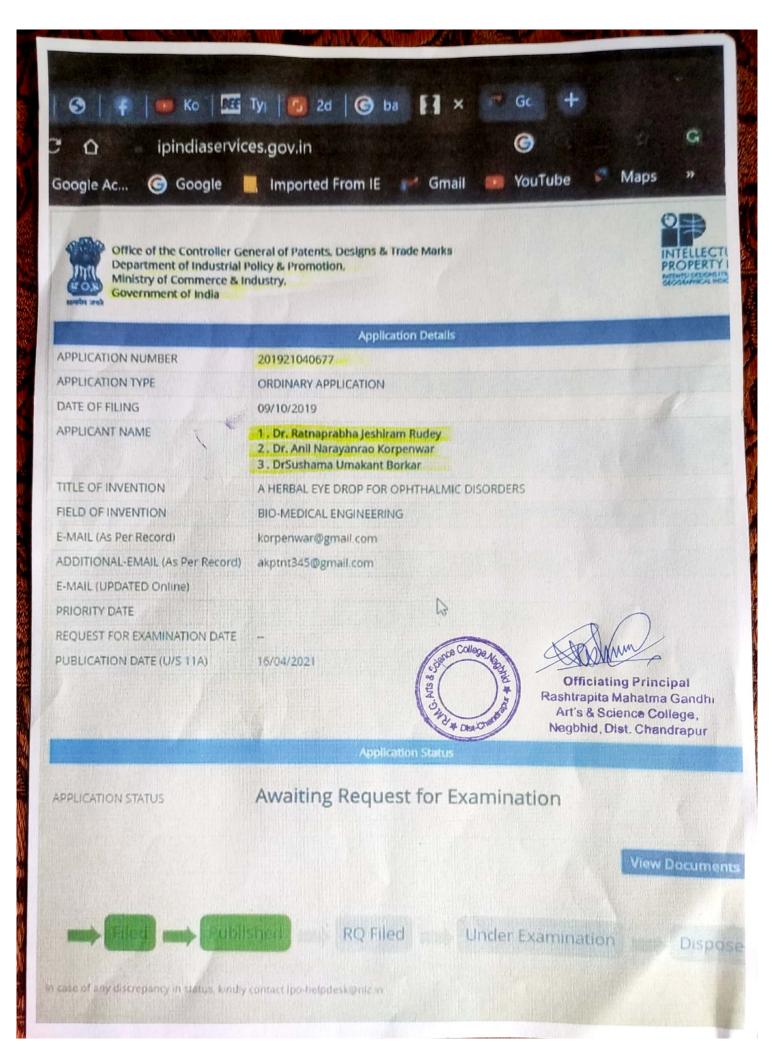
12. The process of 8, wherein said process is carried out in the presence of molecular hydrogen and the catalytic transfer hydrogenation agent.

13. The process of claim 8, wherein HMF and the catalyst are stirred in the presence of the CTH agent and the HMF is converted to DMF; wherein a yield of DMF is greater than 81%; and wherein a conversion of HMF is 100%.

14. The process of claim 9, wherein transition metal(s) content either alone or in combination in said catalyst is from 1 weight % to 5 weight %.

15. The catalyst of claim 1, wherein a BET surface area Os, Ir, Cu, Ni, Re, Cr, Mn, Fe, Zn, Co, and combinations of 60 m<sup>2</sup>/g, and wherein total pore volume of the transition metals of m<sup>3</sup>/g, and wherein total pore volume of the transition metal(s) catalyst is from 1.05 cm<sup>3</sup>/g to 1.2 cm<sup>3</sup>/g.







Manoharbhai Shikshan Prasarak Mandal, Armori's

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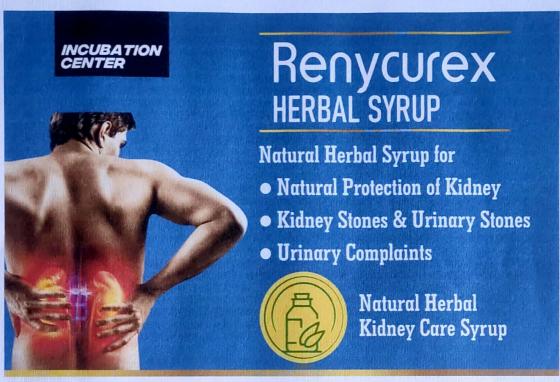
Email: - gdnagbhir72@gmail.com

nagbhid.rmgcollege@gmail.com

- 3.2.1 Institution has created an ecosystem for innovations, Indian Knowledge System (IKS), including awareness about IPR, establishment of IPR cell, Incubation centre and other initiatives for the creation and transfer of knowledge/technology and the outcomes of the same are evident
- Following are the Products of "Incubation Center" Launched in the Market. All these products received approval from Food and Drug Administration (FDA), Maharashtra.
- 1. Renycurex Herbal Syrup
- 2. Bhishmika Herbal Oil
- 3. Lyssomen Herbal Health Capsule
- 4. Inanna Herbal Oil
- 5. Shatawari Kalp Herbal Health Tonic
- 6. Herb-Boon Herbal Powder

Salar College Version Res

# 1. Renycurex Herbal Syrup



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# 2. Bhishmika Herbal Oil

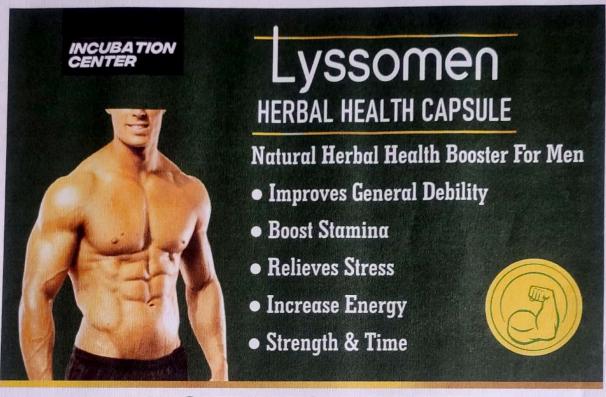


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# 3. Lyssomen Herbal Health Capsule



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### 4. Inanna Herbal Oil



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# 5. Shatawari Kalp Herbal Health Tonic

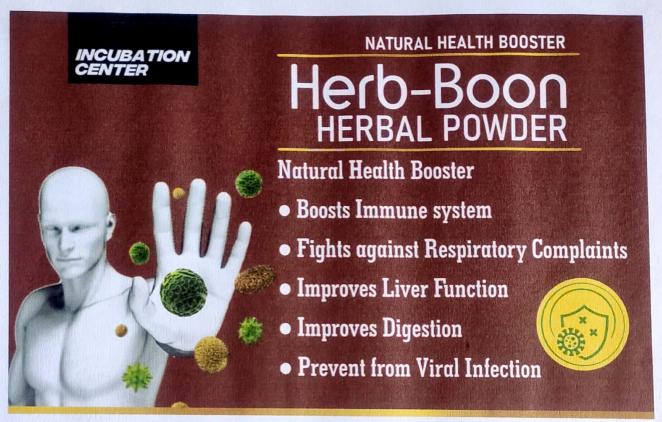


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### 6. Herb-Boon Herbal Powder



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### MFG Product Profile

### Food & Drugs Administration MH

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| MEDICARI ASTHACEPT CAPSULES  | ayu prop med!  odication-doce, as per physician  interical Seed. 70 mg. 2. Buch(Acoria, cahamus) Rhizomes 60 mg. 3. Lata Kastori (Hibiscus, Abelmoschus) Seed 50 mg. 4. arkii  ki mar. ko. 166 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 3. Lata Kastori (Hibiscus, Abelmoschus) Seed 50 mg. 4. arkii  ki mar. ko. 166 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 3. Lata Kastori (Hibiscus, Abelmoschus) Seed 50 mg. 4. arkii  ki mar. ko. 167 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 3. Lata Kastori (Hibiscus, Abelmoschus) Seed 50 mg. 4. arkii  ki mar. ko. 167 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 3. Lata Kastori (Hibiscus, Abelmoschus) Seed 50 mg. 4. arkii  ki mar. ko. 167 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 4. arkii  ki mar. ko. 167 (167). 2. Buch (Acoria, cahamus) Rhizomes 60 mg. 4. arkii  ki mar. ko. 167 (167). 2. Buch (167). 2. arkii ki mg. 4. arkii mg. 4.       | Ayu Ph-P&P                               | Each capsule contains extracts   |
| CAP-AYU-DOM  | AVU PROP MED   | Avu Ph-P&P                               |  |
| 1.Bhuimala(Phyllanthus niruri)   | ICA HOBELIVER TONIC , DOSES AS PER PHYSICIAN Parchane 70 mg 3. Kurk (Procedure Form) Philosophysics (1997)   |  | Fach capsule contains extracts   |
| CAP-AYU-DOM  | I) Khursani ajwän(Hyosymus niger) seed-30mg, 2) Kantakary(Solanum vanthocarpum) root-50mg, 3) Chotta gokharut Tibulas terrestrisifroit-75mg, 4) Chirchirat/Achyraathes aspera)root-50mg, 5) Wekhandt Agruss adjungsterrestrisifroit-75mg, 4) Chirchirat/Achyraathes aspera)root-50mg, 5)   | Ayu Ph-P&P                               | EACH CAPSULE CONTAINS<br>EXTRACTS OF   |
| 1) Kinurani anvaini Hvoveymus  | rakish pari t*Indicanonos per physician Mig processitivas, capatel filling and packing<br>niger) seed 30mg, 2) Kantakary(Solomini xanthocarpum) roos 30mg, 3) Choita gokharu(Tribulas terrestris)/ruit-75mg, 4<br>3mg, 6) Manishhan/Rubia cardiglofathocar-75mg, 4ya Ph (6) units)   | ) Chirchirat Achyra                      | nthes aspera)root-30mg, 5)   |
| OIL - AYU - DOM /  | 1)Sarxo oil(Brasica cumpestris)Seed-95ml. 2)Badam oil(Prunus amygdalus)seed-1ml. 3)Salai guggle<br>(Bowelli serrata)Gum-1gm, 4)Kapoor (Camphora)-1gm, 5)Menthol-1gm, 6)Ajwain ke phool(Thymol)-<br>fom   | Ayu Ph-P&P                               | Each 100ml contains ext of   |
|  | spiakash part Pandication-external massage on Manufacturing process; Ingredient in 3 udded masso oil borled till it doobted cooling add ingresses Seed-95ml, 21Radam oil Primus amygdalus (seed-1ml. 3)Salai gingglet Boxwelli servata/Gum-1gm. 41Kapoor (Camphora cumpesaris) Seed-95ml, 21Badam oil Primus amygdalus (seed-1ml. 3)Salai gingglet Boxwelli servata/Gum-1gm.<br>Ava Ph 1100 ml)  |  |  |
| ORL - AYU - DOM VEDCARE WHITOCEPT SYRUP UT   | ayu prop med// PERINC TONIC deserts no physician   | Ayu Ph-P&P                               | each 100 ML contains   |
|  | i <mark>jerák</mark> Roni 10 gm 2. Kuts (Dotichos byllorus (Seed 2 gm 3. Chandrasur(Lepulum Sativum) Seed 5 gm 4. Kumihha (Careya<br><mark>ibica) Bark [</mark> 11 gm 4yu Ph (100 ml)  | urboreatBack 3 ga                        | 3. Neemt (zardirachita indica)   |
| OTH - AYU-DOM  | ayu prop med  EMACE HEALTH TONIC  SOUR ROUTE (Home 2 Flowers (Flowers)   | Ayu Ph-P&P                               | each 100 gm contains   |
| AVANA SYRUP INDICATION:TOM   | osus) koots (0 gm 2. Ehrycht (Elatoria cardamonum) Fruits 1 gm 3 Sugar (Saccharum officinarum) (10) gm 4xu Ph<br>AYU PROP MED<br>C TOSES AS PH PMYSICIAN<br>Root 15 gm 2 Cinnamon(Dulchim) Cinamom zeylamum-Bark 2gm 3 Black Papper (Kall Mirch) Piper nigram-Seed Jem   | Avu Ph-P&P                               | each 100 ml cantains   |
| EPL - AYU - DOM  | AYU PIROP MED/FOR EXT USE  |  | h)Zyzimber officianalis-Rhizmucs   |
| I Mayour daithers culinaries see   | ation-SKIN CARE AS PER PHYSICIAN   | Ayu Ph-P&P                               | Japhal (Myristica Fragransilra)  |
| (Meristica Fragranstruit 2.5 gm  | Avu Ph. t. GM1   | Ayu Ph-P&P                               | sinensisi citali 5 gm 5 Japhal  EACH CAPSULE CONTAINS  |
| BUT CAPSULE Author-bleavpeal 2017  | 1) Aairan(Premna integrifolia) bar Suyes 3) Somatha (Oroxylum indicum) Roor-Suyes 3) Kiji Maan<br>(Uraria picta) Root-75mg. 4) Indiada mixili 182 principia antiquorum)-75mg. 5) Kirgunatti Gerx<br>(Uraria picta) Root-75mg. 6) History Protesta Section 2008, 75mg [RUMATOL CAPOSTACIALING Principles of Principles and Principles of Pr |  | EXTRACTS OF  |
| 1) Braft Legic marnetosi Fruit-9.<br>8thng, 6) Muradi Helicteres i soraj   | Union 3. Ward Figure homest Late 2 7 70  | a Gandhi                                 | Trigonella fremungi accumisecul-   |
|  | Neghbid Dist Cha   |  | 121 100  |
| Print Date: 04/10/2021, Data   | Counts: (0 - 0)  | SACUOA                                   | A STATE OF THE STA |

### MFG Product Profile

|  | 1)Nishoth(Opercolina terpethum)Root-30gm. 2)Triphala(amla.bchda.harda)Fruit-10gm, 3)Senna<br>(Cassia angustiolia)Lcaf-30gm,4)Ajwain(Carum copticum )Seed-10gm, 5)Esapgot(Plantago<br>ovara)Hask-20gm   | Ayu Ph-P&P  | Lach Hogin contains ext o  |
|--|--|---|--|
| DNishothi Operedina terpethu   | ovara/Hask-20gm, s)Exapgot(Plantago meta) in the processing and packing in the advantage of the processing and packing in Rona-14gm, 2/Traphalatamla behda harda/Friit-10gm, 3/Senna (Cavsia angustitolian)-eaf-30gm, 4/4/waimCars.  KOSHIN CAPSULE  |   |  |
| 20gm Ayu Ph (50 GM)<br>CAP - AYU - DOM   | KOSHIN CAPSULE   |   | )Esapgoli Plantago ovata) Hask-  |
| Mig process mixing capsule filling p<br>to GueliceleTenesporte curclelet   | socking, Indication, Doses-Asper physician<br>at Pariching 50mg, 21 Goldwert February  | Ayu Ph-P&P  | LACH CAPSULE CONTAIN<br>EXTRACTS OF  |
| other) Fruit-90mg Ayu Ph 1345 m<br>ORL - AYU - DOM   | a\u prop med   | graftEclipta alba)Panchan   | g-80mg, 5) Vidang(Emhelia  |
| DECARE SYRUP For IMMUNITY  I Basile Fulsin Osimum sanic tion-  | '.TONIC  | Ayu Ph-P&P  | each 100 ML contain  |
| gm 5. Raisin(Manuka) Friis ven   | - 10800<br>Rou 13 gm 2 Cinnamam(DalchimiCinamon zeylanlam-Bark 2gm 3 Black Fapper(Kali Mirch) Piper nigram-Sea<br>glera-Fran 5 gm 6 Lemon Jince(Vimbm/Citrus meylica 4run 1 ml 18a Ph 1100 ml)<br>AVU PROP MED   | ed 2 gm 4 Dry Ginger (Sunt)   | NZvzimber officianalis-Rhizomes  |
| VANA WHITOCARE SVEID IN DU   | CATION IN AND AND AND AND AND AND AND AND AND AN   |   | The state of the s |
| ark 10 gm 6. Bahoolf Acacia are  | obica)Buck (Ugin 4 vi Ph.) - g.L.)  Antica)Buck (Ugin 4 vi Ph.) - g.L.)  | Careva arborea)Bark 5 gm  | 5. Neem(Azardirachita indica)  |
| TO HATE PACK INDICATION LAND   | ALL THE TOTAL CONTROL OF THE PARTY OF THE PA   |   |  |
| hozome 12 gm Avu Ph (- GM)   | CONDEDONS AS PER PHYSICIAN Edit get Q8 to 100 GM 2 Necami Azadirachita indica) leaf 25 gm 3. Brahmit Centila Asiatica) leaf 30 gm 4. Amlat AND Brah mod Get as a constant and    | Embilea Offinalis) From 15  | gni 5.Piyaz(Allium Opu)  |
| DUADE HAID DACKSDANTE, CO.   | propried for extuse  |   |  |
| hozome 12 gm Ayu Ph (Total 1)<br>DIL - AYU - DOM   |  | Embilea Offinalisi Friat 15   | gm 5. PiyaziAllium cepu)   |
| CMATOLOH Authorsthaypustali y  | 1)Neem oil(Azadirchita indica)-20ml, 2)Sarso oil(Mustard)-79ml, 3)Kala jeera(Carumcarvi)Seed-5gm., - 4)Kapoor-1gm, 5)Gungle (Commifera muku)(Gum-5gm, 6)Salai guggle(Bowellia errata)Gum-7gm, - Nirquodi (Vitex nirguodo) Leaf-10gm, 8)Karanjil(Pongamia glabra)seed-5gm and fundamentskand manays of Mandataunog process woldoned valde.  | Ayu Ph-P&P  | Each 100ml contains ext of   |
| Nagunda lites nargundar Leaf-  | 10gm, 8iKaranji(Pongamia glabraiseed-5em Avu Ph (100 ml)   | kul Gum-Sgm. 6) Solar gingg   | letBoswellia erratarGum-7gm.   |
| EDCARE LITHEEPT CVD1 ID / LITER  | NE TALL  |   |  |
| RO - AYU - DOM   | inte (1901) does at per physician ancepanching Root 10 gm 3. Gokhara(Tribulus (crrestris) Fruit 15 gm 4. Ptal Garud ban(Onosma Bracteatum) Leaf 5 gm, Ara Ptal Garud as a prop med Aorest use.  [35] A prop med Aorest use.  | li(Cocculus hirsutus) Root 3  | gm 5. Arkapushpi(Holostemma  |
| HNHMIKA Pain Oil / AK BUB BURS   | SICIAN   |   | each 100 ML contain  |
| 10 - AYU - DOM   | AYU PROP MED  MMUNITY TONIC DOSES AS PER PHYSICIAN  AND TONIC DOSES AS PER PHYSICIAN   | (100 ml)  |  |
| gm 5. RaismtManuka)Vitis ver   | Root 11 gm 2 Cinnamom(Dalehini)Cinamom zeylanium-Bark 1 gm 3 Black Papper(Kali Mirch) Piner (Seram-See   | d Fom 4 Dry Ginner Smith  | cach 100 ml eaman  |
|  |  |   |  |
|  | il manjishtatrubia cordifolia iroot40mg, lodrat symbolocos vacem <b>osa) bark40mg, triphala-fruit20ms</b> , brungara ityefi<br>osaitlower100mg, iatamagettasi barkasi kan  | ota albatleaves 160ma saem  | each 100 ml cantair  |
| CONCAPSULE / Said Late   | Vayuvidang(Emblica ribes)Fruit-70mg, 4) Nagkeshur(Mesua ferrea)-60mg, 5) Kale mire(Piper<br>nigram)Seed-50mg, 6) Sarpunkha Tenhrosia nurnuren)Deach  | Ayu Ph-P&P  | FACIL CAPSULE CONTAIN  |
| BIRINGRAJ(Eclipla alba)P<br>ing. 6) Sarpunkha(Tephrosia)   | anchange-70mg, 2) Triphala(Amla, Beheda, Harda; 130mg, 3) Valyavidang(Emblica ribes)Fruit-70mg, 4) Navle da.<br>purpurea; Root-60mg Ava Ph. (60 million da. 11)  | artMusius German 60 53  | r 1  |
| YP - AYU - DOM   | 1)Bach (Acorus calamus)Rhizome-3gm, 2) Adulsa(Adhatoda vashka)Leaf-10gm, 3)tulsi(Osimum<br>sanctum)Panchang-1gm, 4)Haldi(Curcuma lunga)Rhizomes-2gm, 5)Sounth(zizimber<br>officinalis)Rhizomes-1gm, 6)K antak-reference  | Avu Ph-P&P  |  |
|  | officinalis)Rhizomes-loss (31' ) and dangaryRhizomes-zgm, 5)Sounth(zizimber  |   | Fach 100ml contains ext o  |
| AP-AVII-DOM  | ip part l'indications ough symp 'Montacuming process fingenient no le 10 et agrafient negliter aux symp base<br>me-dym, 3. Adultact (Adultache visagan) et 10 pm, 3 finds (Ostman sanctum)P anchang-lym, 4 finds (ivection a lung visagan) et 10 pm, 3 finds (Ostman sanctum)P anchang-lym, 4 finds (ivection a lung partie) et 10 pm, 5 finds (ivection a lung partie) et 10 pm, 10 pm | suger is added and syrup base is<br>u)Rhizomes-2gm, 5)Sounth  |  |
| AP-AYU-DOM   | npart f-indication-cough symp Manufacturing prosesses Ingredient to the 16 sea referent medication and extract made by adding water, to this mis-3gm, 3.) Adults at Iddinated a vasaka) East-10gm, 3)tals it Ostoniam, sanction Peachang, 1 gm, 4)Haldit Curviona lunguage and propriet and extract made by adding water, to this incarption Peachang, 1 gm, Sugar syrup base, 4 vii Ph (100 ml).  3) up rop med   |   | ptepared<br>zizimber officinalis/Rhizomes-   |
| AP - AYU - DOM  DEARE STONOGEPT CAPSULES  Of Klursum aim ann Hyoseymus  ekhandt Acorus calamus troot-2  RO - AYU - DOM   | ip part l'indications ough symp 'Montacuming process fingenient no le 10 et agrafient negliter aux symp base<br>me-dym, 3. Adultact (Adultache visagan) et 10 pm, 3 finds (Ostman sanctum)P anchang-lym, 4 finds (ivection a lung visagan) et 10 pm, 3 finds (Ostman sanctum)P anchang-lym, 4 finds (ivection a lung partie) et 10 pm, 5 finds (ivection a lung partie) et 10 pm, 10 pm |   | ptepared<br>zizimber officinalis/Rhizomes-   |
| AP - AYU - DOM DOCARL STONOCEPT CAPSULES J Kluesum aja amt II voseymus ekhandi storus calamus proot-2 RO - AYU - DOM ANNA HAIR OIL hair oil  | ip part I indications outdo symp Manufacturing process ingredient to a large transport medical purposes in the medical purpose in the med   |   | prepared<br>zizimber officinalis/Rhizomess-<br>Each capsule contain-<br>thes aspeca/root-5(lmg, 5)   |
| AP - AYU - DOM DCARL STONOCEPT CAPSULES I Khursum aframit Hyoscymus khandi Acorus calamus iroot - 2 RO - AYU - DOM ANNA HAIR Oll hair ail PANNA HAIR ON - AYU - DOM ANNA HAIR ON - AYU - DOM ANA MERSON - B  | near I malestions ough symp Manufacturing process ingredient to a let one grident degetier and extract made by adding water, to this incurption Planchang - I gm. Sugar syrup base incurption Planchang - I gm. 4) Haldit Curetuma lungo appropriate properties of the state of the st   | Ayu Ph-P&P    Sing 4) Chirchirat Achyram   Ayu Ph-P&P   | prepared attimber officinalis/Rhizomess Each capsule contain thes aspeca/root-50mg, 5) each 100 ML contain   |
| AP - AYU - DOM DORAL STONOGERT C APSULES  I Klursum ajs ann Hyvseymus khandelsoms calamis iroot-2  Klo - AYU - DOM ANNA HAIR Oll. bair all PL - AYU - DOM ANNA HAIR Oll. bair all CANNA HAIR Oll. ba | in part I indication cough symp Manufacturing process ingredient to a life on a picket neglitis and symp same and in most pump lanchang I gm. Sugar symp base ingredient to this incarpiant Panchang I gm. 3) Adulast Aduladed vasaka) Leal- (10gm. 3) Indist(Oximum sanctim) Panchang I gm. 4) Haldit Cureuma lungs app prop med indication disease per physician indication disease per physician inger is seed-3 lung. 2) Kantikasy (Solanum xanthocurpum) root-50mg. 3) Chotta gokhurut Tribulas terrestris) puit-7 sing. 6) Manufathan Cardifoliai Root-75mg. 4 vu Ph. (t Total 360 mg) app prop med/for ext use.  AVU PROP MED/FOR EXT USE  Dearthon FOR FACE/EXTERNAL APPLICATION), DOSES AS PER PHYSICIAN  Darnhaldit Behreites species Root sign. 3. Roktachandam (Pterocarpus xantalinus) Bark I gm. 4. Manijisthut Rubia codinus or of a gm. Avu Ph. (CM).  | Ayu Ph-P&P    Sing 4) Chirchirat Achyram   Ayu Ph-P&P   | prepared attimber officinalis/Rhizomess Each capsule contain thes aspeca/root-50mg, 5) each 100 ML contain   |
| AP - AYU - DOM  DORAR ISTOROGEPT CAPSULES  J Khursam sija aanti Joseymus  sekhanid-korus calannesiroot 2  RO - AYU - DOM  ANNA HAR Oli, bair ait  PL - AYU - DOM  AMANGERIA - Proce pack INI  Masurifishus lens Joul 7gm 2  n 6. Ananimonithemideenus in  PL - AYU - DOM  RO - AND - HARD - Process of the second of t | in part i malestimesoude symp Manufacturing process ingredient to a regiment despite and extract made by adding water to this meeting. A dult said (dilutated a vasaka) Leal-(10pm, 3) information to a regiment despetier and extract made by adding water to this incarption) Panchang-Lym, Sugar syrup base. Live Ph (100 ml)  3) up rop med  and prop med  finitiation discrete per physican ingers seed-3/fing, 2) Kantikugy(Solanum xanthocurption) root-5/fing, 3) Chotta gokharut Tribulas (cryestris)) uit-7  ayu prop med/for ext use.  Sug. 6) Manyishan(Rubia cardifolia)Root-75/mg. A v. Ph. (t. Total \$60 mg)  ayu prop med/for ext use.  AND   | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P   | prepared zizinher officinalis/Rhizonies- Each capsule contain- hex aspera/root-50mg, 5) cach 100 ML contain- cach 100 ML contain-  |
| AP - AYU - DOM  DORAL STONGEEPT CAPSULES  J Riursant cija canti jos citymis  sekhandi Acorus calannis iroot - 2  RO - AYU - DOM  ANNA HAR Oli, bair ail  PL - AYU - DOM  ANNA MISHA LEP 1000 pack INI  Masuri Erhan lens Joul 7gm 2  n 6. Anantmont Hemideenus in  PL - AYU - DOM  RO - AYU - DOM  RO - AND  | in part i "indication-cough symp" Manufacturing process- ingredient to a life of an inchest depetier and extrace made by adding water, to this incarption/Panchang-1gm, 3/4 dulas deviated vasaka) Leal- (10gm, 3/14/s) (Ostmann sanction/Panchang-1gm, 4/Haldit Curcuma lunge appropriate language agent Sugar syrup base Avi Ph (100 ml).  3/4 Prop med Indication desease not physician indication desease not physician in indication desease of indication and indication devolution in indication i   | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P   | prepared zizinher officinalis/Rhizomes- Each capsule contain- hes aspeca/root-5fling, 3) cach 100 ML contains cach Yoli gui contains activities a contains cach Yoli gui contains  |
| AP - AYU - DOM DORAL STONGEEPT CAPSULES J Khursam aja aintilyoseymias khursam aja aintilyoseymias RO-AYU - DOM ANNAHAR OIL - har ail PL - AYU - DOM ANNAHAR OIL - har ail LANA MUKHA LEP rice pack INE MasurtErbian lensyDul 7gm 2 n 6. rhantmonliHemidesmus in IC WOUND CARE LEPAM CREA I Vall (Fieue Benghaltonsis) rous sistumadhiagh cyyrlina gllabra VP - AYU - DOM   | in part i mideather-coule symp Manufacturing process ingredient to Let are given the gettier and evity base in early in a let a significant english by adding water to this incarption Panchang I gm. 4) Halahi Curcuma lunge as you prop med indication descare near the growth base. Average in the part of the land of the growth of the gr   | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Ayu Ph-P&P  ayu Ph-P&P  ayu Ph-P&P  Ayu Ph-P&P  tun) sont 5 gm 5 Vetas(Sali  | Fach capsule contains  Each capsule contains  hes aspeca/root-50mg, 5)  each 100 ML contains  cach 100 ML contains  (Citras reticulata)Eruit chal 7  each 100 gnt contains  (capresty bark 8 gm 6.  Each 100ml contains ext o  |
| AP - AYU - DOM  DORAL STONOGEPT CAPSULES  J Kharsam aja aintHyoseymias  kharsam aja aintHyoseymias  RO - AYU - DOM  ANNA HAR OIL - bar ail  PL - AYU - DOM  ANNA MUKHA LAP rice pack INE  MasurtFrium lens/Dul 7gm 2  in 6. shantmonHemidesmus in  IC WOUND CARE LEPAM CREA  Yalt frees Benghaltonsis pan  salimmadhigh cyyrlica gllabra  VP - AYU - DOM  CON NYBUR Alaba (khar)   | in part i mideather-coule symp Manufacturing process ingredient to Let are given the gettier and evity base in early in a let a significant english by adding water to this incarption Panchang I gm. 4) Halahi Curcuma lunge as you prop med indication descare near the growth base. Average in the part of the land of the growth of the gr   | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Ayu Ph-P&P  ayu Ph-P&P  ayu Ph-P&P  Ayu Ph-P&P  tun) sont 5 gm 5 Vetas(Sali  | Fach capsule contains  Each capsule contains  hes aspeca/root-50mg, 5)  each 100 ML contains  cach 100 ML contains  (Citras reticulata)Eruit chal 7  each 100 gnt contains  (capresty bark 8 gm 6.  Each 100ml contains ext o  |
| AP - AYU - DOM DORAL STONGEEPT CAPSULES J Khursum aja aint Hyoseymus sekhandt-korus calaimstroot - 2 RO - AYU - DOM ANNA HAR OIL hair ait PL - AYU - DOM ANNA HAR ALD Prisc puck INI Masureff-than lens/Dal 7gm 2 n 6. Anautmont/Hemideenins ii PL - AYU - DOM IC WOODD CARE LEPAM CREA YAI (Facile Benghalansis) roo sekhimadhinghyeyrtiira gilahea YP - AYU - DOM CON SYREP Author/sharprakiis Bhringenit/Eclipta atbar/Panch diplotlar Panchange-Sym xuger AP - AYU - DOM   | in an I midication-coulds symp Manufacturing process ingredient to I to 6 are pites the gettine and the starting water to this micropium) Panchang-Igm. Sugar syrup base incarption Panchang-Igm. Manufacturing water to this incarption Panchang-Igm. Maldid Curciuma language as proposed in the proposed of the proposed in the propose   | Ayu Ph-P&P  Sing A) Chirchirat Achyrant  Ayu Ph-P&P  Ayu Ph-P&P  Ayu Ph-P&P  ayu Ph-P&P  itin) cont 5 gm 5 Yetis (Salt  Ayu Ph-P&P  this suger is added and syrup baserultat Swertia charattan Back   | prepared Each capsule contain- Each capsule contain- lies aspeca/root-50mg, 5)  each 100 ML contain- cach 100 ML contain- (Cirras reticulata/Fruit chal 7  cach 100 gru contain- caproses bark 5 gm 6.  Each 100ml contains ext of the prepared (7.5 gm, 5) Guduchi (Trinosporu)   |
| AP - AYU - DOM DORAL STONGEEPT CAPSULES J Khursum aja aint Hyoseymus sekhandt-korus calaimstroot - 2 RO - AYU - DOM ANNA HAR OIL hair ait PL - AYU - DOM ANNA HAR ALD Prisc puck INI Masureff-than lens/Dal 7gm 2 n 6. Anautmont/Hemideenins ii PL - AYU - DOM IC WOODD CARE LEPAM CREA YAI (Facile Benghalansis) roo sekhimadhinghyeyrtiira gilahea YP - AYU - DOM CON SYREP Author/sharprakiis Bhringenit/Eclipta atbar/Panch diplotlar Panchange-Sym xuger AP - AYU - DOM   | in an I midication-coulds symp Manufacturing process ingredient to I to 6 are pites the gettine and the starting water to this micropium) Panchang-Igm. Sugar syrup base incarption Panchang-Igm. Manufacturing water to this incarption Panchang-Igm. Maldid Curciuma language as proposed in the proposed of the proposed in the propose   | Ayu Ph-P&P  Sing A) Chirchirat Achyrant  Ayu Ph-P&P  Ayu Ph-P&P  Ayu Ph-P&P  ayu Ph-P&P  itin) cont 5 gm 5 Yetis (Salt  Ayu Ph-P&P  this suger is added and syrup baserultat Swertia charattan Back   | prepared Each capsule contain- Each capsule contain- lies aspeca/root-50mg, 5)  each 100 ML contain- cach 100 ML contain- (Cirras reticulata/Fruit chal 7  cach 100 gru contain- caproses bark 5 gm 6.  Each 100ml contains ext of the prepared (7.5 gm, 5) Guduchi (Trinosporu)   |
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| AP - AYU - DOM  DORAL STONGEEPT CAPSULES  J Khursam aja aint Hyuseymius  sekhandistoorus calaamsi root - 2  RO - AYU - DOM  ANNA HAR OIL - bar ail  PL - AYU - DOM  ANNA HAR AL IP rice puck INE  Masurt Erbun fensi Dul Tyen 2  a 6. diantimont Hemidesmus ii  PL - AYU - DOM  IC WOUND CARE LEPAM CREA  Tall ficus Benghalensis jon  CON SYRUP Authoristian yeak  AU - DOM  CON SYRUP Authoristian yeak  AU - DOM  CON SYRUP Authoristian yeak  AU - DOM  MICON CAPSULE I leath unic,  J augrantimat (1 phus rounding  MICON CAPSULE I leath unic,  J augrantimat (1 phus rounding  J authoristian yeak  J D Gokkon uf Tribullus  J authoristian yeak  J D J ayury Phy - AYU - DOM  ANA LAXATIVE POWDER CHIL  Terphalat Aimla Behuda, Harda  J D J ayu yyy Ph 10 M  DACR POULAB J AL J SKIN SOI J  Gullah Jal Roxa Centifolio) Pu  HM - AYU - DOM  ANA LAXATIVE POM  DACR POULAB J AL J SKIN SOI J  Gullah Jal Roxa Centifolio) Pu  HM - AYU - DOM  ANA LODOM  ANA LODOM  ANA LODOM  ANA LODOM  ANA LODOM  ANA LOROM CEntifolio) Pu  HM - AYU - DOM  ANA LODOM  ANA LOROM CEntifolio) Pu  HM - AYU - DOM  ANA LOROM CEntifolio) Pu  HM - AYU - DOM  ANA LOROM CEntifolio) Pu  HM - AYU - DOM  ANA LOROM CENTIFOLION  ANA LOROM CENTIFOLION  HM - AYU - DOM  ANA LOROM CENTIFOLION  HM - AYU - DOM  ANA LOROM CENTIFOLION  ANA LOROM CENTIFOLION  HM - AYU - DOM  ANA LOROM CENTIFOLION  ANA LOROM C | in part i indications couch symp Manufacturing process ingression to I for a critical negligible in the same and registers and stress make by adding water to this incarpium) Panchangs I got Manufacturing process ingresion to I for a critical negligible in the activation of the property of the part of the    | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Ayu Ph-P&P  tun) whit 5 gm 5 Yetus (Sali  Ayu Ph-P&P  this suger is added and gyrup basecultur Swertia charattay Back  Ayu Ph-P&P  Shing 4) Methit Trigonella for  Ayu Ph-P&P  Ce4) Dessy-As per physician uni Cralage ya anivvala Bark  Ayu Ph-P&P  iodium (harries 1996 5 Ku)  Ayu Ph-P&P  | prepared attainher officinaliss Rhisomes— Each capsule contain- hex aspeca/root-5(ling, 5)  each 100 ML contains cach Will gri cutains at Circus reticulata/Fruit chal 7  each 100 gri contains verifficial contains ext of Each 100ml contains ext of Each 100ml contains extract cutain/Seed fil mg Sikala jeera Tach 100ml contains extract cutain/Seed fil mg Sikala jeera Tach 100ml contains extract cutain/Seed fil mg Sikala jeera Tach 100ml contains contains a jeera (Computin contains) each 100 gri contains a jeera (Computin contains)  |
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| AP - AYU - DOM DCARL STONGEET CAPSULES D Rhursum air aunt Hysocymus skhandlscorus ealamistiroot - 2 RO - AYU - DOM NASA HAR OIL - bair oil PL - AYU - DOM ANNA HAR DER - face pack INI Masurt Fishian lens Dal 7 gm 2 6. Anantmonit Hemideenins in PL - AYU - DOM (WOUND CARE LEPAM CREA That History Benghalmski) por Annahanghagyeyrinza gilahra VP - AYU - DOM (CON SYRUP Author-bhayprakas Bhringymit leichpta atbarPance AP - AYU - DOM MICON CARSULE / Health unic, D Augarmathar Cyprus roundla gollu autwo Seed 20 mg Ayu 1 79 - AYU - DOM ANAL LAXATIVE POWDER CHIL Triphalatashia, Behada, Harda de J Dom dyny Ph 4 GAD, NA-AYU - DOM ANAL LAXATIVE POWDER CHIL Triphalatashia, Behada, Harda de J Dom dyny Ph 4 GAD, NA-AYU - DOM Galab JalfRoin Centifolia) Pt HM - AYU - DOM DCARP LAXACEPT CHURN dig Triphalatashia, Behada, Harda de J Dom dyn Ph 4 GAD, MA - AYU - DOM DCARP LAXACEPT CHURN dig Triphalatashia, Behada, Harda de J Hym dyn Bur Ph mod J 100 G AP - AYU - DOM   | in part i mideathorsoude symp Manufacuring process ingredient act to or accident degline and rear mode by adding water, to this incarpum) Panchang - Igm. Manufacuring process ingredient act to or accident degline and getter and cive adding water to this incarpum) Panchang - Igm. Manufacuring panchang - Igm. A) Haldri Curciuma funga as prop med.  But about seed-afting - D. Kantinkay/Sodamum xanthoc acpum) root-50mg, 3) Chotta gokhurut Tribulas terrestris] huit-7 sang, 6) Manufashan Rudina cardifolial Root-75mg, 4 vp. Ph. (t. Total \$60 mg)  ayu prop med.  AVU PROP MED/FOR EXT USE.  MAYU PROP MED/FOR EXT USE.  Manufashad-WOUND CARE.   | Ayu Ph-P&P  Sing 4) Chirchirat Achyrant  Ayu Ph-P&P  Nyw Ph-P&P  Nyw Ph-P&P  Ayu Ph-P&P  tun) whit 5 gm 5 Yetus (Sali  Ayu Ph-P&P  this suger is added and gyrup basecultur Swertia charattay Back  Ayu Ph-P&P  Shing 4) Methit Trigonella for  Ayu Ph-P&P  Ce4) Dessy-As per physician uni Cralage ya anivvala Bark  Ayu Ph-P&P  iodium (harries 1996 5 Ku)  Ayu Ph-P&P  | prepared Each capsule contain hex aspecta/root-50ing, 5)  cach 100 ML contain cach 100 mL contain cach 100 gm contain a caproot, bark 8 gm 6.  Each 100 mL contains ext o e is prepared -7. 5 gm. 5) Guchachi (Tanospora Fach capsule contains extract cutum/Seed bit mg 5)Kala feera Unch 100 gm contain a feera (Cummum extract) cach 100 gm contain a feera (Cummum extract) cach 100 mL contain cach 100 mL contain cach 100 gm contains powder  |
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|   | 1)Bhringraj(Eclipta alba)Panchang-5gm,2)Amla(Embilea officinalis)Fruft-2gm, 3)Kurphad(aloe<br>barbadenssis)Leaf-10gm, 4)Nem(Azadirachita indica)Leaf-2gm,Brahmi(Centella asiatica)panchang-<br>2gm, Water base presentis.  | Avu Ph-P&P   | *. Each 100ml contains ext of  |
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| 10 / AVI PROP MEDITOR EXT.  | SOLUTION   | Avu Seh-L-P.C.P  |  |
| Amalaki (Emblica officinalis  | SEEMINGAG extraction arising with preservative filling packing<br>stfruit-5gm, 2) waith ficus bangalensis reont-5gm, 3) juryaandi hibiscus rosas/nensis/flower-3gm, 4) korphadd ab   | ng harbada atata (12   | each 100ml contains  |
| AP - AYU - DOM  | LIVOSPAR CAPSULE   | Commence of the Commence of th |  |
| Author-Bhaspeakash part 1. Mfg. po<br>Bhuianiala(Phyllanthus niru   | recess-Mixing capsule filling packing, Indication-Liver twite Doses-ic per physician (F) Panellano (Mina) (Galache T. T.   | Ayu Ph-P&P   | EACH CAPSULE CONTAINS<br>EXTRACTS OF   |
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| DCARF DBCFPT CAPSULES (in<br>Jamun(Syzygium cumini) See<br>mg Avu Ph (Total 360 mg)                                   | adication/docests per physician<br>d-61mg, 21KavelatMomordica charuntia)Fenit 61mg, 3) Methi/Tri <b>gonella foenimgraecia</b> n/Seed-61mg,4) Bahi  | Ayu Ph-P&P  Il Acacia arabica)Bark-60 ms   | Fach capsule contains 51WadtFiscus BengalensisiBark  |
| ANA P CARE CAPSULE/INDIC  | A YU PROP MED  | Avu Ph-P&P   | Each cancula contains average  |
| perus scatiosus)Rhizome 60  | Rooff 60 mg 2 Bhutanda(Phyllanthus meurit) Panchung 70 mg 3. Kaniakarif Salamum xanthocarpunii Rooff 50 mg<br>mg 6. Padalt Stercospermum suuveolinsi Panchung 60 mg Apa Ph. t. 3600  | 4. Chandrasurr Lepidimi sate   | cum/Seed 60 mg 5.Nagarmotha  |
| 0 HAIR OIL / Hair oil/mfe-sidhly  | AYU PROP MED/FOR EXT USE   | Avu Ph-P&P   | each 100 ml castain  |
| Til Oil (Seusanum Indicum)<br>ılımi (Centila Asistica) leaf 3   | seed 90 mt 2.Jaswand(Hibiscus rosa sinensis)frait 5 gm 3.WadtFines Bengalensis)Jata 7 gm 4. Bhrigrai (Edip<br>gm 4xu Ph (23 GM)  | ta AlbaiPanchang 3 gm 5. An  | nla(Emblica Offinalis)fruit 15 om t  |
| L - AYU- DOM<br>DCARE ORTHOCEPT OIL EXT   | avu prop med/for ext use   | Avu Ph-P&P   | each 100 ML contain  |
| Veem vill Azadirchita indica  | 1- Year Sarra ailt Marraud 70 of 185 1   | nunkuliGum-Som AiSalai m   | and the second of the Committee  |
| AP - AYU-DOM  | <ol> <li>Vayuvidang(Emblica ribes)Fruit-75mg, 2) Kale mire(Piper nigranuin)Fruit-75mg, 3) Sarpunkha<br/>(Tephrosia purpuria)Root-50mg, 4) Lajri(Mimosa pudica)Panchang-50mg, 5) Chitrakmool Plumbage</li> </ol>  |  | FACIL CAPSULF CONTAIN<br>EXTRACTS O  |
|   | result 2 from 2) Kall mixed Discharge and Mrg process Mixing capsule filling and packing   |  |  |
| Planicas Root-75mg, 6) Chand<br>YP - AYU - DOM  | Fruit-15mg, 21 Kale miret Phyer nigrammilFruit-75mg, 33 Sorpunkha (Tephroxia purpuria)Root-50mg, 4) Lairit<br>travurt Lepidum xativum)Seed-75mg Ayu Ph (60 unus)   | Mimosa pudica)Panchang-5   | mg, 51 ChitrakmooltPlumbago  |
|   | 1) Guduchi(Tinospora cordifolia)Panchange-(0gm, 2) Sunth(Zizimber officinalis)Rhizomes-2gm, 3) Chirafta(Swertia chirafta)Bark-3gm, 4) Pi(papda(Fumaria indica)Panchange-4gm, 5) Bhringraj(Eclipalha)Panchang-3gm, 6) Padal(Stereospermum supaveolens)Bark-5gm, 7) Vidarikand(Pueraria tuberosa)Root-5gm, 8   |  | Fach 100ml contains ext  |
| pa)Panchang-3gm. 6) Padali!   | aash par i Muhaatioul-digestive tonie Manufacturing process-Ingredient to 1 to 7 are taken together and extract made by adding on<br>this Panchange-10gm, 2) Sunth (Extraober officinalis) Rhizomes-2gm, 3) Chirariat Swerta chiratia (Bark-1gm, 4)<br>Storeosperinum suparvoctors) Bark-5gm, 7) Vilai (Band-Purearia Interna (Bark-1gm, 4) by 10) climatics   | ater, to this suger is added and syru<br>PitpapdatFumaria indica)Po  | p base is prepared.<br>unchange-4gm, 5) Bhringrai(Eclipto  |
| AP - AYU - DOM  | CAPSULE  CAP | Ayu Sch-I-P&P  | Fach Capsule Contains, extrac  |
| Vidarikand(Pueraria tubero,<br>undus)Rhizome-120mg Ayu S  | sa)root-20mg,2)Amla(Emblica officinalis)Fruit-6(three 30Bbrodon)(Contalla monta of Paul Contalla of Co | Ficus hengalensis)Root-30ma  | (.5) NagarmothatCyperus  |
| AP - AYU - DOM  | SPARTA CAPSULE   | Avu Ph-P&P   | FACIL CAPSULE CONTAIN  |
| uthor Bhavprakash part I Manutas<br>shwagarallia(Withinia sonife<br>diJolia)Panchang-60mg Ava                         | cturing process. Mrs.ing.Capsale filling Packing. Indication Death tong. Doses As per physician.  170) R001-75m2, Lannel Teef (Afueting prise ens) See 4-75m2, Gokhren Fribidus Ferrissus) Fruit Stong d'Onig, State Ph. 1360 mg.  |  | EXTRACTS   |
| RL-AYU-DOM  | ayu prop med   | Ayu Ph-P&P   | cach 100 ML conta  |
| CARE BRENOCEPT SYRUP (*) Brahmi(Bacopa monari)Lea (100 ml)  | (ONECobe as per physician<br>af-10gm, 2) ShankhapitshpitEvolvulus alsinoides)Panchung-10gm, 3) Bach (Acorus calannus)Rhizomes-5gm,   |  |  |
| P - AYU - DOM.  | AYU PROP MED: HEALTH TONIC , DOSES: AS PER PHYSICIAN   | Ayu Ph-P&P   |  |
| amun(Syzygium cumini)See  | d 60 mg 2. Karela(Momordica charuntia)Fruit 60 mg 3. Methit Trigonella Foenumgraeum ) Seed 60 mg 4. h  |  |  |
| - AYU- DOM  | AYU PROP MED/FOR EXT USE   |  |  |
| GULABIAL ARK / INDICATION Juliab Jai(Rosa Centifolia) J   | N. SKIN SOLUTION'External application) Dissects per physician<br>enals 5 ml base material-lega, Glycerine, Preservatives, Sugandhi Dravya (Q.S. Ayu Ph. 6-ml)  | Ayu Ph-P&F   | each 10 ml cant  |
| P - AYU-DOM<br>CARE LIVOCEPT CAPSULES   | ayu pi op tijed/dose-as per physician  | Ayu Ph-P&F   | Fach capsule contains extr   |
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Print Date: 04/10/2021, Data Counts; (0 - 0)



# Sighting record of Black Eagle from Kitali Lake, Maharashtra, India

The Black Eagle *Ictinaetus malaiensis* (Temminck 1822) is a large diurnal raptor of tropical and sub-tropical mountainous forests. (Clark 1994;Cristae 2001). It is the monophyletic species, placed in the family Accipitridae. It has two races, *I. m. malaiensis*, found mainly in China and the Indian race *I. m. perniger* is distributed throughout the Himalayan foothills from Kashmir to Arunachal Pradesh, the Western and Eastern Ghats, the highlands of Madhya Pradesh, and the Chhota Nagpur Plateau regions in Odisha and West Bengal (Naoroji 2006; Rasmussen & Anderton 2012).

On the 9 January 2022, during our birdwatching trip to Satbahini foothills, when we were busy taking photographs of the



Site map of Kitali poind amongs forested hilly ranges and large Ghodazari Lake, showing in inset hydrophytes like Nelumbo sp. providing ideal habitat for waterfowl and Jacana's, also surrounded by paddyfields.

Common Hawk-Cuckoo at a roadside small wetland Kitali pond, we were very surprised to get glimpses of a large black-coloured raptor, perched on *Azadirachta indica*. Due

to vehicular movement on both the sides of the road, the bird got disturbed. It just takeoff

> Image 1: Photo showing location site of Black Eagle over the Azadirecta indica on the embankment of Kitali pond. A. Total black with brownish patch on should except, yellow coloured cere and toes. B. Struggling to perch, reveals underparts wings having lighter bars on primaries and secondaries, unique to Black Eagle. C. Perched Black Eagle revealing lighter bars on tail retrices and pale brownish colour at the bottom line of tail. D. Black Eagle glies passively by spreading flat wings.



ird-0-soar #174 21 July 2022

from the perched site and passively glided overhead twice. It was a real delight to watch such a passive sail of black raptor, giving me sufficient time to capture photographs in Canon 200D camera. Initially, we thought it to be a Greater Spotted Eagle, but after citing few research articles and referencing through field guides of Grimmett et al. (1999) and Ali (2002), it was confirmed as Black Eagle. The body of the bird is entirely black in colour. The beak has yellow cere with a black tip and toes are yellow in colour. A whitish patch is seen at the base of primaries and pale barring on primaries, secondaries and tail coverts.

Kitali Lake (20.502N, 79.645265E) is located near forested area of Satbahini hill ranges in Nagbhid. It lies within the buffer area of newly declared 'Ghodazari Sanctuary' by Maharashtra Government.

The Kitali Lake is 16 km northeast of Nagbhid and 94 km south of Chandrapur. This part of Nagbhid Taluka of Chandrapur District has mixed vegetation of moist and dry deciduous forest interspersed with hill ranges of Shiv Tekdi of Nagbhid, Muktai Hills towards Chimur Taluka and Satbahini Hill nearby study site. Agrarian rural people totally depend upon paddy cultivation for their livelihood. The study site is a perfect wetland habitat for waterfowls like, Greyheaded Swamphen, Bronze-winged Jacana, Pheasant-tailed Jacana and so many waders as the pond is shallow and occupied hydrophytes like *Nelumbo* sp

While going through the published research articles, it was found that, there are only a few published records of the species from Vidarbha region. Black Eagle was reported earlier in Maharashtra from Satpuda hill ranges (Wadatkar et al. 2014), which is about 400 km away from Satbahini Hills.

Although widespread in distribution and enlisted as Least Concern as like other raptors, Black Eagle also shows declining trend (BirdLife International 2022). Earlier, it was a regular winter migrant to forested mountainous ranges and hills in peninsular India, but in the recent past, it is very rare and uncommon to locate them in this part of eastern Vidarbha and is a vagrant species. It was earlier reported from the core zone of Tadoba Andhari Tiger Reserve by Bayani & Dandekar (2017), but there was no photographic record of this raptor, hence, we conclude that sighting of Black Eagle from this unprotected area is a very significant event and further study is recommended for their protection and conservation.

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Citation: Deshmukh, G.D. & N. Borode (2022). Sighting record of Black Eagle from Kitali Lake, Maharashtra, India. Bird-o-soar #174, In: Zoo's Print 37(7): 39–41.

Zoo's Print

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