ABSTRACTS

1) A process for the manufacture of nanocomposites for removal of dyes from waste-waters of industrial effluents comprising the steps of preparing nano-composites from at least one substance of the following group of nanometals and nanometal oxides, namely, An, Ag, Fe, Co, Ni, Pt, Pd, Ru, ZnO, TiO2, Fc2O3, Fe2O4, dispersed on a support material consisting of two or three difference dimensional nanomaterials selected from at least one of the following, namely, carbon nanotubes, thermally exfoliated graphite, chemicaklly exfoliated graphite oxide, graphene sheets, graphene, functionalized carbon nanotubes / graphene; passing the waste waters through at least one nanofiltration memberane which is made of a biocompatible material such as chitosan, polypyrole; passing the waste water thereafter over the nanocomposites to enable the nanocomposites to adsorb the dyes in the said waste waters, the nanocomposites being then treated with an inorganic acid for re-using the said nanocomposites.

We Claim:

- A method of manufacture of *the Carbon Nanostructures including* 1-7 at % *nitrogen modified* carbon nanostructures or 1-7 at % *Boron modified* carbon nanostructures including Mg alloy *nanoparticles catalysts which is about 5-30 wt% on the Carbon Nanostructures support materials. The Carbon Nanostructures include carbon nanotubes and Graphene.*

- *The carbon nanotubes either single walled nanotubes or multiwalled nanotubes and a few layered Graphene sheets are prepared by in situ reduction with B/N modification materials and appropriate salt precursors* of Mg alloy *nanoparticles catalysts by hydrogen exfoliation method or solar exfoliation method or thermal exfoliation method.*

- *A method for preparing modified Carbon Nanostructures with Nitrogen and or Boron by using melamine or boric acid respectively by hydrogen exfoliation method or thermal exfoliation method.*

- *A method for the dispersion of the* alloy *nanoparticles catalysts by using the appropriate salt precursors and reduction by the chemical method or hydrogen exfoliation method. The dispersion of the MG* alloy *nanoparticles*

catalysts is about 5-30 wt% on the nitrogen or boron modified Carbon Nanostructures.

- *Invention of materials having hydrogen storage capacity of around 6.7 wt % at 25°C and at a pressure of 35 bar in Zeolites added with Boron or nitrogen modified Carbon Nanostructures and with Mg and transition metal (Ni, Co, Fe) alloy Nanoparticles as catalysts.*

- *A method for* manufacturing of *Carbon Nanostructures which are modified with above mentioned B/N elements and dispersed with above mentioned Mg and transition metal (Ni, Co, Fe) alloy nanoparticles catalysts* therefrom for hydrogen storage applications when manufactured by a method as claimed in any one of the preceding Claims.

Dated this the day of 2013

M.K.Rao
KAMATH & KAMATH
APPLICANTS' ATTORNEY

E.9 JAN 2014

Figure 1:Raman Spectra of Zeolite-G-Mg_2Ni

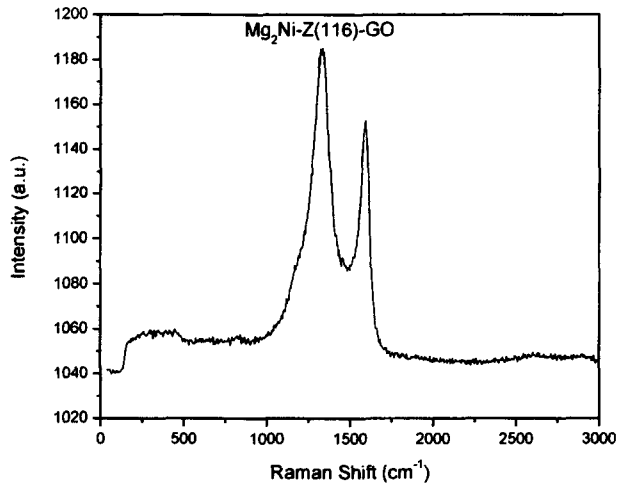
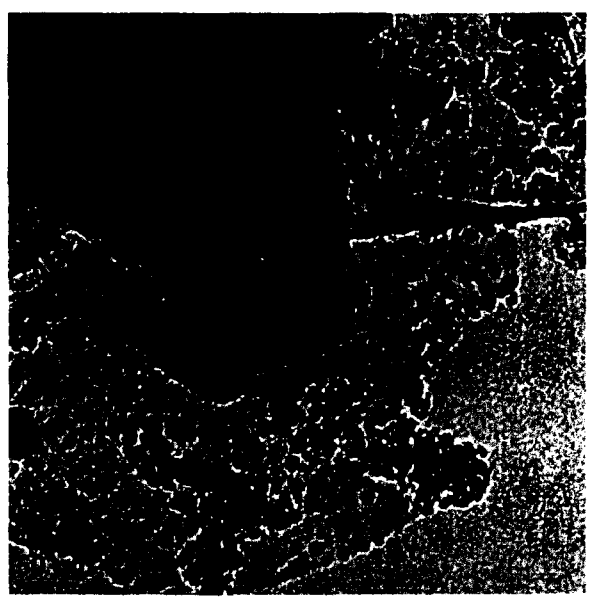


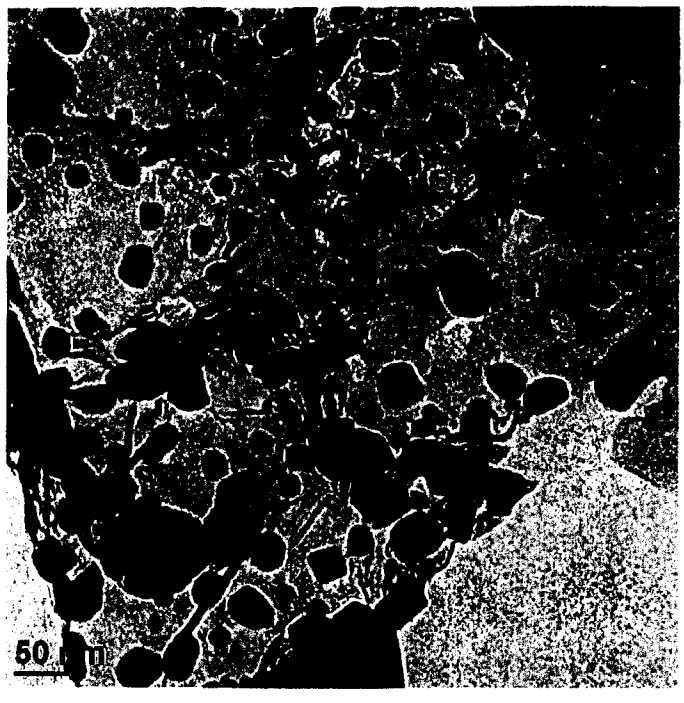
Figure 2 shows the TEM image Zeolite-G-Mg_2Ni



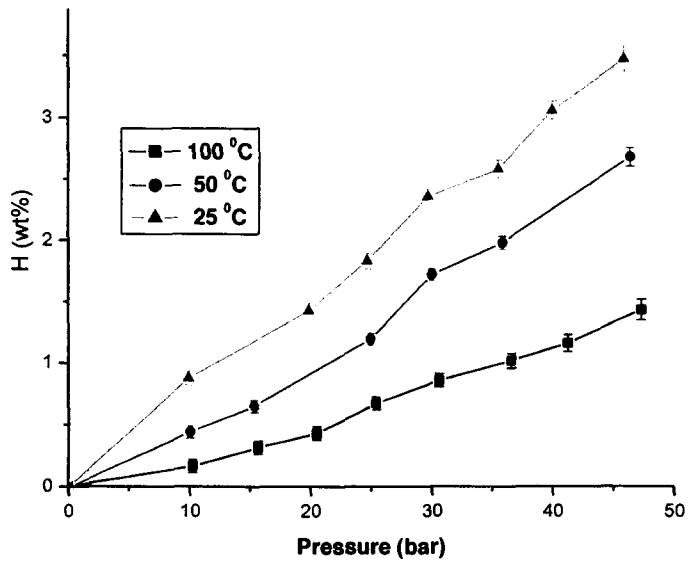
ORIGINAL

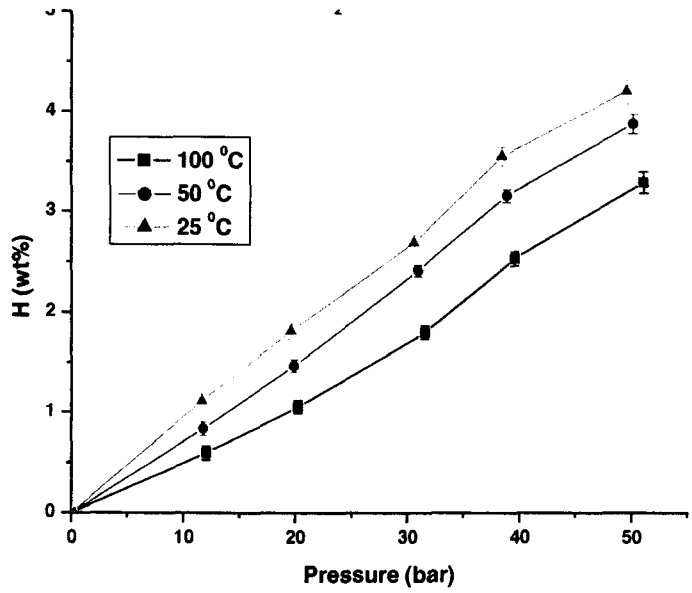
No 103 ICHE/ 2014

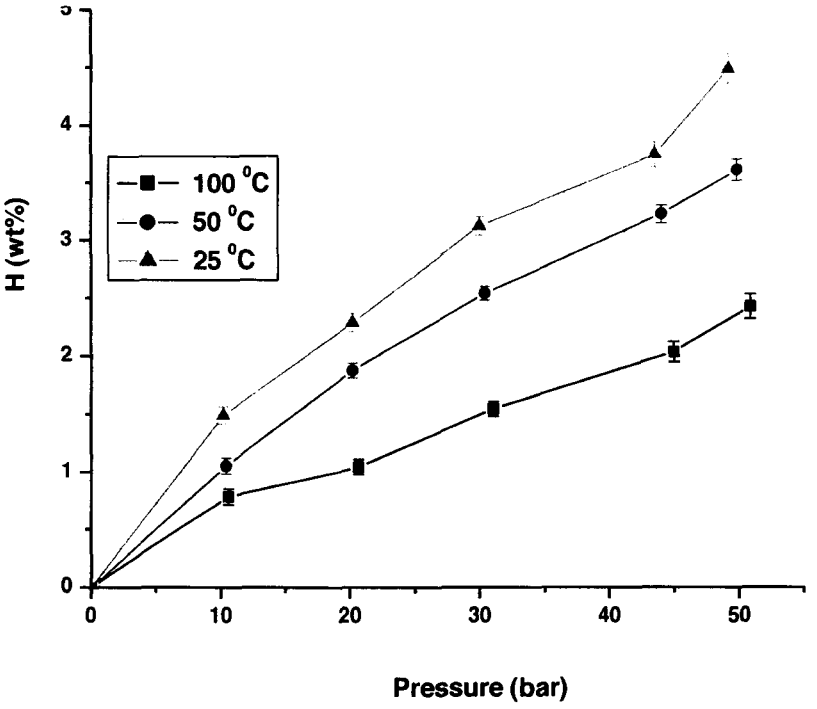
Figure 3 shows the TEM image N-G-Mg_2Ni

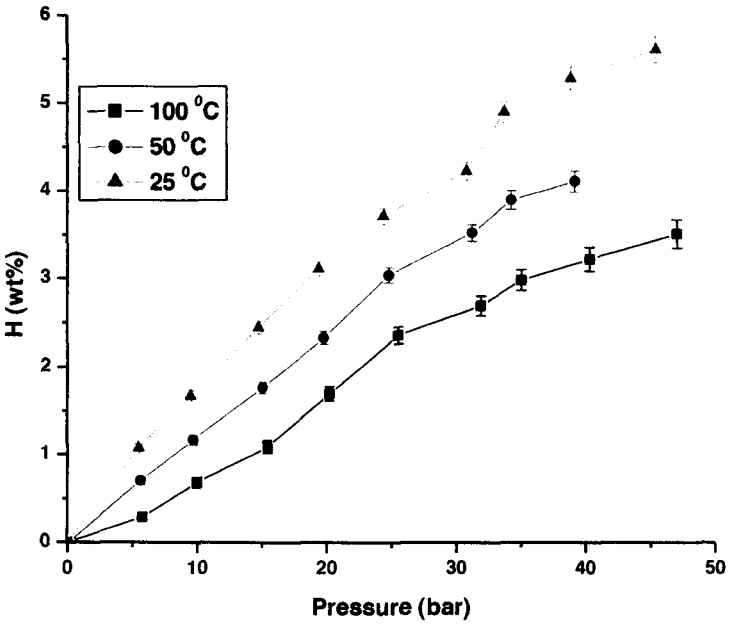


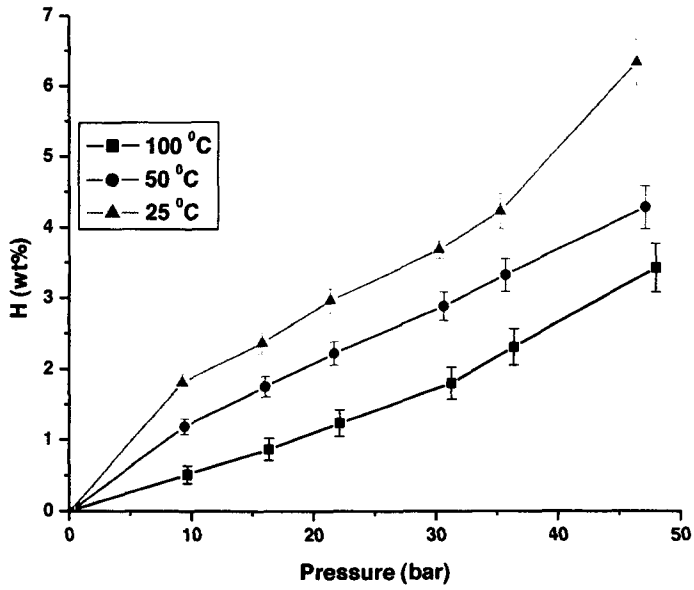
Figures 4-11 give the hydrogen storage capacity in Mg_2Ni-G-MWNT; Mg_2Ni-G; Mg_2Ni-Zeolite-BG; Mg_2Ni-Zeolite-2G; Mg_2Ni-Zeolite-G; Mg_2Ni-zeolite-G-MWNT; Mg_2Ni-BG; Mg_2Ni-NG

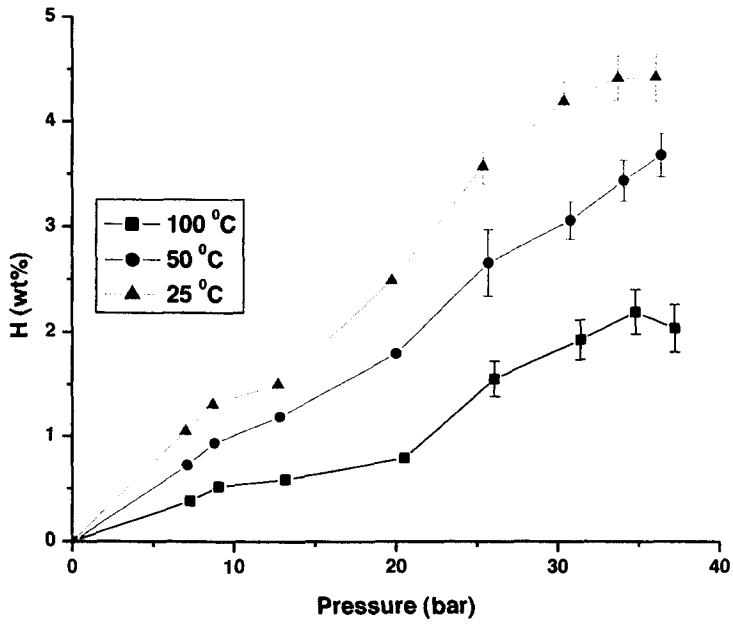


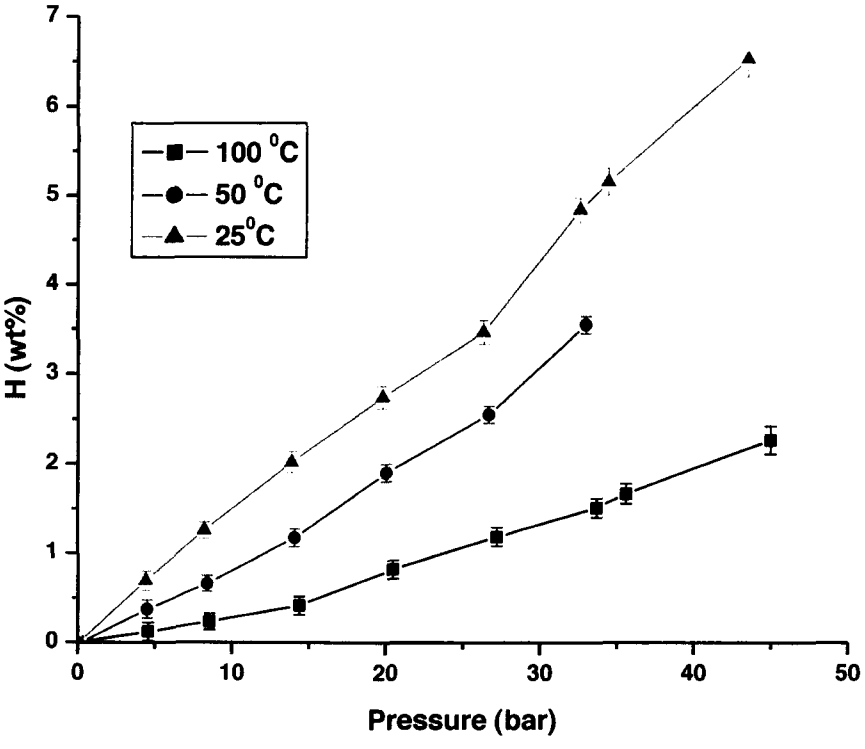


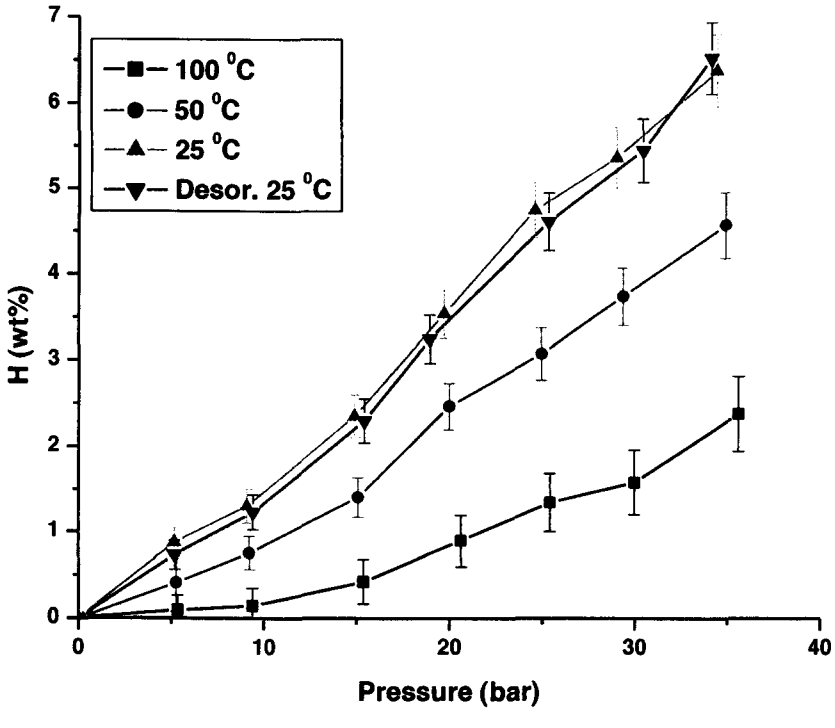

100 °C
50 °C
25 °C
H (wt%)
Pressure (bar)
0
1
2
3
4
0
10
20
30
40
50


100 °C
50 °C
25 °C
H (wt%)
Pressure (bar)
0
1
2
3
4
5
6
0
10
20
30
40
50


100 °C
50 °C
25 °C
H (wt%)
Pressure (bar)
0
1
2
3
4
5
6
7
0
10
20
30
40
50


100 °C
50 °C
25 °C
H (wt%)
Pressure (bar)
0
1
2
3
4
5
0
10
20
30
40


100 °C
50 °C
25°C
H (wt%)
Pressure (bar)
0
1
2
3
4
5
6
7
0
10
20
30
40
50


100 °C
50 °C
25 °C
Desor. 25 °C
H (wt%)
Pressure (bar)
0
1
2
3
4
5
6
7
0
10
20
30
40

THE FOLLOWING SPECIFICATION PARTICULARLY DESCRIBES THIS INVENTION AND THE MANNER IN WHICH IT IS TO BE PERFORMED

This invention relates to '*Synthesis of Zeolites modified Carbon Nanostructures with Mg alloy Nanoparticles as catalysts for the dissociation of hydrogen molecules*' materials therefrom for hydrogen storage.

Hydrogen is the ideal fuel for new types of fuel cell vehicles, but the main issue is how to store hydrogen in the fuel cell vehicles. Hence, the *development of hydrogen storage materials in an important aspect for possible application in Fuel cell runs automobiles. DOE (department of energy, USA) has set the target of developing materias with 5.5 hydrogen wt% for 2015. This invention will definitely draw attention from Companies/industries/R&D sections in India and abroad due to the fact that the development of hydrogen storage material with a minimum of 5.5 wt % is the one of the 'bottleneck' in the use of Fuel cell in automobiles instead of fossil fuels. This present invention removes the 'bottleneck' in the storage of hydrogen in automobiles.*

Storing hydrogen as a compressed gas or liquid has risks of leaks or explosions. Therefore storing hydrogen into solid materials has long been seen as the way forward. The conventional materials cannot store very large amounts of hydrogen, and nanotechnology seems to hold the key to viable solutions to the safe storage of hydrogen

because nanostructured materials have unique and tunable properties, which are well suited to hydrogen storage. Scientists around the world are working on the hydrogen storage properties of different nanostructured materials. The key is to find nanostructured materials, which can store, release and reabsorb hydrogen at ambient temperatures and practical pressures. Therefore, the main challenges to improve the hydrogen storage technologies are to increase the storage weight % using nanotechnology, and reduce the cost of the material.

In this invention, we have demonstrated a hydrogen storage capacity of ~6.7 wt % in Zeolites modified Carbon Nanostructures with Mg alloy Nanoparticles. The Carbon Nanostructures – zeolite composites are zeolite modified exfoliated Nitroge/Boron doped graphitic oxide dispersed with Mg alloy Nanoparticles as catalysts for the dissociation of hydrogen molecules.

A hydrogen storage capacity of ~6.7 wt % is achieved in Carbon Nanostructures – zeolite composites. The Carbon Nanostructures – zeolite composites are zeolite modified exfoliated Nitroge/Boron doped graphitic oxide dispersed with Mg alloy Nanoparticles.

I. The Carbon Nanostructures are hybrid type with carbon nanotubes (CNT =SWNT, MWNT), Graphene and zeolites included with Nitroge/Boron.

II. The catalysts are nanoparticles of Mg-TM alloy Nanoparticles with TM= Fe, Co, Ni.

The process of synthesis of zeolite modified exfoliated Nitroge/Boron doped graphitic oxide dispersed with Mg alloy Nanoparticles as catalysts was performed by a single step process of hydrogen exfoliation cum reduction. The maximum hydrogen storage capacity of ~6.7 wt % is achieved n the hydrogen equilibrium pressures of 35 bar & 25°C which is more than the target set for 2015 by DOE, USA.

The materials are Zeolites modified Carbon Nanostructures with Mg alloy Nanoparticles as catalysts. The following is the description of the synthesis of these materials.

- *The Zeolites are first activated by KOH treatment. This is done by usually grinding KOH and zeolite together for 1 hour. For 500 mg zeolite we usually take 50 mg of KOH. Then spread the ground mixture in a quartz boat and heat it in the furnace at 500 C for 2 h in the presence of Argon.*
- *The activated zeolites are then mixed with appropriate salt precursors of the alloys. In addition, the required amount of Carbon Nanostructures along with the source of Nitrogen or Boron is added. The carbon Nanostructures is Carbon nanotubes and or Graphene. Melamine or boric acid is used as source of nitrogen or boron. The nitrogen or boron in the hexagons of carbon will modify the electronic structure thereby favors the bonding of hydrogen atoms.*
- *Nitrogen or Boron doped hybrid Carbon Nanostructures is used as support materials for the dispersion of catalysts.*

- *The catalysts are one of the alloys Nanoparticles which act as catalysts for the dissociation of hydrogen molecules into hydrogen atoms. The alloys are the combination of Mg and Transition metal (Fe, Co, Ni).*
- *The Mg alloy NPs catalysts dispersion is performed by using the appropriate salt precursors and by the chemical method, hydrogen exfoliation method. The dispersion of the alloy NPs catalysts is about 5-30 wt% on the in Zeolites modified with Boron or nitrogen doped Carbon Nanostructures.*
- *The entire mixture is reduced to the hydrogen storage material by the hydrogen exfoliation method or thermal exfoliation method.*
- *These Boron or nitrogen doped Carbon Nanostructures are with 1-7 at % of N and Boron. Alloy nanoparticles are the combination of Mg and transition metal (Ni, Co, Fe) with 10-30 wt%.*
- *The hydrogen storage capacity and the desorption capacity have been investigated in the temperature range 25C –100°C by pressure reduction technique using Sievert's facility in the hydrogen equilibrium pressure 1-50 bar.*
- *A maximum hydrogen storage capacity of 6.7 wt % is achieved at room temperature and at a moderate pressure of 35 bar& 25°C in Zeolites added with Boron or nitrogen modified Carbon Nanostructures and with Mg alloy Nanoparticles as catalysts.*

The morphology of the prepared Carbon Nanostructures was examined by using Scanning electron microscope (SEM) and transmission electron microscope (TEM). The amount of nitrogen/boron doped was studied by XPS. The amount and the average particle size of the dispersed nanoparticles of alloy/element was investigated by TGA and EDAX. The doping of nitrogen and boron was found to be 3-10 % and the particle size of the Mg alloy nanoparticles was found in the range of 10-15 nm.

Hydrogen adsorption studies were carried out in the pressure range 1-50 bar and in the temperature range 25–100°C using a high-pressure Sieverts' apparatus. The unit has been calibrated with high-purity hydrogen (99.99%) at various initial pressures. At various pressures and temperatures, the volume of gas contained in the empty sample cell was precisely measured after confirming leak-free system. At high pressures, the ideal gas law was corrected using van der Waals equation for the volume of gas molecules and molecular interactions. Each time, over 250 mg of sample was loaded in the sample cell and activated by vacuum heat treatment before exposing to hydrogen to remove the oxide layers. In each activation process, the sample was evacuated to a vacuum of 10^{-6} Torr and heated at 250 °C for 2 h. It was then cooled to 100 °C, and hydrogen was allowed to interact with the sample. The sample was subsequently cooled to 75°C, 50°C, 25°C. The pressure–composition relationships were obtained by calculating the hydrogen storage capacity in wt % from the pressure drop during the hydrogen adsorption at constant temperature. After each cycle, the sample was degassed for 3 h at

250 °C under a vacuum of 10^{-6} Torr. During the experiment the room temperature was maintained at 24 ± 1 °C.

A maximum hydrogen storage capacity of 6.7 wt % is achieved at room temperature (25°C) & at a moderate pressure of 35 bar in Zeolites added with Boron or nitrogen modified Carbon Nanostructures and with Mg alloy Nanoparticles as catalysts.

It is noteworthy:

- *Hydrogen storage is the major application areas of this invention.*
- *This invention involves a significant reduction in the cost of raw materials involved in its preparation because of use of zeolite and graphite.*
- *This invention is a simple and scalable process adaptable to mass manufacturing and immediate adoption as retrofit to existing systems.*
- *A maximum hydrogen storage capacity of 6.7 wt % is achieved at (25°C) and at a moderate pressure of 35 bar which is the same as the target of US Department of Energy.*