## JAI NARAIN VYAS UNIVERSITY JODHPUR



<u>2016 - 2021</u>

3.4.3 Number of Patents published / awarded during the last five years.







INTELLECTUAL PROPERTY INDIA PATENTS [DESIGNS] TRADE MARKS GEOGRAPHICAL INDICATIONS



भारत सरकार GOVERNMENT OF INDIA पेटेंट कार्यालय THE PATENT OFFICE पेटेंट प्रमाणपत्र Patent Certificate (Rule 74 of Patents Rules)

| Patent Number      |     | 271054   |
|--------------------|-----|--|
|                    |     | 2/1054   |
| Application Number | :   | 2774/DEL/2006  |
| Date of Filing     | : " | 22/12/2006   |
| Patentee           | :   | 1.DEPARTMENT OF BIOTECHNOLOGY(DBT)<br>2. J.N.V. UNIVERSITY |

It is hereby certified that a patent has been granted to the patentee for an invention entitled "AFFINITY BIOSENSOR FOR DOPAMINE" as disclosed in the above mentioned application for the term of 20 years from the 22 day of DECEMBER 2006, in accordance with the provisions of the Patent Act 1970.

Controller of Patents

Date of Grant:29/01/2016

Controller General of Patents, Designs & Trademarks

Note: The fees for renewal of this patent, if it is to be maintained, will fall/has fallen due on 22 day of DECEMBER 2008 and on the same day in every year thereafter.

## **Indian Patents**

- **Recently Granted Patents** •
- **Recently Published Patents**

## Title of Invention

## "AFFINITY BIOSENSOR FOR DOPAMINE"

A novel process for the development of a biosensing method having a receptor protein as sensing element for a Surface Plasmon Resonance SPR based affinity biosensor for detection of dopamine DA comprising the incubation of receptor D-RC in the concentration range of 20-30 ug/ml, with DA in the concentration range 0.1 ng/ml -700 ng/ml, followed by injection over DA-BSA conjugate and duration of injection in the range of 3 to 7 minutes preferably 5 minutes leading to a shift of resonance angle by  $0.020^{\circ}$  to  $0.040^{\circ}$ , the decreases in the resonance

Abstract angle shift being in proportion to the concentration of DA; the sensor surfaces being fabricated by physical adsorption of the conjugates on to the transducer surfaces, a microscopic glass slide coated with a ~5 nm layer of chromium and ~ 50nm layer of gold serving as transducer surface attached to the prism of the surface plasmon resonance instrument, wherein the sensor surface is functionalized with a protein conjugate of dopamine and bovine serum albumin with phosphate buffered saline (PBS) 0.1M, pH 7.2 as carrier solution.

## Field of Invention:

This invention relates to the development of a D 3 dopamine receptor based, sensing element for a "surface plasmon resonance" based affinity biosensor for highly sensitive and selective detection of dopamine.

Background of the Invention;

Dopamine is a substance of prime importance for optimal regulation of human nervous system. In the biomedical field, to detect the presence and to monitor the level of dopamine is of immense importance for the clinicians to identify the CNS related disorders as well as to decide the dose estimation of external supplement. Various analytical methods including HPLC, capillary electrophoresis, fluorescence and amperometry have been demonstrated for detection of dopamine. Reviewing the wide number of reports, it can be realized that the selective detection of dopamine remains a challenge because of the presence of large excess of ascorbic acid, uric acid

Full

Text and other analogous endogenous compounds with dopamine in biological samples, which limits their application to invivo analysis. Moreover, they often involve time consuming and complicated procedures. Various strategies have been employed in electrochemical based sensors for alleviation of this selectivity problem such as, coupling with chromatographic separation, surface pretreatment and using permselective membranes. However the problem of selective detection of dopamine remains unsolved.

Biosensors are analytical devices comprising a biological or biologically derived sensing element immobilized at a physicochemical transducer to measure one or more analytes. The sensing element could be enzymes, microorganism, tissue slices, biomimetic catalysts, antibodies, nucleic acids, receptor proteins or synthetic receptors.

In contrast to the most common enzyme based biosensors, the affinity based

biosensing element (antibodies, receptors) do not usually catalyze chemical transformations and rather undergo a physical transformation while interacting with corresponding antigen/ligand which can be detected by physical transducer. Most advanced Surface Plasmon Resonance (SPR) based optical transducers are now well established technology capable of monitoring antibody-antigen based immunoreactions or receptor-ligand based affinity reactions. The binding between immobilized receptor and ligand changes the refractive index, leading to the change in SPR angle, which can be monitored in real time. The magnitude of the change in SPR signal is directly proportional to the mass bound to the surface even at nanogram levels.

It is necessary to develop an appropriate biosensor which can measure dopamine independent of presence of antibodies against it, as antibody production is a reaction phenomenon of body in response to entrance/presence of antigen.

There is no biosensor yet developed to measure the presence of dopamine invivo or invitro conditions.

## Objects of the Invention;

The object of this invention is to develop a receptor based affinity biosensor for highly selective and sensitive detection of dopamine.

Other objective is to develop a sensor capable of performing invivo estimation of dopamine without prior separation in medical diagnostics.

Another object of this invention is to use Surface Plasmon Resonance based optical transducer for dopamine sensing.

Yet another object is to use receptor for developing biosensor which is a natural target for analyte as they possess high affinity and specificity refined by evolutionary process.

Other object of this invention is to avoid the use of antibody as recognizing element which involves challenging the experimental animals.

Another object is to develop the stability of receptor subunits by using immobilization conditions that closely related to natural environment.

Yet another object is to develop a multiple analysis system of hundreds of binding events from very small sample volumes.

## Detailed Description of the Invention;

The principle of the invention is that a biosensor to sense qualitative and quantitative presence of dopamine invitro and invivo can be developed using receptors instead of antibodies.

Inventors have established here receptor based sensing element for a SPR based affinity biosensor for highly sensitive and selective detection of dopamine (DA), employing a D3 dopamine receptor and a home made DA-bo vine serum albumin (DA-BSA) conjugate.

The new methodology presented here using receptor as a recognition element for dopamine detection based on Surface Plasmon Resonance (SPR) technique proved to be a potential tool for highly sensitive and selective detection of dopamine with good reliability and reproducibility. The sensing idea is simple and the high affinity molecular recognition of the dopamine receptor (D-RC) provided remarkable specificity for dopamine (DA) against ascorbic acid (AA), uric acid (UA) and other dopamine analogues viz., 3,4 dihydroxyphenyl acetic acid (DOPAC) and 3-(3,4 dihydroxyphenyl)-alanine (DOPA). The proposed method provides a new and promising route for reliable and economic biochemical diagnosis of risk of dopamine

related disorders.

The sensor surfaces were fabricated by physical adsorption of the conjugates (analytecarrier protein) onto the transducer surfaces. A microscopic glass slide coated with a ~5 nm layer of chromium and ~50 nm layer of gold served as transducer surface and was attached to the prism of the surface plasmon resonance instrument (Model SPR-670, Nippon Laser and Electronics, Japan). The sensor surface was

functionalized with a protein conjugate of dopamine and bovine serum albumin (BSA). Phosphate buffered saline (PBS, 0.1 M, pH 7.2) was used as the carrier solution. A pepsin solution, glysin -HCI buffer, pH 2 was used for regeneration of sensor surface.

The sensitive and selective interactions of the dopamine receptors (D-RC) with dopamine-bo vine serum albumin conjugate (DA-BSA) were monitored using SPR technique. Detection of dopamine (DA) was carried out using the principle of indirect competitive inhibition, which is a promising protocol for sensitive detection of small molecules in techniques like SPR where molecular interaction is being observed. The developed affinity biosensor is remarkably sensitive showing, detection limit 85 pg/ml (ppt, parts per trillion) and highly selective with a response time of 5 minutes. Monitoring dopamine concentration through present "Affinity Sensor" technique will be helpful as an early marker rather than as a diagnosis of CNS-related disorders. The two main novelties lie in the use of SPR as optical transducer and the use of a natural receptor as bio-recognizing element in the development of affinity biosensor for dopamine.

## We Claim;

1. A novel process for the development of a biosensing method having a receptor protein as sensing element for a Surface Plasmon Resonance SPR based affinity biosensor for detection of dopamine DA comprising the incubation of receptor D-RC in the concentration range of 20-30 ug/ml, with DA in the concentration range 0.1 ng/ml -700 ng/ml, followed by injection over DA-BSA conjugate and duration of injection in the range of 3 to 7 minutes preferably 5 minutes leading to a shift of resonance angle by  $0.020^{\circ}$  to  $0.040^{\circ}$ , the decreases in the resonance angle shift being in proportion to the concentration of DA; the sensor surfaces being fabricated by physical adsorption of the conjugates on to the transducer surfaces, a microscopic glass slide coated with a ~5 nm layer of chromium and ~ SOnrn layer of gold serving as transducer surface attached to the prism of the surface plasmon resonance instrument, wherein the sensor surface is functionalized with a protein conjugate of dopamine and bovine serum albumin with phosphate buffered saline (PBS) 0.1M, pH 7.2 as carrier solution.

2. A novel process for the development of a biosensing method as claimed in claim 1, wherein the immobilised assay format could be reused by simple regeneration of its original state by injecting 2-10 jig/ml pepsin solution (glysin -HC1 buffer, pH 2.0) for 10-60 s.

3. The novel process for the development of bio sensing method as claimed in claim 1 wherein the biosensor is providing remarkable sensitivity with a detection limit of 85 pg/ml and a response time of 5 minutes.

4. The process for the development of a biosensing method as claimed in claim 1, wherein the sensing idea is simple and the

high affinity molecular recognition of the D-RC provided remarkable specificity for DA against interference from coexisting compounds such as ascorbic acid, uric acid and other DA analogues.

5. The process for the development of a biosensing method as claimed in claim 1 wherein almost similar resonance angle shift for the interaction of D-RC with DA-BSA conjugate is detected after multiple analysis cycles to be reused for nearly 20 cyles of measurements.

6. The process for the development of a biosensing method as claimed in claim 1, wherein monitoring dopamine concentration is used as an early marker rather than diagnosis.

7. The process for the development of a biosensing method as claimed in claim 1, wherein affinity interaction based biosensor is used for the first time for dopamine detection using receptor as the bio recognition element.

8. The process for the development of a biosensing method as claimed in claim 1, wherein the SPR is used for the first time as physical transducer for sensing dopamine.

## **Documents:**

http://ipindiaonline.gov.in/patentsearch/GrantedSearch/viewdoc.aspx?id=GuleQKMEJIFV2It 0xDDiGg==&loc=+mN2fYxnTC4l0fUd8W4CAA==

## <u>« Previous Patent</u> Next Patent »

| Patent Number                    | 271054            |
|----------------------------------|-------------------|
| Indian Patent Application Number | 2774/DEL/2006     |
| PG Journal Number                | 06/2016           |
| Publication Date                 | 05-Feb-2016       |
| Grant Date                       | 29-Jan-2016       |
| Date of Filing                   | 22-Dec-2006       |
| Name of Patentee                 | J.N.V. UNIVERSITY |
| Applicant Address                | JODHPUR-342003.   |

## **Inventors:**

| # Inventor's Name | Inventor's Address                        |
|-------------------|---|
|                   | J.N.V.UNIVERSITY, DEPARTMENT OF CHEMISTRY |
| I SUNITA KUMBHAT  | JODHPUR-342003. INDIA                     |
|                   | ART, SCIENCE AND TECHNOLOGY CENTER FOR    |
| 2 NORIO MIURA     | COOPERATIVE RESEARCH, KYUSHU              |
|                   | UNIVERSITY,KASUGA-SHI,FUKUOKA, 816-8580.  |

|                                      | DIVISON OF VIROLO  | GY AND MOLECULAR         |
|--------------------------------------|--------------------|--------------------------|
| 2 VINOD IOSUI                        | BIOLOGY, DESERT MI | EDICINE RESEARCH         |
| 5 VINOD JOSHI                        | CENTER,(INDIAN CO  | UNCIL OF MEDICAL         |
|                                      | RESEARCH), JODHPUI | R-342003.                |
|                                      | ART, SCIENCE AND T | ECHNOLOGY CENTER FOR     |
| 4 SHANKADAN                          | COOPERATIVE RESEA  | ARCH,KYUSHU              |
| SHANKARAN                            | UNIVERSITY, KASUG  | A-SHI,FUKUOKA, 816-8580. |
| K.                                   | ART, SCIENCE AND T | ECHNOLOGY CENTER FOR     |
| 5 VENGATAJALABATHY                   | COOPERATIVE RESEA  | ARCH,KYUSHU              |
| GOBI                                 | UNIVERSITY, KASUG  | A-SHI,FUKUOKA, 816-8580. |
| <b>PCT International Cla</b>         | ssification Number | G02B                     |
| PCT International Application Number |                    | N/A                      |
| <b>PCT Internation</b>               | al Filing date     |                          |
| PCT Conventions:                     |                    |                          |

# PCT Application Number Date of Convention Priority Country 1 NA

© All Indian Patents, 2013-2016.

Patent data available in the public domain from Indian Patents Office, Department of Industrial Policy and Promotions, Ministry of Commerce and Industry, Government of India.



European Patent Office 80298 MUNICH GERMANY

Questions about this communication ? Contact Customer Services at www.epo.org/contact

Date

11.01.16

| Reference                                       | Application No./Patent No.<br>13814642.8 - 1352 |  |
|---|---|--|
| Applicant/Proprietor<br>Hindustan Petroleum Cor | poration Ltd., et al                            |  |

## Notification of the data mentioned in Rule 19(3) EPC

In the above-identified patent application you are designated as inventor/co-inventor. Pursuant to Rule 19(3) EPC the following data are notified herewith:

DATE OF FILING

: 23.07.13

PRIORITY : IN/24.05.13/ INA MM18522013

TITLE

: CATALYTIC DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON OXIDES FREE HYDROGEN AND BAMBOO SHAPED CARBON NANOTUBES

DESIGNATED STATES

: AL AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HR HU IE IS IT LI LT LU LV MC MK MT NL NO PL PT RO RS SE SI SK SM TR





European Patent Office 80298 MUNICH GERMANY

Date

Questions about this communication ? Contact Customer Services at www.epo.org/contact

## 

De Vries & Metman **Overschiestraat 180** 1062 XK Amsterdam PAYS-BAS

16.03.16

| Reference   | Application No./Patent No.  |
|---|-----------------------------|
| EP23421-HS/pb   | 13814642.8 - 1370 / 3003552 |
| Applicant/Proprietor<br>Hindustan Petroleum Corporation Ltd., et al |                             |

## Communication of European publication number and information on the application of Article 67(3) EPC

The provisional protection under Article 67(1) and (2) EPC in the individual Contracting States becomes effective only when the conditions referred to in Article 67(3) EPC have been fulfilled (for further details, see information brochure of the European Patent Office "National Law relating to the EPC" and additional information in the Official Journal of the European Patent Office).

Pursuant to Article 153(3) EPC the publication under Article 21 PCT of an international application for which the European Patent Office is a designated or elected Office takes the place of the publication of a European patent application.

The bibliographic data of the above-mentioned Euro-PCT application will be published on 13.04.16 in Section I.1 of the European Patent Bulletin. The European publication number is 3003552.

In all future communications to the European Patent Office, please quote the application number plus Directorate number.

## For the Examining Division



## LIS 20160129424A1

## (19) United States

## (12) Patent Application Publication (10) Pub. No.: US 2016/0129424 A1 Pant et al.

#### (54) CATALYTIC DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON OXIDES FREE HYDROGEN AND BAMBOO SHAPED CARBON NANOTUBES

- (71) Applicants: HINDUSTAN PETROLEUM CORPORATION LTD., Mumbai (IN); INDIAN INSTITUTE OF TECHNOLOGY, DELHI. New Delhi (IN): CENTRE FOR HIGH TECHNOLOGY (CHT), Noida (IN)
- (72) Inventors: Kamal Kishore Pant, New Delhi (IN); Sushil Kumar Saraswat, New Delhi (IN); Annaji Rajiv Kumar Tompala, Bangalore (IN): Kanaparthi Ramesh, Bangalore (IN); Venkata Chalapathi Rao Peddy, Bangalore (IN); Venkateswarlu Choudary Nettem, Bangalore (IN); Sri Ganesh Gandham, Bangalore (IN)
- 14/893,643 (21) Appl. No.;
- (22) PCT Filed: Jul. 23, 2013
- PCT/IN2013/000460 (86) PCT No.: § 371 (c)(1). (2) Date: Nov. 24, 2015

## May 12, 2016 (43) Pub. Date:

(30)Foreign Application Priority Data

**Publication Classification** 

| (51) | Int. CL    |           |
|------|------------|-----------|
|      | B01J 23/80 | (2006.01) |
|      | C01B 3/26  | (2006.01) |
|      | C01B 31/02 | (2006.01) |
|      | B01J 37/08 | (2006.01) |
|      | B01J 37/18 | (2006.01) |
|      |            |           |

(52) U.S. Cl. CPC B01J 23/80 (2013.01); B01J 37/08 (2013.01); B01J 37/18 (2013.01); C01B 31/0233 (2013.01); C01B 3/26 (2013.01); C01B 3L/026 (2013.01); C01B 2202/36 (2013.01); C01B 2202/34 (2013.01); C01P 2004/04 (2013.01); C01P 2004/13 (2013.01); C01P 2004/61 (2013.01); C0/B 2203/0277 (2013.01); C0/B 2203/1076 (2013.01); C01B 2203/1241 (2013.01)

#### (57) ABSTRACT

In accordance with the present subject matter there is provided a process for catalytic decomposition of lower hydrocarbons to produce carbon oxides free hydrogen and bamboo shaped carbon nanotubes over a catalyst composition. The process for catalytic decomposition of lower hydrocarbons comprises contacting lower hydrocarbon over a catalyst composition, where the catalyst composition comprising, a catalyst, at least one modifying agent and a support material.



Home (http://ipindia.nic.in/index.htm) About Us (http://ipindia.nic.in/about-us.htm) Who's Who (http://ipindia.nic.in/whos-who-page.htm) Policy & Programs (http://ipindia.nic.in/policy-pages.htm) Achievements (http://ipindia.nic.in/achievements-page.htm) RTI (http://ipindia.nic.in/right-to-information.htm) Feedback (https://ipindiaonline.gov.in/feedback) Sitemap (shttp://ipindia.nic.in/itemap.htm) Contact Us (http://ipindia.nic.in/contact-us.htm) Help Line (http://ipindia.nic.in/helpline-page.htm)

Skip to Main Content Screen Reader Access (screen-reader-access.htm)



## (http://ipindia.nic.in/index.htm)



(http://ipindia.nic

### Patent Search

| Invention Title   | CATALYTI<br>NANOTU   | C DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON OXIDES FREE HYDROGEN AND BAMBOO S<br>BES        | HAPED CA |
|---|--|---|----------|
| Publication Number  | 36/2016  |   |          |
| Publication Date  | 31/08/2016   |   |          |
| Publication Type  | INA  |   |          |
| Application Number  | 1852/MU  | M/2013  |          |
| Application Filing Date   | 24/05/20   | 13  |          |
| Priority Number   |  |   |          |
| Priority Country  |  |   |          |
| Priority Date   |  |   |          |
| Field Of Invention  | CHEMICA  | L   |          |
| Classification (IPC)  | C01B 31/   | 02,B01J 23/88   |          |
| Inventor  |  |   |          |
| Name  | Address  |   | Country  |
| PANT, Kamal Kishore   | INDIAN INS   | STITUTE OF TECHNOLOGY DELHI (IIT DELHI), HAUZ KHAS, NEW DELHI-110 016                                   | India    |
| SARASWAT, Sushil Kumar  | INDIAN INSTITUTE OF TECHNOLOGY DELHI (IIT DELHI), HAUZ KHAS, NEW DELHI-110 016   |   | India    |
| TAMPALA, Annaji Rajiv<br>Kumar  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066       |   | India    |
| RAMESH, Kanaparthi  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066       |   | India    |
| RAO, Peddy Venkata<br>Chalapathi  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066       |   | India    |
| CHOUDARY, Nettem<br>Venkatewarlu  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066       |   | India    |
| GANESH, Gandham Sri   | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066       |   | India    |
| PEDDY, VENKATA<br>CHALAPATHI RAO  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066 INDIA |   | India    |
| NETTEM,<br>VENKATESWARLU<br>CHOUDARY  | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066 INDIA |   | India    |
| GANDHAM, SRI GANESH   | HINDUSTAN PETROLEUM CORPORATION LTD., HPCL CORPORATE R&D CENTRE, 1ST FLOOR, ADARSH ECO PLACE, 176 EPIP<br>KUNDENAHALLI HOBLI, WHITEFIELD, BANGALORE- 560 066 INDIA |   | India    |
| Applicant   |  |   |          |
| Name  |  | Address   | Country  |
| HINDUSTAN PETROLEUM<br>CORPORATION LTD.   |  | HINDUSTAN PETROLEUM CORPORATION LTD, PETROLEUM HOUSE, 17 JAMSHEDJI TATA ROAD, CHURCHGATE, MUMBAI 400020 | India    |
| INDIAN INSTITUTE OF TECH<br>DELHI   | INOLOGY,   | INDIAN INSTITUTE OF TECHNOLOGY DELHI (IIT DELHI), HAUZ KHAS, NEW DELHI- 110 016                         | India    |
| CENTRE FOR HIGH TECHNOLOGY CENTRE FOR HIGH TECHNOLOGY (CHT), OIDB BHAWAN, TOWER 'A', 9TH FLOOR, PLOT NO. 2, SECTOR- 73, N<br>Uttar Pradesh 201301 |  | India   |          |

#### Abstract:

In accordance with the present subject matter there is provided a process for catalytic decomposition of lower hydrocarbons to produce carbon oxides free hydroger bamboo shaped carbon nanotubes over a catalyst composition. The process for catalytic decomposition of lower hydrocarbons comprises contacting lower hydrocar catalyst composition, where the catalyst composition comprising, a catalyst, at least one modifying agent and a support material. .





भारत सरकार GOVERNMENT OF INDIA पेटेट कार्यालय THE PATENT OFFICE पेटेट प्रमाणपत्र PATENT CERTIFICATE (Rule 74 of The Patents Rules)

पेटेंट सं. / Patent No.

294099

416/DEL/2011

जावेदन सं. / Application No.

फाइल करने की तारीख। Date of Filing

17/02/2011

1.SHARMA Peeyush Kumar 2.BHANDARI Anil

पेटेंटी / Patentee

प्रमाणित किया जाता है कि पेटेंटी को उपरोक्त आवेदन में यथाप्रकटित METHOD AND APPARATUS FOR REMOVAL AND BIODEGRADATION OF ENGINE OIL FROM CONTAMINATED WATER BODIES नामक आविष्कार के लिए, पेटेंट अधिनियम, १९७० के उपबंधों के अनुसार आज तारीख़ 17th day of February 2011 से बीस वर्ष की अवधि के लिए पेटेंट अनुदत्त किया गया है।

It is hereby certified that a patent has been granted to the patentee for an invention entitled METHOD AND APPARATUS FOR REMOVAL AND BIODEGRADATION OF ENGINE OIL FROM CONTAMINATED WATER BODIES as disclosed in the above mentioned application for the term of 20 years from the 17th day of February 2011 in accordance with the provisions of the Patents Act, 1970.



अनुदान की तारीख : 09/03/2018 Date of Grant :

टिपाणी - इस पेटेट के नवीकरण के लिए फीस, बॉद इसे बनाए रखा जाना है, 17th day of February 2013को और उन्होंने के अध Note. - The lees for renewal of this patent, if it is to be maintained will fall / has fallen due on 17th funsame day in every year thereafter.

कमांक : 011105680 SL No :





US010010874B2

## (12) United States Patent

## Pant et al.

#### (54) CATALYTIC DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON **OXIDES FREE HYDROGEN AND BAMBOO** SHAPED CARBON NANOTUBES

- (71) Applicants: Hindustan Petroleum Corporation Ltd., Mumbai (IN); Indian Institute of Technology, Delhi, New Delhi (IN); Centre for High Technology (CHT), Uttar Pradesh (IN)
- (72) Inventors: Kamal Kishore Pant, New Delhi (IN): Sushil Kumar Saraswat, New Delhi (IN); Annaji Rajiv Kumar Tompala, Bangalore (IN); Kanaparthi Ramesh, Bangalore (IN); Venkata Chalapathi Rao Peddy, Bangalore (IN): Venkateswarlu Choudary Nettem, Bangalore (IN); Sri Ganesh Gandham, Bangalore (IN)
- (73) Assignces: Hindustan Petroleum Corporation Ltd., Mumbai (IN); Indian Institute of Technology (IIT Delhi), New Delhi (IN); Centre for High Technology (CHT), Uttar Pradesh (IN)
- (\*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.
- (21) Appl. No.: 14/893,643
- (22) PCT Filed: Jul. 23, 2013
- (86) PCT No.: PCT/IN2013/000460 § 371 (c)(1), Nov. 24, 2015 (2) Date:
- (87) PCT Pub. No.: WO2014/188439 PCT Pub. Date: Nov. 27, 2014
- (65) **Prior Publication Data**

US 2016/0129424 A1 May 12, 2016

#### US 10,010,874 B2 (10) Patent No.:

#### (45) Date of Patent: Jul. 3, 2018

- (30)**Foreign Application Priority Data** 
  - May 24, 2013 (IN) ..... 1852/MUM/2013
- (51) Int. Cl. (2006.01)B01J 23/80 C01B 3/26 (2006.01)(Continued)
- (52) U.S. Cl. CPC ..... B01J 23/80 (2013.01); B01J 21/04 (2013.01); B01J 37/0201 (2013.01); (Continued)

(58) Field of Classification Search CPC .. B01J 23/80; B01J 37/08; B01J 37/18; C01B 31/0233; C01B 3/26; C01B 31/026; (Continued)

#### (56)**References** Cited

#### U.S. PATENT DOCUMENTS

2003/0108480 A1\* 6/2003 Baker ..... B82Y 30/00 423/447.3

#### OTHER PUBLICATIONS

Ashok, et al., Catalytic Decomposition of Methane to Hydrogen and Carbon Nanofibers over Ni-Cu-SiO2 Catalysts, Energy & Fuels 2009; 23: 5-13.\*

#### (Continued)

Primary Examiner - Daniel C McCracken (74) Attorney, Agent, or Firm - Marshall, Gerstein & Borun LLP

#### (57)ABSTRACT

In accordance with the present subject matter there is provided a process for catalytic decomposition of lower hydrocarbons to produce carbon oxides free hydrogen and bamboo shaped carbon nanotubes over a catalyst composition. The process for catalytic decomposition of lower hydrocarbons comprises contacting lower hydrocarbon over a catalyst composition, where the catalyst composition com-

(Continued)



prising, a catalyst, at least one modifying agent and a support material.

#### 19 Claims, 12 Drawing Sheets

.....

(51) Int. Cl.

| C01B 31/02  | (2006.01) |
|-------------|-----------|
| B01J 37/08  | (2006.01) |
| B01J 37/18  | (2006.01) |
| B01J 21/04  | (2006.01) |
| B01J 37/02  | (2006.01) |
| C01B 32/162 | (2017.01) |
| C01B 32/17  | (2017.01) |
| B01J 37/03  | (2006.01) |
| B82Y 40/00  | (2011.01) |
|             |           |

(52) U.S. Cl.

 2203/1241 (2013.01); C01P 2004/04 (2013.01); C01P 2004/13 (2013.01); C01P 2004/61 (2013.01)

#### (58) Field of Classification Search

CPC ...... C01B 2202/36; C01B 2202/34; C01B 2203/0277; C01B 2203/1076; C01B 2203/1241; C01P 2004/04; C01P 2004/13; C01P 2004/61 See application file for complete search history.

(56) References Cited

#### OTHER PUBLICATIONS

Zhang et al., "Hydrogen Production via the Direct Cracking of Methane Over Silica-Supported Nickel Catalysts", Applied Catalysis A: General 167, 1998, pp. 161-172.

Saraswat et al., "Ni—Cu—Zn/MCM-22 Catalysts for Simultaneous Production of Hydrogen and Multiwall Carbon Nanotubes via Thermo-Catalytic Decomposition of Methane", International Journal of Hydrogen Energy 36, 2011, pp. 13352-13360.

Patel et al., "Renewable Hydrogen Generation by Steam Reforming of Acetic Acid Over Cu—Zn—Ni Supported Calcium Aluminate Catalysts", http://pubs.acs.org, doi: 10.1021/bk-2012-1092.ch009, Jan. 20, 2012, 27 pages.

\* cited by examiner



Figure 1



Figure 2



Figure 3



Figure 4



Figure 5



Figrue 6



Figure 7



Figure 8



Figure 9



Figure 10



Figure 11



Figure 12

#### CATALYTIC DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON **OXIDES FREE HYDROGEN AND BAMBOO** SHAPED CARBON NANOTUBES

#### TECHNICAL FIELD

The subject matter described herein in general relates to a catalyst composition for decomposition of lower hydrocarbons comprising of a catalyst and modifying agent on a 10 support material. The present disclosure also relates to a process for preparation of a catalyst composition for the decomposition of lower hydrocarbons. The present disclosure further relates to a process for producing carbon oxides free hydrogen and bamboo shaped carbon nanotubes by the 15 catalytic decomposition of lower hydrocarbons in the presence of active, catalyst at suitable reaction temperatures and atmospheric pressure.

#### BACKGROUND

Petroleum fuels have long been predominantly used in industry and transport world-over. However, these fuels have a limitation of availability, and they also produce high levels of emissions especially carbon oxides viz carbon 25 dioxide (CO<sub>2</sub>), carbon monoxide (CO). Hydrogen (H<sub>2</sub>) has been suggested to be a good alternative to replace conventional petroleum fuels. H<sub>2</sub> utility as a substitution to fossil fuels has attracted much attention in the last two decades because of its successful demonstration in space technolo- 30 gies and fuel cells, although it necessitates the use of a complex and costly manufacturing process. Conventional methods for production of hydrogen are methane steam reforming, methane partial oxidation or coal gasification routes. However, in addition to production of hydrogen, 35 these methods also produce considerable amounts of carbon oxides (CO<sub>x</sub>) like carbon dioxide, carbon monoxide and their separation is costly and detrimental to the electrodes used in fuel cells. Thus, hydrogen obtained through conventional methods has to be purified to render it free from  $CO_x = 40$ for fuel cell application and other applications. The conventional methods used for hydrogen production involve multistep operations and are not commercially viable.

The production of  $CO_x$  free hydrogen could be advantageous in terms of environmental and economic aspects. The 45 routes proposed for the production of clean hydrogen are ammonia (NH<sub>3</sub>) decomposition, hydrogen splitting and catalytic decomposition of methane (CDM) or lower hydrocarbons. The later method is inexpensive as compared to the former methods for production of CO<sub>x</sub> free hydrogen. The 50 catalytic decomposition of methane can be presented as following.

#### CH<sub>4</sub>→C+2H<sub>2</sub> ∆H=17.8 kcal/mole

CDM to pure hydrogen has several advantages as com- 55 pared to conventional hydrogen production methods. CDM yields pure hydrogen at less severe conditions. Further, the process results in simultaneous production of high value nano-carbons along with hydrogen. This makes the above process as an important alternative process to produce COx 60 free hydrogen. On the other hand, an important disadvantage of the process is that it produces carbon deposits on the catalyst that result in the catalyst deactivation. The regeneration of the deactivated catalyst is done by the combustion of carbon deposits which leads to generation of carbon 65 dioxide. CDM on Ni, Fe and Co based catalysts on inert support materials to give hydrogen is reported. Ni based

2

catalysts are more active and stable than other transition metals and results in higher hydrogen and carbon yield for methane decomposition (Zhang et al. Catal. Lett. 2004, 7).

Methane is decomposed to yield CO<sub>x</sub> free hydrogen and carbon on catalyst based on Ni<sub>x</sub>Mg<sub>y</sub>O (where x and y represent the mole content of Ni and Mg). Addition of Cu as a promoter to the catalyst composition enhances both methane decomposition and solid carbon yield and increases the catalyst life time up to 19 h (US2005/0063900). Methane decomposition in the presence of a catalyst based on. Fe and Ni to give hydrogen enriched fuel and carbon nanotubes have been carried out by using microwave irradiation. In this process, both the catalyst and methane were exposed to microwave irradiation at a selected microwave power (US2008/0210908). Catalysts synthesized by admixing Fe salt and in combination with Ni, Pd, and Mo have been used for decomposition of light hydrocarbons to hydrogen and carbon nanomaterial. The binary metal salts having at least Fe as one metal and in combination with Ni, Pd, Mo were <sup>20</sup> found to be active for the production of hydrogen at the temperature range of 500 to 1000° C. (U.S. Pat. No. 6,875, 417). Multi walled, size controlled carbon nanotubes were produced by the decomposition of carbon containing compounds over supported transitional metal based catalysts. The typical support materials used for the process were SiO<sub>2</sub>, SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub>, aerogel Al<sub>2</sub>O<sub>3</sub> and MgO (U.S. Pat. No. 7,214,360).

Unsupported nano-sized nickel oxide particles have been utilized for the production  $CO_x$  free hydrogen by methane decomposition especially at low temperatures, between 300 to 500° C. It has been observed that the catalyst performance is strongly dependent on the particle size of the catalyst (US2009/0140215). Production of carbon nanotubes can be achieved with the dimension of 3-150 nm having the aspect ratio of more than 100 by the decomposition of hydrocarbons over solid catalyst containing Co, and Mn on an inert support (US2009/0140215). Lower hydrocarbon in the presence of low concentration of oxidizing/reducing gas or moisture can be subjected to decomposition in the presence of catalyst to yield functional carbon nanomaterials and hydrogen. The amorphous carbon produced on the catalyst is removed (U.S. Pat. No. 7,767,182). Silica supported Ni catalyst have been used for catalytic methane decomposition at low reaction temperatures (550° C.) and these catalysts have produced long cylindrical hollow carbon filaments (Zhang et al. Appl. Catal. A: Gen 1998, 161).

One major drawback of the processes for the conversion of methane to give carbon oxides free hydrogen and carbon nanotubes is the rapid deactivation of the active catalyst. Further, the active catalyst deactivates at a rapid rate due to higher amorphous carbon deposition. The conversion of methane is in the range of 50-60% with low carbon yield. Although considerable research has been done on the activity of different catalysts for methane decomposition reaction, there is no effective catalyst available that operates with lesser deactivation for a longer time. While the concepts for catalytic decomposition of methane for production of hydrogen have been shown in the state of the art methods, there still exists a need to develop novel catalytic compositions for decomposition of lighter hydrocarbons to yield carbon nanotubes and  $CO_x$  free hydrogen

#### SUMMARY

The present disclosure relates to a catalyst composition comprising: a catalyst in an amount in the range of 10% to 70% w/w of the total weight of the composition; at least one

modifying agent in an amount in the range of 1% to 14% w/w of the total weight of the composition; and a support material in an amount in the range of 20% to 70% w/w of the total weight of the composition. The catalyst composition described herein is used for decomposition of lower 5 hydrocarbons to produce carbon oxides free hydrogen and carbon nanotubes.

The present disclosure also relates to a process for producing a catalyst composition, the process comprising: dissolving at least two salts selected from the group con- 10 sisting of Ni, Co, Fe, Cu, and Zn in water to obtain a solution; adding a support material to the solution to obtain a paste; drying the paste at a temperature of 120° C. for a period of 1 to 48 h to obtain a dried paste; calcining the dried paste at a temperature range of 400-600° C. for a period of 15 1 to 6 h to obtain a precursor catalyst; and reducing the precursor catalyst by hydrogen and in the presence of nitrogen in a temperature range of 380-450° C. for a period of 1 to 6 h to obtain a catalyst composition.

The present disclosure further relates to a process for 20 producing carbon oxides free hydrogen and carbon nanotubes, the process comprising; placing a catalyst composition in a reactor; wherein the catalyst composition comprises of a catalyst in an amount in the range of 10% to 70% w/w of the total weight of the composition; at least one modifying 25 block diagrams herein represent conceptual views of illusagent in an amount in the range of 1% to 14% w/w of the total weight of the composition; and a support material in an amount in the range 20% to 70% w/w of the total weight of the composition; passing a flow of lower hydrocarbon in the presence of an optionally used carrier gas in the range of 30 10-100 mL/min over the catalyst composition at a temperature of 550-800° C. at atmospheric pressure; and recovering the carbon oxide free stream and carbon nanotubes wherein the stream comprising of hydrogen, unreacted lower hydrocarbon and the optionally used carrier gas.

These and other features, aspects, and advantages of the present subject matter will be better understood with reference to the following description and appended claims. This summary is provided to introduce a selection of concepts in a simplified form. This summary is not intended to identify 40 key features or essential features of the claimed subject matter, nor is it intended to be used to limit the scope of the claimed subject matter

#### BRIEF DESCRIPTION OF THE DRAWINGS

The detailed description is described with reference to the accompanying figures. In the figures, the left-most digit(s) of a reference number identifies the figure in which the reference number first appears. The same numbers are used 50 throughout the drawings to reference like features and components.

FIG. 1 graphically illustrates CDM at various reaction temperatures between 700 to 800° C. The relationship between methane conversions against reaction time is illus- 55 trated.

FIG. 2 graphically illustrates the effect of reaction temperature and GHSV (L/ $h \cdot g_{cat}$ ) on the CDM.

FIG. 3 graphically illustrates the CDM as a function of time at different temperatures on Cu(5%)Zn(5%)-Ni(60%)/ 60 Al<sub>2</sub>O<sub>3</sub>.

FIG. 4 graphically illustrates the hydrogen yield as a function of time at different temperatures over Cu(5%)Zn (5%)-Ni(60%)/Al<sub>2</sub>O<sub>3</sub>.

FIG. 5 graphically illustrates the CDM on the effect of 65 partial pressure of methane on methane conversion over Cu(5%)Zn(5%)-Ni(60%)/Al<sub>2</sub>O<sub>3</sub>.

FIG. 6 graphically illustrates the CDM on the effect of partial pressure of methane on hydrogen yield over Cu(5%) Zn(5%)-Ni(60%)/Al<sub>2</sub>O<sub>3</sub>.

FIG. 7 graphically illustrates the CDM on efficiency as a function of time for different separation cycles without any oxidation treatment.

FIG. 8 graphically illustrates the CDM on hydrogen yield as a function of time for different separation cycles without any oxidation treatment.

FIG. 9 graphically illustrates the influence of hydrogen carrier gas on the hydrogen yield on Cu(5%)Zn(5%)-Ni (60%)/Al<sub>2</sub>O<sub>3</sub> catalyst.

FIG. 10 graphically illustrates SEM micrographs of 60% Ni/Al<sub>2</sub>O<sub>3</sub> and Cu(5%)Zn(5%)-Ni(60%)/Al<sub>2</sub>O<sub>3</sub> spent catalyst after reaction at 750° C.

FIG. 11 graphically illustrates TEM micrographs showing the appearance of CNTs catalyst after a CDM run at 750° C.

FIG. 12 graphically illustrates HRTEM image of bamboo shaped multi-walled CNT illustrating wall graphite layer (arrow 1), compartment graphitic (arrow 2), after compartmentalization (arrow 3) and graphite plane distance (arrow 4) on catalyst C.

It should be appreciated by those skilled in the art that any trative systems embodying the principles of the present subject matter.

#### DETAILED DESCRIPTION

The present invention now will be described more fully hereinafter. Indeed, the invention may be embodied in many different forms and should not be construed as limited to the embodiments set forth herein; rather, these embodiments are provided so that this disclosure will satisfy applicable legal requirements. As used in the specification, and in the appended claims, the singular forms "a", "an", "the", include plural referents unless the context clearly dictates otherwise

The subject matter described herein relates to catalytic decomposition of lower hydrocarbons to produce carbon oxides free hydrogen and bamboo shaped carbon nanotubes. Carbon oxides comprises of carbon dioxide and carbon monoxide.

The objective of the present disclosure relates to a catalyst composition which is used for decomposition of lower hydrocarbons to produce carbon oxides free hydrogen and carbon nanotubes.

Yet another objective of the present disclosure relates to a catalyst composition comprising: a catalyst in an amount in the range of 10% to 70% w/w of the total weight of the composition; at least one modifying agent in an amount in the range of 1% to 14% w/w of the total weight of the composition; and a support material in an amount in the range of 20% to 70% w/w of the total weight of the composition.

An embodiment of the present disclosure provides a catalyst composition, wherein the catalyst is selected from the group consisting of Ni, Co, and Fe. The metal in the catalyst composition may be present in their elemental form or as metal oxide or as metal salt or mixtures thereof.

Another embodiment of the present disclosure provides a catalyst composition, wherein the catalyst is in an amount in the range of 50 to 70% w/w of the total weight of the composition.

In another embodiment of the present disclosure, provides a catalyst composition, wherein the modifying agent is selected from the group consisting of Cu, Zn, oxides of Cu, oxides of Zn, and mixtures thereof.

The present disclosure further relates to a catalyst composition, wherein the support material is selected from the group consisting of alumina, zirconia, titania, silica, niobia, 5 zeolites, MCM-22, mesoporous oxides and microporous oxides, preferably selected from the group consisting of  $Al_2O_3$ , SiO<sub>2</sub>, and MCM-22 in an amount in the range of 20% to 40% w/w of the total weight of the composition. The preferred support material is  $\gamma$ -alumina.

In accordance to the present disclosure, the catalyst composition, more preferably, is 60% w/w Ni, 5% w/w Cu, 5% w/w Zn and 30% w/w Al<sub>2</sub>O<sub>3</sub>. The catalyst composition depicted as  $Cu(5\%)Zn(5\%)-Ni(60\%)/Al_2O_3$  contains 30% w/w Al<sub>2</sub>O<sub>3</sub>.

In further embodiment of the present disclosure, the lower hydrocarbon is selected from the group consisting of methane, ethane, and mixtures thereof.

The present disclosure relates to a process for producing a catalyst composition, the process comprising; dissolving at 20 least two salts selected from the group consisting of Ni, Co, Fe, Cu, and Zn in water to obtain a solution; adding a support material to the solution to obtain a paste; drying the paste at a temperature of  $120^{\circ}$  C. for a period of 1 to 48 h to obtain a dried paste; calcining the dried paste at a temperature range 25 of 400-600° C. for a period of 1 to 6 h to obtain a precursor catalyst; and reducing the precursor catalyst by hydrogen and in the presence of nitrogen in a temperature range of 380-450° C. for a period of 1 to 6 h to obtain a catalyst composition. 30

An embodiment of the present disclosure relates to a process, wherein salts of Ni are selected from the group consisting of nickel nitrate, nickel chloride, nickel formate, nickel acetate and nickel carbonate. Salts of nickel can be simply any organic or inorganic metal salts containing Ni. 35 An embodiment of the present disclosure relates to a process, wherein the preferred salt of Ni is nickel nitrate.

Yet another objective of the present disclosure relates to a process, wherein salts of Co are selected from the group consisting of cobalt nitrate, cobalt chloride, cobalt formate, 40 cobalt acetate, and cobalt carbonate. Salts of Co can be simply any organic or inorganic metal salts containing Co. An embodiment of the present disclosure relates to a process, wherein the preferred salt of Co is cobalt nitrate.

In another embodiment of the present disclosure, provides 45 a process, wherein salts of Fe are selected from the group consisting of iron nitrate, iron chloride, iron formate, iron acetate, and iron carbonate. Salts of iron can be simply any organic or inorganic metal salts containing iron. An embodiment of the present disclosure relates to a process, wherein 50 the preferred salt of Fe is iron nitrate.

The salts of Cu of the present disclosure are selected from the group consisting of copper nitrate, copper hydroxide, copper carbonate, copper formate and copper acetate. Salts of copper can be simply any organic or inorganic metal salts 55 containing copper. An embodiment of the present disclosure relates to a process, wherein the preferred salt of Cu is copper nitrate.

The present disclosure further relates to a process, wherein salts of Zn are selected from the group consisting of 60 zinc acetate, zinc hydroxide, zinc formate, zinc carbonate and zinc nitrate. Salts of zinc can be simply any organic or inorganic metal salts containing zinc. An embodiment of the present disclosure relates to a process, wherein the preferred salt of Zn is zinc nitrate. 65

The support material used in the present disclosure is selected from the group consisting of alumina, zirconia, titania, silica, niobia, zeolites, MCM-22, mesoporous oxides and microporous oxides, preferably selected from the group consisting of  $Al_2O_3$ ,  $SiO_2$ , and MCM-22. An embodiment of the present disclosure relates to a process, wherein the preferred support is  $\gamma$ -alumina. Alumina is cost effective support used in the industry for preparation of catalyst.

The present disclosure further relates to a process, wherein water used for the preparation of solid gels is distilled and deionized. Any other purified form of water preferably non-ionic can also be used.

In accordance to the present disclosure, the catalyst composition is a mixed oxide with meso or micro pores with nano or microsize of metal (NiO) particles. The metal oxide (Ni) structure is confirmed by the x-ray diffraction,

In accordance to the present disclosure, the catalyst composition is a mixed oxide or oxides with macro or meso or micro pores with either ordered or disordered pore geometry with nano or microsize of metal active particles. The average NiO crystallite size is in the range from 20 to 35 nm. Small crystallites are preferred for the catalyst composition.

The present disclosure further relates to a process for producing a catalyst composition comprising; dissolving nitrate salts of Ni, Cu, and Zn in water to obtain a solution; adding alumina to the solution to obtain a paste; drying the paste at a temperature of  $120^{\circ}$  C. for a period of 24 h to obtain a dried paste; calcining the dried paste at a temperature range of 550° C. for a period of 6 h to obtain a precursor catalyst; and reducing the precursor catalyst by hydrogen in the presence of nitrogen preferably in the temperature range of 450° C. for a period of 6 h to obtain a catalyst composition.

The present disclosure further relates to a process for producing carbon oxides free hydrogen and carbon nanotubes, the process comprising; placing a catalyst composition in a reactor; passing a flow of lower hydrocarbon in the presence of an optionally used carrier gas in the range of 10-100 mL/min over the catalyst composition at a temperature of 550-800° C. at atmospheric pressure; and recovering the carbon oxide free stream and carbon nanotubes wherein the stream comprising of hydrogen, unreacted lower hydrocarbon and the optionally used carrier gas.

The present disclosure further relates to a process for producing carbon oxides free hydrogen and carbon nanotubes, the process comprising; placing a catalyst composition in a reactor; wherein the catalyst composition comprises of a catalyst in an amount in the range of 10% to 70% w/w of the total weight of the composition; at least one modifying agent in an amount in the range of 1% to 14% w/w of the total weight of the composition; and a support material in an amount in the range 20% to 70% w/w of the total weight of the composition; passing a flow of lower hydrocarbon in the presence of an optionally used carrier gas in the range of 10-100 mL/min over the catalyst composition at a temperature of 550-800° C. at atmospheric pressure; and recovering the carbon oxide free stream and carbon nanotubes wherein the stream comprising of hydrogen, unreacted lower hydrocarbon and the optionally used carrier gas.

The reactor used in the present disclosure for decomposition of lower hydrocarbons is selected from the group consisting of fixed bed tubular reactor and fluidized bed reactor, preferably from fixed bed tubular reactor.

An embodiment of the present disclosure relates to a process, wherein the lower hydrocarbon in passed over the catalyst at a rate of 10-100 mL/min with GHSV in the range of (600-12000 ml/h·g<sub>cat</sub>). The preferred GHSV is 3600 ml/h·g<sub>cat</sub>

Yet another objective of the present disclosure relates to a process, wherein the carrier gas is selected from the group comprising of nitrogen and hydrogen. A further embodiment of the present disclosure relates to a process, wherein the carrier gas is hydrogen.

The catalyst composition disclosed in the present disclosure is heated at a temperature is 700-800° C. Yet another objective of the present disclosure relates to a process, wherein the catalyst is heated at a temperature is  $750^{\circ}$  C.

In another embodiment of the present disclosure provides a process, wherein the carbon nano-tubes are bamboo-like carbon nanotubes.

In another embodiment of the present disclosure, provides a process for preferentially producing high purity bamboolike carbon nanotubes.

In another embodiment of the present disclosure, the high purity of bamboo shaped carbons is obtained with outer and inner diameter in the range of 60-55 nm and 25-30  $\mu$ m and 2-3  $\mu$ m in length.

In another embodiment of the present disclosure, wherein the catalyst composition exhibited high stability of more than 70 h at  $750^{\circ}$  C.

Yet another objective of the present disclosure relates to a process, wherein high purity bamboo shaped carbon nano- 25 tubes are separated by centrifugation.

Yet another objective of the present disclosure relates to a process, wherein the process comprising; placing a catalyst composition in fixed bed tubular reactor; wherein the catalyst composition comprises of 60% w/w Ni, 5% w/w Cu, 5% 30 w/w Zn and 30% w/w Al<sub>2</sub>O<sub>3</sub>; passing a flow of methane in the presence of a carrier gas at the rate of 20 ml/min, over the catalyst composition at a temperature of 750° C. at atmospheric pressure; and recovering the carbon oxide free stream and carbon nanotubes wherein the stream comprising 35 of hydrogen, unreacted methane and the optionally used carrier. The optionally used carrier gas can be hydrogen or nitrogen.

The following definitions and analytical instrumentation tools are used in this description of the invention:

(a) The BET (Brunauer-Emmett-Teller) surface area analyzer provides precise surface area evaluation of catalytic materials that are formed and used in this description;

(b) The SEM (Scanning electron microscope) provides precise information about the sample surface including 45 external morphology (texture), chemical composition, and crystalline structure and orientation of materials making up the sample surface;

(c) The HRTEM (High-resolution transmission electron microscopy) is an imaging mode of the transmission elec- 50 tron microscope (TEM) that allows the imaging of the crystallographic structure of a sample at an atomic scale. This method of analysis provides a way to study nano-scale properties of crystalline material;

(d) Thermo-gravimetric analysis or thermal gravimetric 55 analysis (TGA) is a method of thermal analysis in which changes in physical and chemical properties of materials are measured as a function of increasing temperature providing information about chemical phenomena including chemisorptions, dehydration, decomposition of solid-gas 60 reactions;

(e) Temperature-programmed reduction (TPR) is an analytical tool used for the characterization of metal oxides, mixed metal oxides, and metal oxides dispersed on a support material providing quantitative information on yields of the 65 reducibility of the oxide's surface, as well as the heterogeneity of the reducible surface;

(f) Carbon nanotubes (CNTs) are allotropes of carbon with a cylindrical nanostructure and have unusual properties which are valuable for nanotechnology applications in the field of electronics, optics and material science.

(g) Bamboo-like Carbon Nano-Tubes (BCNTs) are carbon nanotube structures which have bamboo like knots in the tubular structure formed.

(h) Gas hourly space velocity (GHSV=Reactant Gas Flow Rate/Reactor Volume) is a measure for relating the reactant gas flow rate to the reactor volume.

(i) Sonication is a process of applying ultra-sound energy to agitate particles in a sample.

(j) Doping is an intentional process to introduce impurities into a given pure substance so as to modify its properties.(k) Raman spectroscopy is a method to analyze the charac-

teristic of a material by the way it scatters incidental light.

The objective of the present disclosure relates to a compositions and a process for production of carbon nanotubes (CNT's) and oxide free hydrogen through decomposition of 20 lighter hydrocarbons, like methane, over a multi-metal catalyst. Under optimum reaction conditions, and with catalysts of the present subject matter, the methane is decomposed to yield oxide free hydrogen and CNT's. The hydrogen thus obtained can be used as a fuel and the CNT's, which are 25 allotropes of carbon, possess valuable applications in the field of electronics, optics and material science.

The routes proposed for production of clean hydrogen are  $NH_3$  decomposition, hydrogen splitting and catalytic decomposition of hydrocarbons, like methane. Catalytic decomposition of methane to yield pure hydrogen is an important and economic alternative to produce COx free hydrogen. This process has several advantages as compared to conventional hydrogen production. This method yields pure hydrogen at less severe conditions. Further advantage of the process is that the production of high value nano carbon simultaneously with hydrogen. The catalytic decomposition of methane can be presented as following:

#### CH<sub>4</sub>→C+2H<sub>2</sub> $\Delta$ H=75.6 kJ/mol.

40

Catalytic decomposition of methane to hydrogen based on Ni, Fe and Co based catalysts on inert support materials has been reported in the state of the art methods. In one conventional method, it has been observed that methane is decomposed to hydrogen and carbon and no carbon oxides was formed, on Ni<sub>x</sub>Mg<sub>y</sub>O (where x and y represent the mole content of Ni and Mg), when the reaction was carried out under 425-625° C.

In another implementation, the effect of Cu on catalyst composition has been studied. Cu was used as a promoter to enhance the catalytic activity of the composition Ni<sub>x</sub>Mg<sub>y</sub>-Cu<sub>z</sub>O where x, y and z showed the mole content of Ni, Mg and Cu respectively. It was observed that addition of Cu enhances the methane conversion and solid carbon yield during catalytic decomposition of methane, as compared to performing the reaction under similar optimum reaction parameters, in the absence of Cu. In an illustrative example, 78% Ni/MgO resulted maximum methane conversion and solid carbon yield 24.4% and 143 g-C/g-Cat for the life time of 17 h at 575° C. Addition of Cu promoter enhanced the maximum Methane conversion and solid carbon yield up to 46.5% and 263 g-C/g-Cat at 725° C.

In another implementation, the production of carbon nanotubes was achieved by the decomposition of hydrocarbons over solid catalyst contains Co, Mn on an inert support. A maximum, yield of 50.3 gCNT/ $g_{cat}$  was obtained for the reaction duration of 108 min at the reaction temperature of 650° C.

State of the Art methods have shown that the synthesized catalysts usually resulted in methane conversion in the range of 50-60% with lower yield of carbon. Apart from it, the synthesized catalyst deactivates at a faster rate due to higher amorphous carbon deposition on the surface of the catalyst. <sup>5</sup> This results in deactivation of the catalyst and the regeneration of the deactivated catalyst is conventionally achieved by combustion of the carbon deposits that lead to oxide production.

The present study mainly aims at developing a catalyst composition, process analysis as well as kinetic study for methane decomposition to produce high yield of COx-free hydrogen along with carbon nanomaterials.

According to an implementation of the present subject 15 matter, the catalyst composition comprising a support material, a mixed metal oxide with active components of Fe, Ni, Co and a modifying agent, where the modifying agent can include either oxides of Cu, Zn, K etc., or derivatives of Cu or Zn or a combination of both. In said implementation, the 20 purpose of using Cu or Zn oxides or derivatives or a combination of both as promoters is to enhance the catalytic activity of the composition. Further, the catalytic support can include one of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> or MCM-22. The mixed metal oxides can include oxides of various active metals like Ni, 25 Co and Fe in various oxidation states.

In one implementation, the catalysts of the present disclosure can be represented by Ni<sub>x</sub>Cu<sub>y</sub>Zn<sub>z</sub> over an Al<sub>2</sub>O<sub>3</sub> support, wherein the x, y and z reflect the weight percentage of Ni, Cu and Zn respectively. The weight percentage of Ni, Cu and Zn ranged from 50-70% for Ni and 3-7% for Cu and 3-7% for Zn. However, the catalyst was found to be most effective when the percentage weights of Ni, Cu and Zn were 60%, 5% and 5% respectively. The purpose of addition of 35 small amount of Cu is to enhance the reducibility of dispersed Ni<sup>2+</sup> species and the thermal stability of catalyst. One of the preferred methods for preparing the catalyst Ni<sub>x</sub>- $Cu_{\nu}Zn_{z}$  over an Al<sub>2</sub>O<sub>3</sub> support of the present subject matter is by co-precipitation method. In the said method, co- 40 precipitated catalysts were synthesized by co-precipitation of their respective nitrates in an aqueous solution and the precipitates subsequently washed, dried and calcined at 550° С.

In another method, the catalyst was prepared by wet 45 impregnation method. In the said method, wet impregnated catalysts were prepared by impregnating metals precursor (Ni, Cu and Zn) on  $\gamma$ -Alumina (BET surface area=197 m<sup>2</sup>/g) in a rotary vacuum evaporator for 2 h followed by dying and calcination at 550° C. for 5 h to promote thermal decomposition of nitrates into their respective oxides. The method of the preparation of the catalyst can be achieved with other preparation methods such as anchoring, deposition, precipitation, grafting, chemical vapor deposition etc that are 55 available in the state of the art.

In one implementation, the catalyst composition of the present subject matter is capable of producing substantially carbon oxide-free hydrogen and substantially pure carbon by the decomposition of methane over a relatively low tem-60 perature range of 550° to 800° C. The stability of the catalytic composition to perform methane decomposition is evaluated under different conditions of temperature ranging from 550-800° C. and further, the stability was also evaluated under varied pressure conditions. Further, the CNT's 65 thus obtained upon catalytic decomposition of methane, that are deposited on the surface of the catalyst are separated

from the catalyst using instrumentation techniques like sonication, centrifugation, etc.

#### EXAMPLES

The following examples are given by way of illustration of the present invention and should not be construed to limit the scope of present disclosure. It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory only and are intended to provide further explanation of the claimed subject matter.

The activity of catalysts for methane conversion and hydrogen yield was compared in a fixed-bed quartz reactor (i.d.=1.9 cm, o.d.=2.1 cm and length=60 cm) in a conventional down flow mode. The catalyst (1.0 g) was loaded in the reactor, and then packed with small amount of inert to prevent the channeling. A chrome-alumina-type thermocouple was placed into the annular space between the reactor, and the furnace to minimize the temperature difference between the catalyst bed and the thermocouple. The feed was introduced through a mass flow controller (Bronkhorst High-Tech, Netherlands) by passing the reactant gas mixture of  $CH_4/N_2$  over the catalyst bed. Prior to activity tests, all calcined samples in the oxidized state were reduced in-situ with a total volumetric flow rate of 60 ml/min in a mixture N<sub>2</sub> and H<sub>2</sub> (1:1 ratio) at 450° C. for 5 h. Initially, pure nitrogen was allowed into the reactor, to create an inert atmosphere in the reactor. Catalytic CH<sub>4</sub> decomposition was performed at atmospheric pressure by passing a flow of methane along with N<sub>2</sub> or H<sub>2</sub> in the range of 10-100 mL/min and reaction temperature at 550-800° C. The composition of the outlet gas from the reactor was determined by Nucon-5700 gas chromatograph (GC), equipped with a thermal conductivity detector, a carbosphere column (80-100 mesh) with argon as the carrier gas.

#### Example 1: Fabrication and Growing of Bamboo Shaped Carbon-Nanotubes (BCNT's)

Experiments were performed to investigate the effect of reaction temperature and gas hourly space velocity (GHSV,  $h^{-1}$ ) on methane conversion and bamboo shaped carbon nano Tubes (BCNTs) formation at experimental conditions. The blank experiment was also carried out in the reactor filled with quartz particles with a feed consisting of methane and nitrogen in a ratio of 1:1 at a GHSV of 3.6 L/h·g<sub>cat</sub> and in the temperature range of 550-800° C. The conversion of methane was less than 2% in all of these experiments, revealed that the methane decomposition was negligible without catalyst in the above temperature ranges.

To explore the catalytic stability of the catalyst, methane conversion as a function of time on stream was measured at reaction temperatures of 700, 750 and 800° C. and the results are reported in FIG. 1. It can be seen that the initial methane conversion was increased with the reaction temperature but the stability of the catalyst decreased at higher temperature (800° C.). The highest conversions of methane at three different temperatures were 47%, 62%, and 70%, respectively. It was also observed that the catalyst was fairly stable at 700 and 750° C., and 45% and 57% methane conversion could be obtained steadily for approximately 15 h. However, at 800° C., catalyst started declining in its catalytic activity just after 5 h on stream, indicating that the catalyst lost its activity and stability at high temperatures due to sintering of Ni particle and catalysts deactivation.

35

50

Example 2: Kinetics of Decomposition Reaction

Preliminary experiments were carried out at different GHSV and particle size to eliminate mass transfer and diffusional resistance. The absence of diffusional resistance <sup>5</sup> was confirmed estimated by Weisz-Prater criterion. According to this criterion, in the absence of pore diffusion resistance, the following condition has to be satisfied:

$$\varphi = \frac{r_0 \rho R_p^2 \tau}{\epsilon D_{CH4-N2} C_S} < 1 \tag{1}$$

Where,  $r_0$  is the initial rate of the reaction,  $\rho$  is the apparent 15 density,  $R_p$  is the radius of the catalyst particle,  $\tau$  is the tortuosity,  $\epsilon$  is the bed porosity. In the present system, the average values for  $\tau$  and  $\epsilon$  were taken to be 3 and 0.62, respectively. Apparent density  $\rho$  was estimated to be 0.2  $g_{cat}$ /cm<sup>3</sup>,  $R_p$  was determined to be 1.5 mm,  $D_{CH_4 \cdot N_2}$  is the <sup>20</sup> diffusion coefficient of the methane-nitrogen mixture and C<sub>s</sub> is the methane concentration at the external surface of the catalyst particle. For all the experiments, Weisz modulus is <1 which demonstrates that there is no diffusional limitation 25 in the catalyst bed. The GHSV of methane distinctly affected the catalytic performance of Ni-Cu-Zn/Al<sub>2</sub>O<sub>3</sub> catalyst in the temperature range of 600-800° C. (FIG. 2). The maximum methane conversion was obtained at 3.6 L/h·g<sub>cat</sub> due to higher residence time in the reactor. Conversion increased 30 significantly (26-70%) with increasing the reaction temperature from 550°-800° C. at constant GHSV (3.6 L/h·g<sub>cat</sub>) and similar trends were also observed at other GHSV.

#### Example 3: Effect of Reaction Temperature on BCNT's Structure

The morphology and size of the nano-tubes formed on 60% Ni-5% Cu-5% Zn-30%  $Al_2O_3$  catalyst was investigated by increasing the reaction temperature from 550-800° C. at <sup>40</sup> GHSV 3.6 L/h·gcat. FIG. **2** shows the methane conversion with respect to increase in GHSV. In addition to bamboo shaped carbon nano-tubes, some amorphous carbon was also found on the catalysts which may be due to the lower catalytic activity of the nickel particles in which the, carbon <sup>45</sup> was poorly interacted with the nickel catalyst.

## Example 4: Effect of Temperature on Methane Decomposition

The nanoporous 60% Ni-5% Cu-5% Zn-30% Al<sub>2</sub>O<sub>3</sub> catalyst was prepared by wet impregnation method. Prerequisite amounts of the transition metal nitrates (Ni:Cu:Zn, 60:5:5) were dissolved in distilled de-ionized water with the 55 required wt % of alumina. The final paste was then dried overnight in an oven and subsequently calcined at 550° C. For each experiment, catalyst was placed in the center of reactor and then purged with nitrogen for 30 min, to ensure the inert atmosphere in the reactor. The catalyst was reduced 60 insitu with hydrogen for 5-6 h at temperature of 450° C. Experiments were carried out to explore the relationship between methane conversion and reaction temperature varied in a range of 730 to 770° C. Total input gas flow rate was fixed at the optimum value of 40 mL/min. Accordingly, gas 65 hourly space velocity (GHSV) was 2.4 L/h·g<sub>cat</sub>. It was observed that even in the closer range, the optimum tem-

perature at which maximum methane conversion and hydrogen production were obtained was 750° C. (FIGS. **3** and **4**).

Example 5: Effect of Partial Pressure on Methane

Experiments were performed to investigate the effect of wider range of partial pressure of methane on methane conversion and CNTs formation at experimental conditions. Effect of partial pressure of methane  $(P_{CH4})$  on methane conversion and hydrogen yield was investigated by varying the methane partial pressure from 0.1 to 0.8 atm at 750° C. This was accomplished by conducting the experiments at different flow rates of methane and nitrogen and at different space time (W/FAo=616.33-4930.66 g-cat min/mole of methane). The effect of different methane partial pressures on methane conversion and hydrogen yield is shown in FIGS. 5 and 6. An increase in partial pressure  $(P_{CH4})$ lowered the methane conversion due to decrease in the residence time of methane in the reactor. The maximum conversion (81%) and hydrogen yield (91%) was obtained for a space time of 2465.33 g-cat min/mole of methane and  $P_{CH4}0.2$ . It was concluded that the optimum partial pressure would be in between 0.2 and 0.3 which is in accordance with our previous results. An increase of partial pressure of methane (from 0.25 to 1.0 atm) is also unfavorable for the TCD reaction. Regeneration study was carried out on 60% Ni-5% Cu-5% Zn-30% Al<sub>2</sub>O<sub>3</sub> catalyst at 750° C. for up to four regeneration cycles. It was observed that no significant loss of activity (93 to 85%) occurred during repeated decomposition/regeneration cycles.

When the reaction temperature was increased to  $750^{\circ}$  C., the bamboo-shape of the CNTs was fully developed. In this condition, amorphous carbon was again observed, but this time the formation of amorphous carbon was probably due to the decomposition of carbon on the CNTs. At  $750^{\circ}$  C., short length bamboo-shaped tubes were produced with some carbon encapsulated metal particles. Further, it was also noticed that a larger diameter CNTs are synthesized at higher reaction temperature. This was mainly due to the aggregation of the nickel particles at high temperatures, leading to the formation of larger catalyst particles and hence contributing CNTs of larger diameter. The BCNTs produced consist of the nano-tubes with the average outer diameters of 30 and 35 nm, at 700 and 750° C. respectively, while at 800° C., the average outer diameter CNTs raised to 50 nm.

#### Example 6: Methane Decomposition after Successive Separation of Nano-Fibers

Reactions were conducted and hydrogen yield was observed for different regenerative cycles after nano-fiber separation from the catalyst without further treatment ie., after sonic separator (without using TGA). The effect of separation of carbon nano-fibers without the burning of amorphous carbon for four consecutive cycles was investigated on methane conversion efficiency. The reaction was carried out at optimum conditions (750° C. and 0.25 partial pressure) for 5 h in each cycle. After each cycle, the catalyst was separated from carbon nano-fibers using sonication for 15 min and the reaction was again carried out on the separated catalyst. This was repeated for three more cycles. The decrease in the methane conversion efficiency and hydrogen yield are given in FIGS. 7 and 8 respectively.

It is seen from the plots that the conversion efficiency is decreasing gradually until 3 cycles, after which there is a steep decrease in conversion efficiency of the catalyst which

40

falls down to 5% approximately. Hydrogen yield is also found to be following the same decrement pattern to around 10%.

#### Example 7: Centrifugation as a Separation Process of Carbon Nano-Fibres

A high speed cooling centrifuge (~18000 rpm, ~35000 g) was used as the separation equipment in addition to ultrasonic agitator to see if separation efficiency could be <sup>10</sup> improved further. The spent catalyst after initial sonication using ethanol as a solvent is decanted and the supernatant solution containing carbon nanofibers is collected. It is further separated in the centrifuge operated at 15000 rpm for 30 min at 20° C. The centrifuged solution is again decanted <sup>15</sup> and the supernatant solution consisting of CNTs is dried in the oven for 6 h and the catalyst residue for 0.5 h at 100° C. The separated catalyst residue can further be sent for complete regeneration from amorphous carbon or used in the reaction without further processing as required. <sup>20</sup>

#### Example 8: Influence of Hydrogen as a Carrier Gas

The study on the effect of hydrogen instead of nitrogen as a carrier gas was carried out to avoid additional separation <sup>25</sup> of product hydrogen gas was carried out. The advantage of this process is that the same hydrogen produced in the reaction can also be used as the carrier gas by recycling. The flow rates of hydrogen were varies from 13 to 40 ml/min and methane from 10 to 48 ml/min for attaining partial pressure <sup>30</sup> of 0.2 to 0.8. The highest methane conversion was obtained for the partial pressure of methane 0.8. This result is different to the result of N<sub>2</sub> gas as carrier where the maximum conversion of methane was obtained at 0.2 partial pressure of methane. The reaction was conducted at 750° C. <sup>35</sup> at atmospheric pressure over 60% Ni-5% Cu-5% Zn-30% Al<sub>2</sub>O<sub>3</sub> catalyst. Each data point was recorded after the reaction time of 6 h (FIG. **9**).

#### Example 9: Test Results

SEM and TEM images of deactivated catalysts revealed that the deposited carbon is filamentous in nature and the Ni has occupied at the tip of the filaments, and the length and nature of the filaments varied with the catalyst compositions. 45 SEM micrographs of 60% Ni/Al $_2O_3$  and 60% Ni-5% Cu-5% Zn-30% Al<sub>2</sub>O<sub>3</sub> spent catalyst after reaction at 750° C. (FIG. 10). TEM and HTREM images confirmed the high purity bamboo-shaped structure of CNTs produced over 60% Ni-5% Cu-5% Zn-30%  $Al_2O_3$  catalyst with outer and inner 50 diameters in the range of 35-40 nm and 10-15 nm, respectively and 2-3 µm in lengths (FIGS. 11 and 12). The filamentous nature of this carbon was also confirmed by Raman spectroscopy. Approximately 84% of the carbon was deposited on the 60% Ni-5% Cu-5% Zn-30% Al<sub>2</sub>O<sub>3</sub> catalyst, 55 which was comparatively higher than rest of the catalysts. The thick walled CNTs prevented nickel inside the tubes from burning out and the final residual weight percentage remained after TGA treatment was mainly due to the weight of the nickel. Furthermore, FTIR shows the presence of 60 some metal and hydroxyl groups (O-H) impurities present on the as-grown CNTs.

The effect of temperature on the morphology and size of the nano-tubes formed was also investigated by increasing the reaction temperature from 700-800° C. at GHSV 600 65 mL/h·gcat. It was noticed that a larger diameter CNTs are synthesized at higher reaction temperature. This was mainly

due to the aggregation of the nickel particles at high temperatures, leading to the formation of larger catalyst particles which produces CNTs of larger diameter. It was observed that the BCNTs produced consists of the nano-tubes with the average inner diameters of  $23\pm 2$  and  $30\pm 2$  nm, at 700 and 750° C. respectively, while at 800° C., the average inner diameter CNTs increased to  $45\pm 2$  nm.

Example 10: Regeneration Study of the Catalyst

The biggest challenge for hydrogen production by catalytic cracking is the regeneration of the spent catalyst. Catalyst deactivation during the course of a process is often expected. Carbon may plug the pore, such that access is denied for the reactants to many crystallites inside these pores. In extreme cases, strong carbon filaments accumulate in pores to the extent that they exert stress leading to fracture of the support material and ultimately causing disintegration of catalyst pellets and plugging of reactor voids. The revers-<sup>20</sup> ibility of the catalyst activity is a very important factor for catalysts. Therefore the interest in these catalysts lies not only in their high catalytic activity, but also in the possibility of regenerating them several times so that their "lifetime" is compatible with the cost of their production. In fact, catalyst regeneration may be futile in view of the changes caused to the catalyst support structure as a result of the filament growth process for methane decomposition. Thus if the decomposition of methane is to be utilized for the production of hydrogen in a continuous process, catalysts with high activity and the possibility of regenerating them several times is needed.

Oxidation  $(C+O_2\rightarrow CO_2)$  was conducted to regenerate the catalyst activity after reaction. The oxygen regeneration was continued until no  $CO_x$  was detected in the products via GC (usually after a period of 4 h). There was slight loss of activity (approximately ±5%) during regeneration cycle. After switching from oxygen to methane, there was a drastic increase in carbon oxides being observed with their concentration declining rapidly during the first 4-5 min of the decomposition reaction. Metal oxides are formed during the regeneration cycle. The formation of carbon oxides at the beginning of the decomposition step could have been produced by the methane interaction with metal oxides and oxygen that adsorbed on the catalyst surface. The hydrogen yields were slightly decreased for all catalysts.

#### Example 11: Change in the Mass of the Sample as a Function of Temperature

TGA is used to measure changes in the mass of the sample as a function of temperature in a controlled air atmosphere. The mass of the spent catalyst was monitored, against time and temperature in the air using NETZSCH TG 209 F3 instrument with heating rate of 10° C./min. The sample was continuously weighed while being heated to 800° C. in an oxidative atmosphere to study thermal stability of CNTs and the amount of metal impurities. For all catalyst, significant weight loss started at 550° C. and the weight loss continued to increase rapidly at higher temperature, until a stable plateau region appeared around 700° C. 88-90 wt % occur around 550° C. A higher oxidation temperature of BCNTs above 550° C. can be ascribed to the purer, less defective CNT samples. A similar observation was also reported in the literature in the temperature range 500-650° C. and attributed to the oxidation of multiwall CNT (Mordkovich, 2007). The residual weight was observed to be 21-25 wt % that implies 65-85% of the mass on the spent catalyst is carbon.

30

50

65

The formation of carbon is also confirmed by XRD patterns. In addition, a minor weight loss was observed at temperature below 500° C. in the TGA spectrum corresponding to a negligible amount of amorphous carbon in catalyst.

#### Example 12: Separation of Carbon Nano-Fibers from the Catalyst

A high speed cooling centrifuge (~18000 rpm, ~35000 g) was used as the separation equipment in addition to ultra- 10 sonic agitator to see if separation efficiency could be improved further. The spent catalyst after initial sonication using ethanol as a solvent is decanted and the supernatant solution containing carbon nanofibers is collected. It is further separated in the centrifuge operated at 15000 rpm for 15 consisting of methane, ethane, and mixtures thereof. 30 mins at 20° C. The centrifuged solution is again decanted and the supernatant solution consisting of CNTs is dried in the oven for 6 h and the catalyst residue for 0.5 h at 100° C. The separated catalyst residue can further be sent for complete regeneration from amorphous carbon or used in the 20 reaction without further processing as required.

Advantages Gained in the Example Illustrative Process in this Subject Matter:

The above mentioned implementation examples as described on this subject matter and its equivalent thereof 25 have many advantages, including those which are described below:

- 1. Ni<sub>x</sub>Cu<sub>y</sub>Zn<sub>z</sub> on alumina catalyst resulted in greater than 90% methane conversion (greater than 85% hydrogen vield) at 750° C.
- 2. The addition of small amounts of Cu as modifying agent (5 wt %) increased the conversion of methane from 78 to 82% at the reaction temperature of 750° C.
- 3. Addition of both Cu and Zn modifying agent (5 wt %) on highest methane conversion compared to other catalysts, where conversion was more than 86%.
- 4. The high purity CNTs were recovered using sonication method.
- 5. The Ni<sub>x</sub>Cu<sub>y</sub>Zn<sub>z</sub> on alumina catalyst exhibited a life time  $_{40}$ of the catalyst in the range of 70-75 h when operated at 750° C.

Although the subject matter has been described in considerable detail with reference to certain examples and implementations thereof, other implementations are pos- 45 sible. As such, the spirit and scope of the appended claims should not be limited to the description of the preferred examples and implementations contained therein.

#### We claim:

1. A catalyst composition comprising:

- a catalyst in an amount in a range of 60% to 70% w/w of a total weight of the composition, the catalyst being selected from the group consisting of Ni, Co, and Fe;
- at least one first modifying agent in an amount in a range 55 of 3% to 7% w/w of the total weight of the composition, the first modifying agent being selected from the group consisting of Cu, oxides of Cu, and mixtures thereof;
- at least one second modifying agent in an amount in a range of 3% to 7% w/w of the total weight of the 60 composition, the second modifying agent being selected from the group consisting of Zn, oxides of Zn, and mixtures thereof; and
- a support material in an amount in a range of 20% to 40% w/w of the total weight of the composition.

2. The catalyst composition as claimed in claim 1, wherein the support material is selected from the group consisting of alumina, zirconia, titania, silica, niobia, zeolites, MCM-22, mesoporous oxides and microporous oxides.

3. The catalyst composition as claimed in claim 1, wherein the catalyst composition is 60% w/w Ni, 5% w/w Cu, 5% w/w Zn, and 30% w/w Al<sub>2</sub>O<sub>3</sub>.

4. The catalyst composition as claimed in claim 1, wherein the support material is selected from the group consisting of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, and MCM-22.

5. The catalyst composition as claimed in claim 1, wherein the catalyst composition is capable of catalyzing decomposition of lower hydrocarbons to produce carbon oxides-free hydrogen and carbon nanotubes.

6. The catalyst composition as claimed in claim 5, wherein the lower hydrocarbons are selected from the group

7. A catalyst composition comprising:

- a catalyst in an amount in a range of 50% to 70% w/w of a total weight of the composition;
- at least one first modifying agent in an amount in a range of 3% to 7% w/w of the total weight of the composition. the first modifying agent being selected from the group consisting of Cu, oxides of Cu, and mixtures thereof;
- at least one second modifying agent in an amount in a range of 3% to 7% w/w of the total weight of the composition, the second modifying agent being selected from the group consisting of Zn, oxides of Zn, and mixtures thereof; and
- a y-alumina support material in an amount in a range of 20% to 40% w/w of the total weight of the composition.

8. The catalyst composition as claimed in claim 7, wherein:

the catalyst is selected from the group consisting of Ni, Co, and Fe.

9. The catalyst composition as claimed in claim 7, Ni/Al2O3 catalyst increased the conversion and conferred 35 wherein the catalyst composition is 60% w/w Ni, 5% w/w Cu, 5% w/w Zn, and 30% w/w y-alumina.

> 10. A process for producing a catalyst composition comprising a catalyst in an amount in a range of 10% to 70% w/w of a total weight of the composition, at least one modifying agent in an amount in a range of 1% to 14% w/w of the total weight of the composition, and a support material in an amount in a range of 20% to 70% w/w of the total weight of the composition, the process comprising:

dissolving at least two salts selected from the group consisting of Ni, Co, Fe, Cu, and Zn in water to obtain a solution:

adding a support material to the solution to obtain a paste: drying the paste at a temperature of 120° C. for a period of 1 h to 48 h to obtain a dried paste;

- calcining the dried paste at a temperature range of 400° C.-600° C. for a period of 1 h to 6 h to obtain a precursor catalyst; and
- reducing the precursor catalyst by hydrogen and in the presence of nitrogen in a temperature range of 380° C.-450° C. for a period of 1 h to 6 h to obtain a catalyst composition.

11. The process as claimed in claim 10, wherein salts of Ni are selected from the group consisting of nickel nitrate, nickel chloride, nickel formate, nickel acetate, and nickel carbonate.

12. The process as claimed in claim 10, wherein salts of Co are selected from the group consisting of cobalt nitrate, cobalt chloride, cobalt formate, cobalt acetate, and cobalt carbonate.

13. The process as claimed in claim 10, wherein salts of Fe are selected from the group consisting of iron nitrate, iron chloride, iron formate, iron acetate, and iron carbonate.

14. The process as claimed in claim 10, wherein salts of Cu are selected from the group consisting of copper nitrate, copper hydroxide, copper carbonate, and copper acetate.

**15**. The process as claimed in claim **10**, wherein salts of Zn are selected from the group consisting of zinc acetate, 5 zinc hydroxide, zinc formate, zinc carbonate, and zinc nitrate.

**16**. The process as claimed in claim **10**, wherein the support material is selected from the group consisting of alumina, zirconia, titania, silica, niobia, zeolites, MCM-22, 10 mesoporous oxides and microporous oxides.

**17**. The process as claimed in claim **10**, wherein the catalyst composition is a mixed oxide with meso or micro pores with nano or microsize of metal (NiO) particles.

18. The process as claimed in claim 10, wherein the 15 catalyst composition is 60% w/w Ni, 5% w/w Cu, 5% w/w Zn, and 30% w/w  $Al_2O_3$ .

19. The process as claimed in claim 10, wherein the support material is selected from the group consisting of  $Al_2O_3$ ,  $SiO_2$ , and MCM-22. 20

\* \* \* \* \*

## (12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

## (19) World Intellectual Property

Organization

International Bureau



(43) International Publication Date 27 November 2014 (27.11.2014)

- (51) International Patent Classification: Bθ1J 37/03 (2006.01) Bθ1J 37/02 (2006.01) Bθ1J 21/04 (2006.01) B82Y 40/00 (2011.01) Bθ1J 23/80 (2006.01)
- (21) International Application Number: PCT/IN2013/000460
- (22) International Filing Date: 23 July 2013 (23.07.2013)
- (25) Filing Language: English
- (26) Publication Language: English
- (30) Priority Data: 1852/MUM/2013 24 May 2013 (24.05.2013) IN
- (71) Applicants: HINDUSTAN PETROLEUM CORPORA-TION LTD. [IN/IN]; Petroleum House, 17 Jamshedji Tata Road, Churchgate, Mumbai 400 020 (IN). INDIAN IN-STITUTE OF TECHNOLOGY, DELHI [IN/IN]; Hauz Khas, New Delhi 110 016 (IN). CENTRE FOR HIGH TECHNOLOGY [IN/IN]; OIDB Bhawan, Tower 'A', 9th Floor, Plot No. 2, Sector- 73, Noida 201 301 (IN).
- (72) Inventors: PANT, Kamal Kishore; Indian Institute Of Technology Delhi (IIT Delhi), Hauz Khas, New Delhi 110 016 (IN). SARASWAT, Sushil Kumar; Indian Institute Of Technology Delhi (IIT Delhi), Hauz Khas, New Delhi 110 016 (IN). TOMPALA, Annaji Rajiv Kumar; Hindustan Petroleum Corporation Limited, Corporate R & D Centre, 176, Adarsh Eco Place, 1st Floor, EPIP Zone II, Whitefield Bangalore 560 066 (IN). RAMESH, Kanaparthl; Hindustan Petroleum Corporation Limited, Corporste R & D Centre, 176, Adarsh Eco Place, 1st Floor, EPIP Zone II, Whitefield Bangalore 560 066 (IN). PEDDY, Venkata Chalapathi Rao; Hindustan Petroleum Corporation Limited, Corporate R & D Centre, 176, Adarsh Eco Place, 1st Floor, EPIP Zone II, Whitefield Bangalore 560 066 (IN). NETTEM, Venkateswarlu Choudary; Hindus-

2014/188439 A1

## (10) International Publication Number WO 2014/188439 A1

tan Petroleum Corporation Limited, Corporate R & D Centre, 176, Adarsh Eco Place, 1st Floor, EPIP Zone II, Whitefield Bangalore 560 066 (IN). GANDHAM, Sri Ganesh; Hindustan Petroleum Corporation Limited, Corporate R & D Centre, 176, Adarsh Eco Place, 1st Floor, EPIP Zone II, Whitefield Bangalore 560 066 (IN).

- (74) Agents: RAE, Konpal et al.; B-6/10, Safdarjung Enclave, New Delhi 110 029 (IN).
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

#### Declarations under Rule 4.17:

of inventorship (Rule 4.17(iv))

### Published:

with international search report (Art. 21(3))

(54) Title: CATALYTIC DECOMPOSITION OF LOWER HYDROCARBONS TO PRODUCE CARBON OXIDES FREE HY-DROGEN AND BAMBOO SHAPED CARBON NANOTUBES

(57) Abstract: In accordance with the present subject matter there is provided a process for catalytic decomposition of lower hydrocarbons to produce carbon oxides free hydrogen and bamboo shaped carbon nanotubes over a catalyst composition. The process for catalytic decomposition of lower hydrocarbons comprises contacting lower hydrocarbon over a catalyst composition, where the catalyst composition comprising, a catalyst, at least one modifying agent and a support material.



## OFFICIAL JOURNAL OF THE PATENT OFFICE

| निर्गमन सं. 07/2020 | शुक्रवार | दिनांक: 14/02/2020 |
|---------------------|----------|--------------------|
| ISSUE NO. 07/2020   | FRIDAY   | DATE: 14/02/2020   |

## पेटेंट कार्यालय का एक प्रकाशन PUBLICATION OF THE PATENT OFFICE

The Patent Office Journal No. 07/2020 Dated 14/02/2020

#### (12) PATENT APPLICATION PUBLICATION

(19) INDIA

(22) Date of filing of Application :13/08/2018

(54) Title of the invention : SLOTTING MACHINE

(43) Publication Date : 14/02/2020

| (51)   | (71)Name of Applicant :               |
|--|---------------------------------------|
| International :B23Q0011000000,B23D0049160000,B23Q0001480000,H04W0052240000,B23D000706000 | 0 1)Quantum University                |
| classification   | Address of Applicant :22km Mile       |
| (31) Priority  | Stone, Mandawar, Roorkee- D.Dun       |
| Document :NA   | Highway (NH-73), Roorkee - 247167,    |
| No   | Uttarakhand, India. Uttarakhand India |
| (32) Priority NA   | (72)Name of Inventor :                |
| Date   | 1)Mohd Vaseem                         |
| (33) Name  | 2)Mohd Jarrar                         |
| of priority :NA  | 3)Gulshan Chauhan                     |
| country  | 4)Chanderkant                         |
| (86)   | 5)Kaushik Karan                       |
| International  |                                       |
| Application :NA  |                                       |
| No :NA   |                                       |
| Filing   |                                       |
| Date   |                                       |
| (87)   |                                       |
| International NA   |                                       |
| Publication  |                                       |
| No   |                                       |
| (61) Patent  |                                       |
| of Addition  |                                       |
| ίο ·NΔ   |                                       |
| Application NA   |                                       |
| Number   |                                       |
| Filing   |                                       |
| Date   |                                       |
| (62)   |                                       |
| Divisional to  |                                       |
| Application :NA  |                                       |
| Number :NA   |                                       |
| Filing   |                                       |
| Date   |                                       |

(57) Abstract :

The present invention relates to a slotting machine fabricated from mild steel material, comprising a ram 4 for reciprocating a tool 2 in vertical directions, a motor 7 to provide power to the machine, a cutting tool 2 for machining the work pieces 1, a tool holder 9 for clamping of cutting tool 2 , a worm gear 5 for speed reduction, a spur gear 6 for transmission of power, a table 8 for holding the work pieces 1, a work piece holder 10 for clamping the work piece 1 in position, a cross slide 3 on which the work piece 1 moves in longitudinal and transverse direction, a frame for supporting all the aforementioned elements.



No. of Pages : 12 No. of Claims : 9



## OFFICIAL JOURNAL OF THE PATENT OFFICE

| निर्गमन सं. 32/2020      | शुक्रवार | दिनांक: 07/08/2020 |
|--------------------------|----------|--------------------|
| <b>ISSUE NO. 32/2020</b> | FRIDAY   | DATE: 07/08/2020   |

## पेटेंट कार्यालय का एक प्रकाशन PUBLICATION OF THE PATENT OFFICE

The Patent Office Journal No. 32/2020 Dated 07/08/2020

## **INTRODUCTION**

In view of the recent amendment made in the Patents Act, 1970 by the Patents (Amendment) Act, 2005 effective from 01<sup>st</sup> January 2005, the Official Journal of The Patent Office is required to be published under the Statute. This Journal is being published on weekly basis on every Friday covering the various proceedings on Patents as required according to the provision of Section 145 of the Patents Act 1970. All the enquiries on this Official Journal and other information as required by the public should be addressed to the Controller General of Patents, Designs & Trade Marks. Suggestions and comments are requested from all quarters so that the content can be enriched.

## (Om Prakash Gupta) CONTROLLER GENERAL OF PATENTS, DESIGNS & TRADE MARKS

7<sup>TH</sup> AUGUST, 2020

(12) PATENT APPLICATION PUBLICATION

(22) Date of filing of Application :13/08/2018

(43) Publication Date : 07/08/2020

## (54) Title of the invention : BIOMETRIC BASED AIRCRAFT SYSTEM WITH REMOTE CONTROL

| (51) International classification                               | :A63H0027000000,<br>B64C0039020000,<br>H04N0021440800,<br>H02P0005740000,<br>H05K0005020000 | <ul> <li>(71)Name of Applicant :</li> <li>1)Quantum University<br/>Address of Applicant :22km Mile Stone, Mandawar,Roorkee-<br/>D.Dun Highway (NH-73), Roorkee - 247167, Uttarakhand, India.<br/>Uttarakhand India</li> </ul> |
|---|---|---|
| (31) Priority Document No                                       | :NA   | (72)Name of Inventor :  |
| (32) Priority Date  | :NA   | 1)Gulshan Chauhan   |
| (33) Name of priority country                                   | :NA   | 2)Mohd Vaseem   |
| (86) International Application No                               | :NA   | 3)Kumar Satyam  |
| Filing Date   | :NA   | 4)Abhishek Gupta  |
| (87) International Publication No                               | : NA  | 5)Aman Kumar  |
| (61) Patent of Addition to Application<br>Number<br>Filing Date | :NA<br>:NA  |   |
| (62) Divisional to Application Number                           | :NA   |   |
| Filing Date   | :NA   |   |

#### (57) Abstract :

The present invention relates to a system for controlling aircrafts comprising, a transmitter for transmitting a control signal that is given by a user, a receiver for receiving the control signal, a fuselage for housing various components and is made of a light weight material, a biometric sensor for verifying identity of the user and generating a command signal upon successful verification, a control unit mounted inside the fuselage for receiving control and command signal to supply electrical power from a battery, at least one BLDC motor with a propeller to provide thrust upon receiving the electrical power, and multiple servo motors connected to the control unit for controlling the yaw, pitch and roll of the fuselage.



No. of Pages : 11 No. of Claims : 8

(12) PATENT APPLICATION PUBLICATION

(21) Application No.201811030434 A

(19) INDIA

(22) Date of filing of Application :13/08/2018

(43) Publication Date : 07/08/2020

|  | :B62K0011000000, | (71)Name of Applicant :                                     |
|--|------------------|---|
|  | G05D0001020000,  | 1)Quantum University  |
| (51) International classification                | B62B0005000000,  | Address of Applicant :22km Mile Stone, Mandawar, Roorkee-   |
|  | B62D0037000000,  | D.Dun Highway (NH-73), Roorkee - 247167, Uttarakhand, India |
|  | A63H0017000000   | Uttarakhand India   |
| (31) Priority Document No                        | :NA              | (72)Name of Inventor :                                      |
| (32) Priority Date                               | :NA              | 1)Chanderkant Vaid  |
| (33) Name of priority country                    | :NA              | 2)Gulshan Chauhan   |
| (86) International Application No                | :NA              | 3)Rajneesh Raghav   |
| Filing Date                                      | :NA              | 4)M. Kannan   |
| (87) International Publication No                | : NA             | 5)Mohd Vaseem   |
| (61) Patent of Addition to Application Number:NA |                  |   |
| Filing Date                                      | :NA              |   |
| (62) Divisional to Application Number            | :NA              |   |
| Filing Date                                      | :NA              |   |

### (54) Title of the invention : SOLAR POWERED AUTONOMOUS VEHICLE

(57) Abstract :

The present invention relates to a solar powered autonomous one wheeled vehicle comprising of a frame 1 including a first foot placement section 2 and a second foot placement section 3; a wheel 5 disposed between the foot placement sections; shock absorbers 8 engaged to the wheels; solar panels 10 coupled to the frame for generating power; a battery 12 to store the power; a motor for rotating the wheel; a motor controller 11 for self stabilization of the vehicle, wherein the controller comprises of a sensor module to measure orientation information of the vehicle and a motor driver to control the motor; and an IOT module for establishing communication with a user by a user interface, wherein the module allows a user to control the vehicle via a mobile/web application.



No. of Pages : 19 No. of Claims : 10





क्रमांक : 011115258 SL No :



भारत सरकार GOVERNMENT OF INDIA पेटेंट कार्यालय THE PATENT OFFICE पेटेंट प्रमाणपत्र PATENT CERTIFICATE (Rule 74 Of The Patents Rules)

पेटेंट सं. / Patent No.

आवेदन सं. / Application No.

317949

415/DEL/2011

17/02/2011

फाइल करने की तारीख / Date of Filing

en e la Décisió que el recordiga

1.SHARMA Peeyush Kumar 2.BHANDARI Anil

पेटेंटी / Patentee

प्रमाणित किया जाता है कि पेटेंटी को उपरोक्त आवेदन में यथाप्रकटित DESIGNING OF AN IN SITU BIO-FILM DEVELOPMENT AND BIOREMEDIATION APPARATUS FOR THE TREATMENT OF HYDROCARBON CONTAMINATED WATERS नामक आविष्कार के लिए, पेटेंट अधिनियम, १९७० के उपबंधों के अनुसार आज तारीख 17th day of February 2011 से बीस वर्ष की अवधि के लिए पेटेंट अनुदत्त किया गया है।

It is hereby certified that a patent has been granted to the patentee for an invention entitled DESIGNING OF AN IN SITU BIO-FILM DEVELOPMENT AND BIOREMEDIATION APPARATUS FOR THE TREATMENT OF HYDROCARBON CONTAMINATED WATERS as disclosed in the above mentioned application for the term of 20 years from the 17th day of February 2011 in accordance with the provisions of the Patents Act, 1970.



अनुदान की तारीख : 08/08/2019 Date of Grant :

okryté

पेटेंट नियंत्रक Controller of Patent

टिप्पणी - इस पेटेंट के नवीकरण के लिए फीस, यवि इसे बनाए रखा जाना है, 17th day of February 2013को और उसके परवात प्रत्येक वर्ष मे उसी दिन देव होगी। Note. - The fees for renewal of this patent, if it is to be maintained will fall / has fallen due on 17th day of February 2013 and on the same day in every year thereafter.

## 1. IN202041051571 - DESIGN AND DEVELOPMENT OF SOIL INSPECTION ROBOT FOR AGRICULTURAL FIELDS

[EN] DESIGN AND DEVELOPMENT OF SOIL INSPECTION ROBOT FOR AGRICULTURAL FIELDS

National Biblio. Data Description Claims Documents

PermaLink Machine translation -

Office

India

Application Number 202041051571

Application Date

28.11.2020

Publication Number 202041051571

Publication Date 04.12.2020

Publication Kind

A IPC

C11B 9/00

Applicants

Dr. Ravikumar Gurusamy Murali Krishna Kotha Dr. Priyesh P. Gandhi Dr.R. Ablin Tarun Gehlot Mohammad Shabaz Akhil Kotha Dr. Punith Kumar M Dr. Punith Kumar S Dr.C. Samson Jerold Samuel Rohan Kotha Dr. T.S. Karthik Dr.John T Abraham Dr. Pavithra G. Dr. T.C. Manjunath

Inventors

Dr.Ravikumar Gurusamy Murali Krishna Kotha Dr. Priyesh P. Gandhi Dr.R.Ablin Tarun Gehlot Mohammad Shabaz Akhil Kotha Dr.Punith Kumar M B Dr. Senthil kumar S Dr.C. Samson Jerold Samuel

## Abstract

Title

[EN] Our India is agricultural country. In the entire resource of India agriculture plays a vital role. There are so many methods are already implemented in the agriculture domain but in modern technology every system has a problem and it needs another methodology to resolve the problem. By monitoring all the implemented technology we found one thing that the number of sensors used is large and they use the tedious way of identification of parameters. In order to reduce the number of sensors and the difficulty of the detection methods we introduce the rail dripping system along with a new method to grow the plants in a another way called "Hyperponic". The objective of the project is by using the RTOS and PIC controller we can get a new highly efficient way of automatic irrigation system and the requirement of land cultivation also reduced. In future we can easily extend the system for RED area. The water absorption rate is calculated in this method by doing the the tetterment to formers iffe and their economic condition.

|  | <br>04/12/2020   | REQUEST FOR EXAMINATION DATE<br>PUBLICATION DATE (U/S 11A)                 |
|--|--|--|
|  |  | PRIORITY DATE  |
|  |  | E-MAIL (UPDATED Online)  |
|  | vijay.21a@gmail.com  | ADDITIONAL-EMAIL (As Per Record)   |
|  | vijay.21a@gmail.com  | E-MAIL (As Per Record)   |
|  | AGRICULTURE ENGINEERING  | FIELD OF INVENTION   |
| TION ROBOT FOR AGRICULTURAL FIELDS   | DESIGN AND DEVELOPMENT OF SOIL INSPEC  | TITLE OF INVENTION   |
|  | 202041051571<br>ORDINARY APPLICATION<br>26/11/2020<br>1. Dr.Ravikumar Gurusamy<br>2. Murali Krishna Kotha<br>3. Dr. Priyesh P. Gandhi<br>4. Dr.R.Ablin<br>5. Tarun Gehlot<br>6. Mohammad Shabaz<br>7. Akhil Kotha<br>8. Dr.Punith Kumar M B<br>9. Dr. Senthil kumar S<br>10. Dr.C.Samson Jerold Samuel<br>11. Rohan Kotha<br>12. Dr. T.S. Karthik<br>13. Dr.John T Abraham<br>14. Dr. Pavithra G.<br>15. Dr. T.C.Manjunath | APPLICATION NUMBER<br>APPLICATION TYPE<br>DATE OF FILING<br>APPLICANT NAME |
|  | Application Details  | GEOGRAPHICAL INDICATIONS   |
| Controller General of Patents,Designs and<br>Department of Industrial Policy and<br>Ministry of Commerce a | GOVERNMENT OF INDIA  | PROPERTY INDIA<br>PATENTS I DESIGNS I TRADE MARKS                          |

(19) INDIA

(22) Date of filing of Application :12/05/2021

(43) Publication Date : 21/05/2021

## (54) Title of the invention : WOUND HEALING TOPICAL PHARMACEUTICAL COMPOSITION

| (51) International classification                               | :A61K0009000000,<br>A61K0047100000,<br>A61K0009060000,<br>A61K0047140000,<br>A61K0047320000 | <ul> <li>(71)Name of Applicant :</li> <li>1)Vishnu Dutt JOSHI<br/>Address of Applicant :House No. 357, Sector 9, Chopasni<br/>Housing Board, Nandanvan, Jodhpur, Rajasthan, India, 342008<br/>Rajasthan India</li> </ul> |
|---|---|--|
| (31) Priority Document No                                       | :NA   | 2)Ashish JOSHI   |
| (32) Priority Date  | :NA   | 3)Peeyush Kumar SHARMA   |
| (33) Name of priority country                                   | :NA   | 4)Anil BHANDARI  |
| (86) International Application No                               | :NA   | (72)Name of Inventor :   |
| Filing Date   | :NA   | 1)Vishnu Dutt JOSHI  |
| (87) International Publication No                               | : NA  | 2)Ashish JOSHI   |
| (61) Patent of Addition to Application<br>Number<br>Filing Date | :NA<br>:NA  | 3)Peeyush Kumar SHARMA<br>4)Anil BHANDARI  |
| (62) Divisional to Application Number                           | :NA   |  |
| Filing Date   | :NA   |  |

(57) Abstract :

The present invention relates to a topical antioxidant pharmaceutical composition and the method of preparation thereof. Topical antioxidant pharmaceutical composition is effective for treating and healing incision wound. Pharmaceutical composition is used for topical application for the treatment of ailments like: submucosal fibrosis, traumatic ulcers, stomatitis glositis and anal fissures.

No. of Pages : 39 No. of Claims : 16





भारत सरकार GOVERNMENT OF INDIA पेटेंट कार्यालय THE PATENT OFFICE पेटेंट प्रमाणपत्र PATENT CERTIFICATE (Rule 74 Of The Patents Rules)

पेटेंट सं. / Patent No.

आवेदन सं. / Application No.

21/03/2011

380374

ţ.

780/DEL/2011

फाइल करने की तारीख / Date of Filing

पेरेंटी / Patentee

1.VEERMA RAM 2.ANIL BHANDARI 3.SANJAY SHARMA 4.TARUN JAIN and 5.RATENDRA KUMAR

प्रमाणित किया जाता है कि पेटेंटी को उपरोक्त आवेदन में यथाप्रकटित ANTI DIABETIC COMPOSITION OF FRACTIONS OF THE FRESH COW URINE AND PROCESS FOR THE SAME नामक आविष्कार के लिए, पेटेंट अधिनियम, १९७० के उपबंधों के अनुसार आज तारीख 21st day of March 2011 से बीस वर्ष की अवधि के लिए पेटेंट अनुदत्त किया गया है।

It is hereby certified that a patent has been granted to the patentee for an invention entitled ANTI DIABETIC COMPOSITION OF FRACTIONS OF THE FRESH COW URINE AND PROCESS FOR THE SAME as disclosed in the above mentioned application for the term of 20 years from the 21st day of March 2011 in accordance with the provisions of the Patents Act, 1970.



अनुदान की तारीख : 27/10/2021 Date of Grant :



पेटेंट नियंत्रक Controller of Patent

टिप्पणी - इस पेटेंट के नवीकरण के लिए फीस, यदि इसे बनाए रखा जाना है, 21st day of March 2013 को और उसके पश्चात प्रत्येक वर्ष में उसी दिन देय होगी। Note. - The fees for renewal of this patent, if it is to be maintained will fall / has fallen due on 21st day of March 2013 and on the same day in every year thereafter.

क्रमांक : 011139915 SL No :





Australian Government

**IP** Australia

# CERTIFICATE OF GRANT

#### Patent number: 2021106619

The Commissioner of Patents has granted the above patent on 17 November 2021, and certifies that the below particulars have been registered in the Register of Patents.

### Name and address of patentee(s):

Gaurav Indra of Assistant Professor, Department of Information Technology, Indira Gandhi Delhi Technical University, Kashmere Gate New Delhi India

Kailash Pati Dutta of Assistant Professor, Department of Electronics and Communication Engineering, Cambridge Institute of Technology, Ranchi Jharkhand 835103 India

S N Panda of Professor, Department of Chemistry, Vikash Degree College, Bargarh Odisha 768028 India

Pankaj Mudholkar of Assistant Professor, Dept: Master of Computer Applications, Thakur Institute of Management Studies, Career Development & Research Mumbai Maharashtra 400101 India

Megha Mudholkar of Assistant Professor, Dept: Master of Computer Applications, Thakur Institute of Management Studies, Career Development & Research Mumbai Maharashtra 400101 India

Jay Prakash Srivastava of Assistant Professor, Department of Mechanical Engineering, SR University, Ananthasagar, Hasanparthy, Warangal Urban Telangana 506371 India

Rakhi Tiwari of Assistant Professor, Department of Mathematics, NITISHWAR College, A Constituent unit of Babasaheb Bhimrao Ambedkar Bihar University Muzaffarpur Bihar 842002 India

Niranjanamurthy M of Assistant Professor, Dept. of Computer Applications, M S Ramaiah Institute of Technology Bangalore 560054 India

Tarun Gehlot of Department of Structural Engineering, MBM Engineering College, University- Jai Narain Vyas University, Jodhpur Rajasthan 342001 India

G.Hudson Arul Vethamanikam of Assistant Professor, Department of Business Administration, Ayya Nadar Janaki Ammal College Sivakasi Tamilnadu 626124 India

Sachin Gupta of Assistant Professor, Department of Business Administration, Mohanlal Sukhadia University, Udaipur Rajasthan 313001 India

Mahmood Alam of Assistant Professor, Department of Mechanical Engineering, Guru Gobind Singh Educational Society's Technical Campus, Kandra, Chas, Bokaro Jharkhand 827013 India

Muthukumar Subramanian of DEAN & Professor, Department of Computer Science and Engineering, Nagarjuna College of Engineering & Technology, Bangalore Karnataka 562110 India

### Title of invention:

INTELLIGENT SYSTEM FOR POLLUTION MANAGEMENT TO MONITOR POLLUTANTS RELEASED INTO THE AIR FROM INDUSTRIES USING IOT

#### Name of inventor(s):



Dated this 17th day of November 2021

**Commissioner of Patents** 

PATENTS ACT 1990

this IP Right