2013

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THE JIGGLED BED REACTOR, A NEW FLUIDIZED BED REACTOR FOR CATALYST TESTING

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ABSTRACT

The jiggle bed reactor (JBR) is a state-of-the-art batch fluidized micro reactor designed and developed to test catalysts for endothermic reactions. This paper illustrates the function and operation of the JBR, including its fluidization dynamics and heat transfer parameters. This paper then reviews the specific performance of the JBR reactor for catalytic gasification of a model compound and raw pyrolysis bio-oils.

INTRODUCTION

Many of the commercial reactors that perform reactions with a solid catalyst use fluidized beds. The laboratory reactors that are currently used for catalyst testing (5,6) suffer from two major limitations:

- To fluidize the particles in a small reactor and still achieve the same gas residence time as with commercial reactors, the gas is recirculated with an impeller. This requires a mechanical seal that cannot perform well at high temperatures and pressures.
- Many industrially important catalytic reactions are endothermic. In current testing reactors, heat is provided by heat transfer from the wall of the gas recirculation loop. Because of the low wall to gas heat transfer coefficient, the wall temperature is much higher than the temperature of the catalyst bed, resulting in parasitic thermal cracking reactions near the wall region.

The objective of development of the jiggle bed reactor was to allow for catalyst testing in a reactor with no mechanical seal and with a wall temperature that would be within a few degrees of the catalyst temperature.

THE JIGGLE BED REACTOR SETUP

The main structure of the jiggle bed reactor is shown in Figure 1. The JBR is a micro reactor to test catalysts and investigate the effect of operating conditions, for gas-solid, endothermic, catalytic reactions. This reactor is designed to overcome the limitations of the conventional test reactors, which were reviewed in the previous section. The first innovation is that, in the JBR, fluidization is achieved by moving the reactor up and down with the appropriate amplitude and frequency, instead of forcing a gas through the bed. The second innovation is that heating is provided by induction, using small metal pins within the catalyst bed.

Induction heating allows for excellent temperature, with a fast response to changes in temperature at endothermic reactions.

Since induction heating is used, the reaction chamber is made of a non-conductive material to prevent shielding of the magnetic field. In addition, it is necessary to ensure that it can withstand temperatures as high as 900 °C. Moreover, the material must be non-porous and strong enough to sustain the vibrating conditions. Therefore, a non-porous ceramic crucible made of 99.8% alumina is utilized. The crucible has an I.D. of 2.54 cm and a height of 7.3 cm. The heating element of the JBR is an assembly of eight Inconel wires with a 0.32 cm diameter and a 7.0 cm length, which are circularly placed inside the ceramic crucible with equal distance from wall and center of the crucible. When a high frequency current is applied through the copper coil (item 11 in Figure 1), the associated magnetic field induces a current through the Inconel wires. Due to the high frequency of the magnetic field, the energy of the induced currents is lost rapidly in the form of heat which is uniformly transferred from the surface of the wires into the bed of catalyst particles.

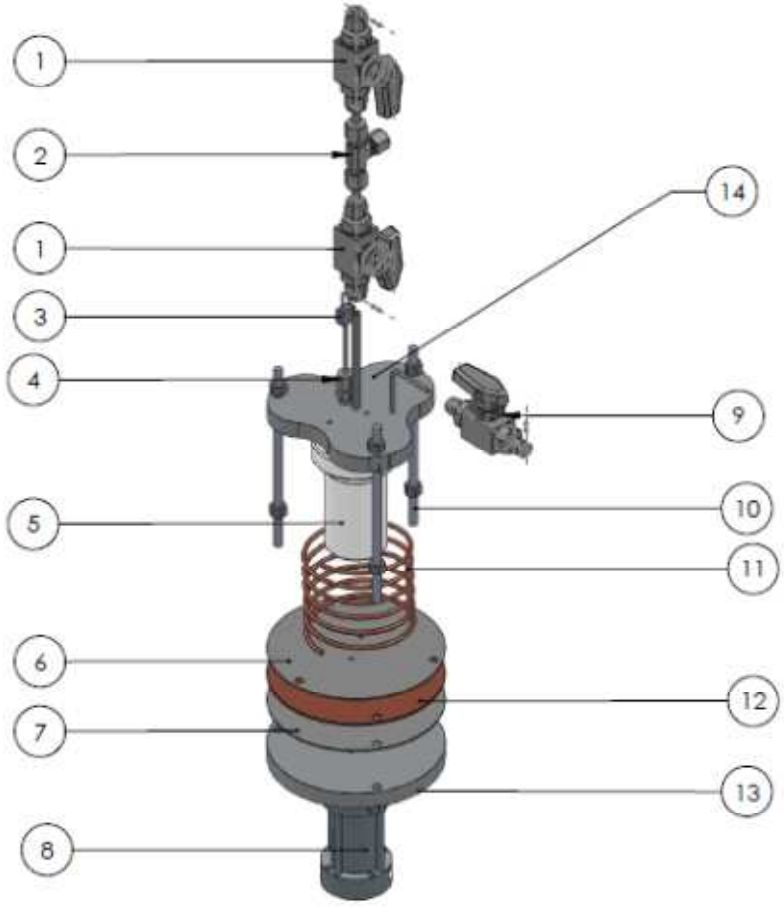

Figure 1. Diagram of the jiggle reactor: 1. on/off feed valves 2. Inlet of carrier gas 3. Thermocouple 4. Inlet of feed and carrier gas 5. Ceramic crucible with insulation 6. Insulation disk 7. Insulation disk 8. Linear pneumatic actuator 9. Outlet gas valve 10. Stainless steel support rods 11. Copper coil 12. Copper disk 13. Aluminum disk mounted on the actuator 14. Stainless steel scalloped disk.

The endothermic reactions take place batch-wise in the JBR. Before injecting the feed, inert gas flushes any air from the reactor. Reactor agitation is initiated at the frequency and amplitude that guarantee effective mixing of the solid particles throughout the entire volume, and heating is applied to achieve the desired reaction temperature. The feed is then introduced in the reactor through an air lock. During the reaction, induction heating maintains a constant reaction temperature throughout the catalyst bed. At the end of each run, the lower inlet valve and the outlet valve are opened (valves 1 and 9 in Figure 1), and inert gas flows through valve 1 to flush the reaction products into a gas sampling bag.
In order to generate an effective gas solid contact within the reactor, it is essential to carefully select the amplitude and frequency of the vertical movement of the pneumatic actuator that moves the reactor up and down. When the catalyst bed expands over the entire length of the crucible, the majority of the gas in the upper part of the crucible is displaced downward. When the bed contracts downward, the gas in the lower regions of the crucible is displaced upward. Thus, the alternating expansion/contraction of the catalyst bed induces intense axial and radial mixing of the gas and solid phases. With the right amplitude and frequency, the catalyst particles are fluidized and well mixed by this motion, with the catalyst bed expanding and contracting in *rapid succession*, as shown by Figure 2. Frequency of the vertical movement was varied between 3 and 6 Hz and its amplitude was varied between 6 and 9 cm. This alternating movement of the particles through the reactor ensures effective contact between the gas and the solid catalyst.

The reactor was operated in batch mode and the reported gas residence time was the time interval between the feedstock injection and the flushing out of the product gases. In a separate study (1), the reactor was tested for the reforming of acetic acid and provided results similar to the results obtained in a continuous, pilot plant fluidized bed.

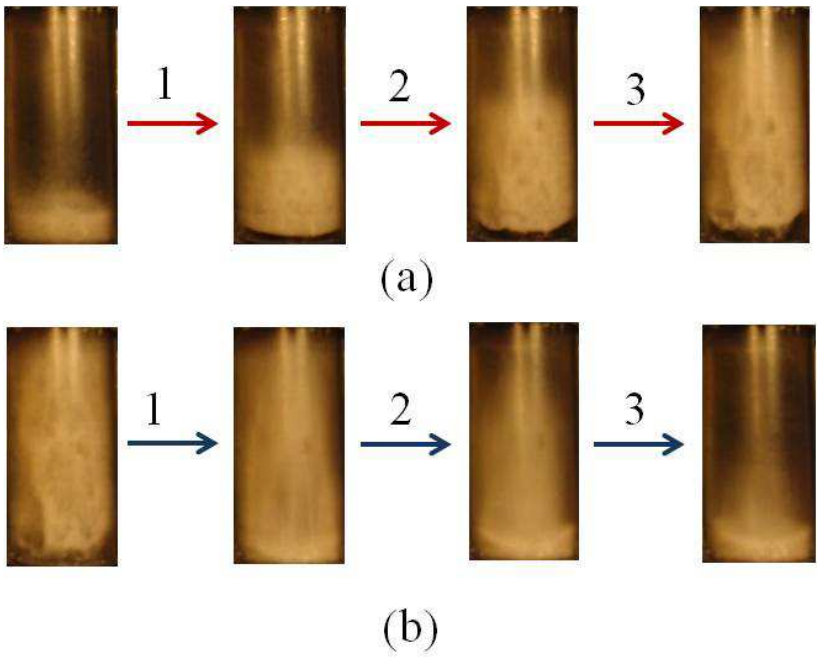

Figure 2. Sequences of mixing of catalyst particles in the jiggle bed reactor: (a) bed expansion during downward actuator retraction (b) bed contraction during upward actuator extension.

STUDY OF FLUIDIZATION DYNAMICS IN JBR

A visual set-up with a transparent crucible, shown in Figure 3, was originally developed to investigate the fluidization of the catalyst particles. The quality of the distribution of catalyst particles over the entire length of the crucible was studied as a function of the amplitude and frequency of the pneumatic actuator, for various amounts of bed particles of various size distributions. Sand particles with size distributions of 75-149 µm, 149-212 µm, and 212-355 µm were loaded in a transparent crucible with dimensions similar to those of the ceramic crucible, to form a bed of particles. The bed mass ranged from 5 to 20 g. The pressure of the air driving the actuator was varied from 138 to 690 kPa to achieve an actuator frequency ranging from 3 to 6Hz. Level switches were used to vary the amplitude of the crucible motion from 64 to 89 mm.

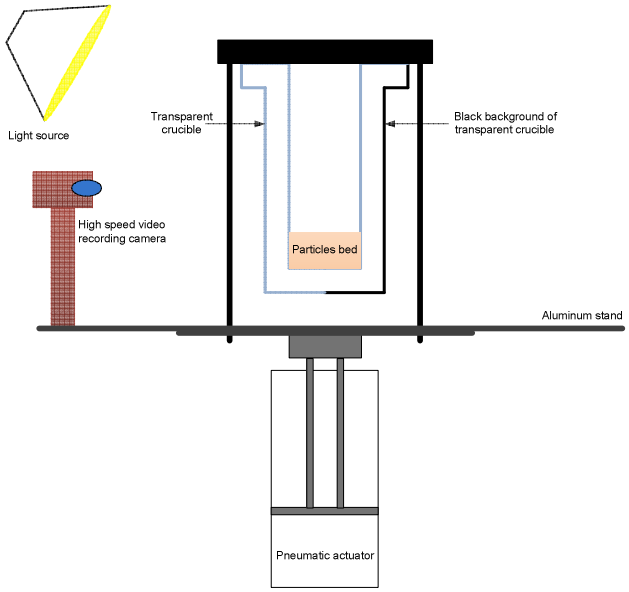

Figure 3. Visual set-up to investigate mixing of sand particles in a transparent crucible

In order to develop to determine when a uniform distribution and effective fluidization of the sand particles is achieved, a high speed video camera (210 frames per second) was used to capture the movement of the solids within the transparent crucible, and computerized image processing helped analyze the recorded videos. For each operating condition, about 14 seconds of the related video was converted to 3000 images in JPEG format. The images were converted to gray images that were digitized so that each pixel had a color value between 1 (for black pixels) and 256 (for white pixels). Then, the coefficient of variation of the pixel gray values was obtained in the space coordinates within the crucible volume:

$$CV_{space} = \left(\frac{\sigma}{\mu}\right)_{space} = \frac{\sqrt{\frac{1}{N_i}\sum_i\left(y_{i,j} - \frac{\sum_i y_{i,j}}{N_i}\right)^2}}{\frac{\sum_i y_{i,j}}{N_i}} \quad (1)$$

where $y_{i,j}$ is the color value of a pixel in picture i at time j, N_i is the number of pixels in picture i, σ is the standard deviation of color values, and μ is the average of color values.

A signal processing program, using the Fourier transform, provided the power spectrum that illustrated the variation with time of CV_{space}, for different amplitudes and frequencies. The power for the dominant frequency characterizes the fluidization intensity: a higher power indicates a more regular fluidization.

Investigating the effect of the size distribution and mass of the catalyst particles showed that these parameters can be adjusted to optimize the fluidization quality. For example, Figure 4 shows that the bed mass had a significant impact on the fluidization dynamics. The power of the CV_{space} signal at its dominant frequency changed dramatically with the bed mass. This variation was non monotonic, with the highest power being achieved for the intermediate bed mass of 10 g, for all particle size distributions. The size distribution of the bed particles also had a significant impact on the fluidization dynamics, as shown in Figure 5. The power of the CV_{space} signal at its dominant frequency varied with the size distribution of the bed particles. The highest power was obtained for the intermediate particle size distributions of 149 – 212 µm. Therefore, it was concluded that it was best to use a bed of 10 g with a 149 – 212 µm particle size distribution.

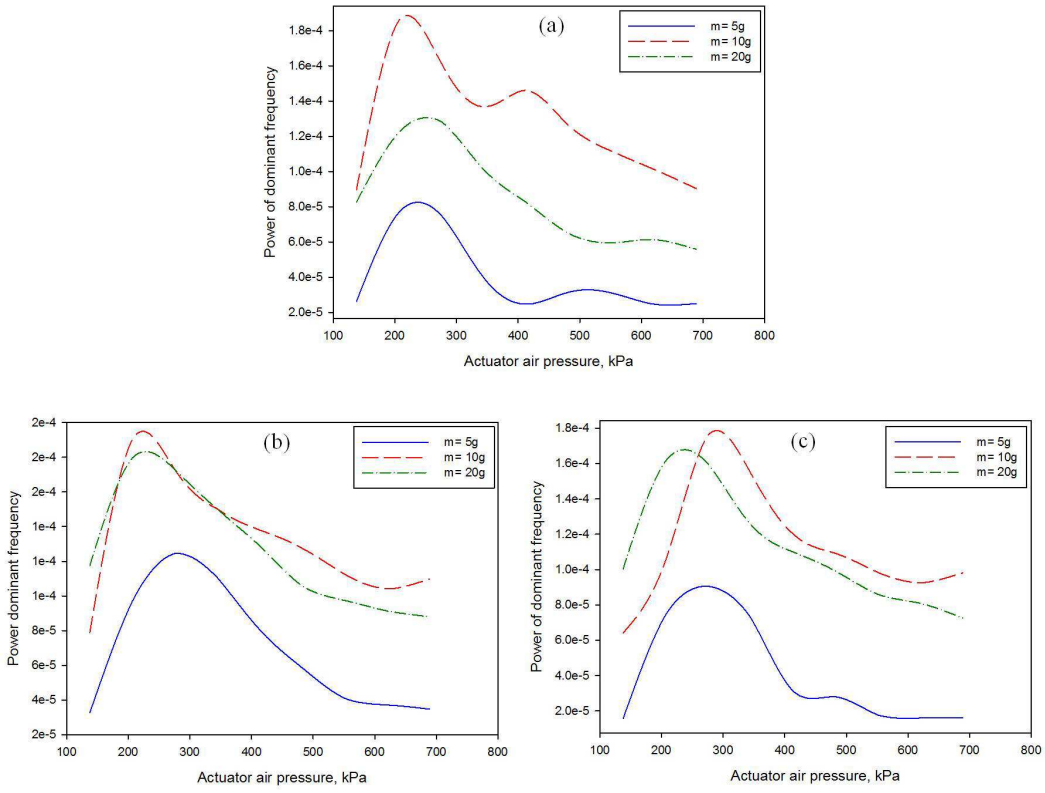

Figure 4. Effect of mass of the particles on dominant frequency power of the particle mixing; sand particles with size of (a) 212-355 µm, (b) 149-212 µm and (c) 75-149 µm

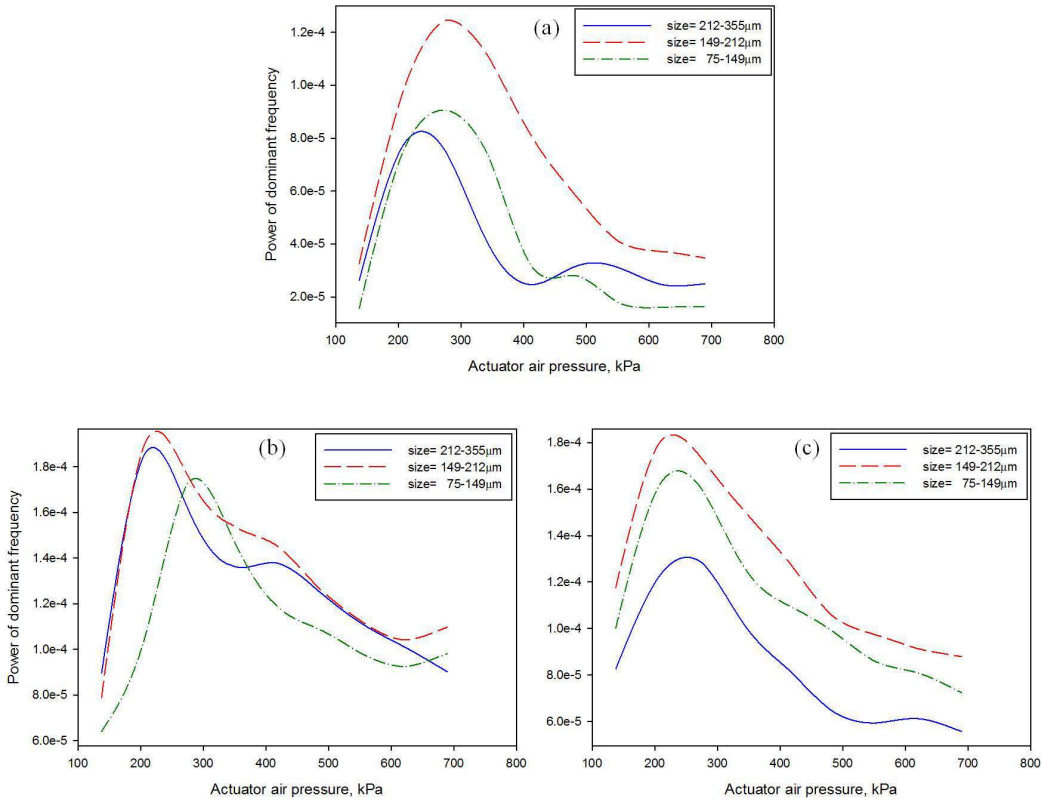

Figure 5. Effect of size distribution of the sand particles on dominant frequency power of the particle mixing; mass of sand particles: (a) 5 g, (b) 10 g and (c) 20 g

STUDY OF HEAT TRANSFER IN THE JBR

Heat transfer parameters in the jiggle bed reactor (JBR) were estimated through heat balance equations for cooling and heating the reactor. When the power supply is switched off, the temperature of the catalyst bed drops quickly due to heat losses from the reactor:

$$m_s C_{P,s} \frac{dT_s}{dt} = -h_o A_o (T_s - T_\infty) \quad (2)$$

where m_s is the mass of bed solids, $C_{P,S}$ is the specific heat capacity of the solids, T_s is the bed temperature, h_0 is the heat loss coefficient, A_0 is the heat loss surface area and T_∞ is the outside temperature.

During heating up of the catalyst bed within the JBR, the metal wires are heated by induction and heat is transferred from the wires to the bed, with a power P. Heat is lost through the reactor wall, as during cooling:

$$m_s C_{P,s} \frac{dT_s}{dt} = P - h_o A_o (T_s - T_\infty) \quad (3)$$

The temperature difference between the metal wires and the bed was determined by means of label temperature indicators (from Omega) applied to the metal wires. The temperature difference between wire and bed ranged from 2 to 6 °C when the induction heating power outlet varied between 10 and 20 %. The heat transfer coefficients and power consumption are presented in Table 1 for various

power levels. The heat transfer coefficient obtained at the higher power level of 20 % was more realistic and was more relevant to this study, since it provided a steady state temperature of around 800 °C, a typical temperature desirable for gasification reactions. According to the correlation from Molerus et al. (4) for heat transfer between a tube and a regular, gas-fluidized bed, the heat transfer coefficient obtained at a power level of 20% would correspond to a ratio of fluidization velocity to minimum fluidization velocity of about 30, which is typical of a well-bubbling fluidized bed. This confirms the excellent quality of the fluidization achieved within the JBR.

Table 1. Consumed power, heat loss heat transfer coefficient and heat transfer coefficient between wires and bed; air pressure 207 kPa, sand particles 10 g

Power level (%)	$h_0\left(\frac{w}{m^2.°C}\right)$	$P\ (w)$	$h_w\left(\frac{w}{m^2.°C}\right)$
10	0.265	1.671	45
12	0.257	2.462	80
15	0.270	4.063	220
20	0.269	6.058	493

VALIDATION OF THE JBR FOR CATALYTIC GASIFICATION

In order to obtain reliable and reproducible data, the functionality of the jiggle bed reactor was first investigated by carrying out catalytic steam reforming of acetic acid, which is a bio-oil model compound. Tests were conducted at different operating conditions and the results were also compared with literature data obtained from conventional test reactors.

In order to conduct a test with the desired molar steam to carbon ratio, a solution of acetic acid in de-ionized water was prepared. Then, precise samples of 4µl were injected into the reactor by means of capillary tubes. Catalytic gasification was conducted at 700 °C, using a commercial, nickel based, steam reforming catalyst (X), with a particle size ranging from 220 to 350 µm. After collecting produced gases in sampling bags, each sample was analyzed with a Varian CP4900 micro GC.

As presented in Table 2, the results of the acetic acid steam reforming experiments conducted in the jiggle bed reactor were comparable to literature data obtained under similar operating conditions, with a much larger, conventional gas fluidized bed.

Table 2. Comparison between data from catalytic steam reforming of acetic acid in JBR with data in literature. Temperature = 700 °C.

Molar steam to carbon ratio = 6				Molar steam to carbon ratio = 3			
	Catalysts tested by Medrano *et al.* (3)		This study	Catalysts tested by Vagia and Lemonidou (2)			This study
Catalyst	*Ni/Al,Ca0.5*	*Ni/Al,Mg0.2*	*X*	*5%Ni*	*10%Ni-1*	*10%Ni-2*	*X*
H_2	0.84	0.87	0.84	0.88	0.83	0.87	0.78
CO	0.18	0.14	0.17	0.27	0.30	0.31	0.27
CO_2	0.71	0.85	0.79	0.73	0.67	0.69	0.65
CH_4	0.00	0.00	0.04	0.00	0.03	0.00	0.06
C_2H_4+C_2H_6	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Conversion	0.90	0.99	1.00	1.00	1.00	1.00	1.00

CATALYTIC GASIFICATION OF BIO-OILS IN THE JIGGLE BED REACTOR

Catalytic gasification was then performed with the same catalyst (X) using two bio-oils: a hardwood bio-oil (DMB) with a general formula of $CH_{2.624}O_{1.121}N_{0.007}$ and a 28 wt% water content, from Dynamotive, and a birchwood bio-oil produced at ICFAR (BWB) with a general formula of $CH_{2.071}O_{0.920}N_{0.005}$ and 50 wt% water.

Figure 6 shows that the yields of hydrogen and CO increased with increasing temperature and catalyst mass, while the production of hydrocarbons declined drastically. Figure 7, which compares the molar H_2/CO ratios obtained from catalytic gasification of the DMB and the BWB bio-oils, shows that larger values of the molar H_2/CO ratio were achieved with the BWB. Since the BWB bio-oil had a larger water content, the presence of additional steam promotes the water-gas shift reaction with the gasification reactor, and may eliminate the need for a separate a water gas shift reactor downstream.

CONCLUSIONS

The jiggle bed reactor is a novel and effective batch micro reactor for the effective testing of different catalysts for endothermic reactions at low cost and minimum time, while precisely simulating the fluidization dynamics and heat transfer conditions typical of industrial fluidized bed reactors. This reactor can be used also to investigate a variety of endothermic reactions with diverse feedstocks.

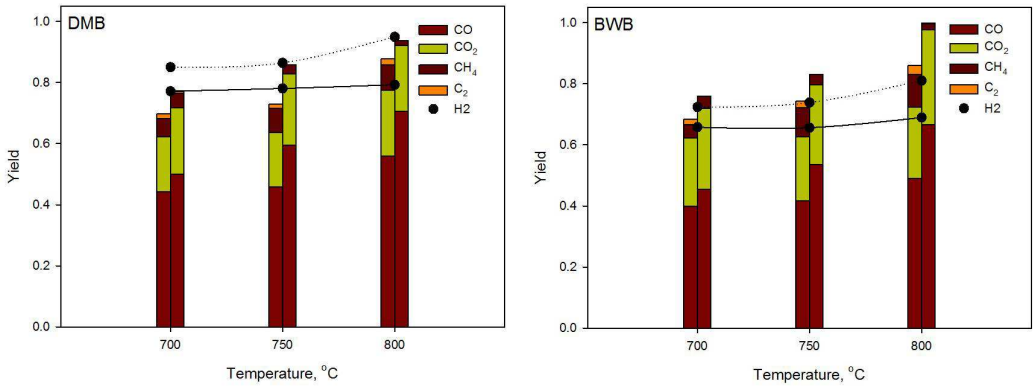

Figure 6. *Yield of product gases from catalytic cracking of the DMB and the BWB versus temperature. Residence time= 30s; left hand side bars: mass of the catalyst X= 0.5 g; right hand side bars: mass of the catalyst X= 1 g.*

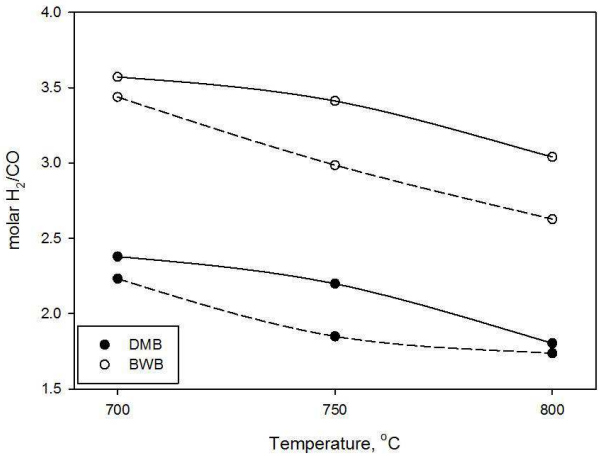

Figure 7. *Yield of molar H_2/CO ratio from catalytic cracking of the DMB and the BWB; residence time= 30 s. Mass of the catalyst X= 0.5g (continuous lines) and 1g (dashed lines)*

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