

## Chemical Looping Combustion of Rice Husk

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### ABSTRACT

A thermodynamic investigation of direct chemical looping combustion (CLC) of rice husk is presented in this paper. Both steam and CO<sub>2</sub> are used for gasification within the temperature range of 500–1200°C and different amounts of oxygen carriers. Chemical equilibrium model was considered for the CLC fuel reactor. The trends in product compositions of the fuel reactor, were determined. Rice husk gasification using 3 moles H<sub>2</sub>O and 0 moles CO<sub>2</sub> per mole carbon (in rice husk) at 1 bar pressure and 900°C was found to be the best operating point for hundred percent carbon conversion in the fuel reactor. Such detailed thermodynamic studies can be useful to design chemical looping combustion processes using different fuels.

**Key Words-** Chemical looping combustion, NiO, Rice husk, Thermodynamic study.

### I. Introduction

CO<sub>2</sub> emission to the environment is mainly contributed by industrial power generation. Concerns for climate change phenomenon have started special procedures to reduce CO<sub>2</sub> emissions using CO<sub>2</sub> capture and storage techniques that capture CO<sub>2</sub> from energy-intensive processes and then store it in suitable geologic locations [1]. The concerns of increase in greenhouse gas emissions and an inevitable global warming crisis is reported by researchers. CO<sub>2</sub> is also largely generated by fossil fuel processing and it is a crucial greenhouse gas that affects the climate change [2]. Fossil fuels are at present the major energy resources and are likely to dominate for more several decades. Hence it is essential to continue the use of fossil fuels but also reduce the CO<sub>2</sub> emissions to the atmosphere [3].

The expectations from the Carbon capture and storage (CCS) technologies to help curb the greenhouse gas emissions and ensure a sustainable development of power generation and other energy-intensive industrial sectors are extremely high. Amongst them, chemical looping systems display a promising option to capture CO<sub>2</sub> with lower cost and energy penalty [1]. Chemical looping combustion (CLC) is a promising technology to utilize fossil fuel for combustion and prevent CO<sub>2</sub> dilution in the nitrogen rich flue gases. In this process, a solid oxygen carrier supplies the oxygen needed for CO<sub>2</sub> and water formation that is a nitrogen free product mixture. This avoids the major cost of CO<sub>2</sub> separation from the flue gases and also reduces the NO<sub>x</sub> formation. A good oxygen carrier readily reacts with the fuel gas and shall be reoxidized upon being contacted with oxygen. An oxygen carrier is generally prepared by a metal oxide and an inert binder that provide the oxygen storage, fluidizability and mechanical strength [4]. Chemical looping

combustion (CLC) is a non-flame two-step combustion which produces a pure CO<sub>2</sub> stream for easy compression and sequestration. The process comprises of two interconnected fluidized bed reactors where the fuel reactor is a bubbling fluidized bed and the air reactor is a conventional circulating fluidized bed. It uses an oxygen carrier - a highly-reactive metal particle, to avoid the direct contact of air and fuel during the combustion, to indirectly transport oxygen from the air to the fuel. The products of combustion are kept separated from the rest of the flue gases namely nitrogen and excess oxygen [5]. The CLC system is composed of two reactors, an air and a fuel reactor, as shown in fig. 1

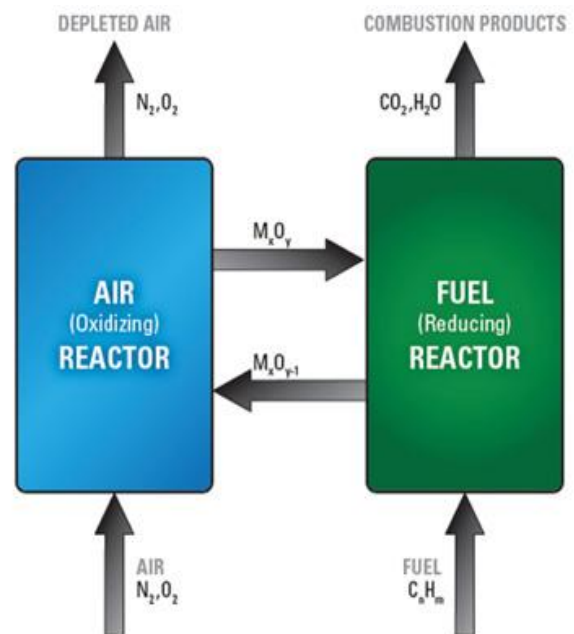
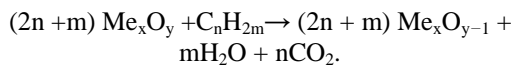
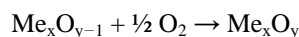


Fig. 1- Chemical-looping combustion system.

In CLC, the solid oxygen carrier is circulated between the air and fuel reactors. The fuel is fed into the fuel reactor where it is oxidized by the lattice oxygen of the oxygen carriers and the reaction in the fuel reactor is almost endothermic.



Once fuel oxidation is completed; the reduced metal oxide  $\text{Me}_x\text{O}_{(x-1)}$  is transported to the air reactor where it is reoxidized according to the reaction:



The flue gas stream from the air reactor will have a high temperature and contain  $\text{N}_2$  and some unreacted  $\text{O}_2$ .

Transition metal oxides, such as Ni, Fe, Cu, and Mn oxides are reported as reactive oxygen carriers. Ni-based oxygen carriers have exhibited the best reactivity and stability during multi-redox cycles [6]. Henrik Leion et al. has investigated the feasibility of using three different solid fuels in chemical-looping combustion (CLC) using NiO as oxygen carrier in a laboratory fluidized-bed reactor system at 970°C. They reported that the NiO particles also showed good reactivity with methane and a syngas mixture of 50%  $\text{H}_2$  and 50% CO also showed good fluidizing properties without any signs of agglomeration [7]. Tobias Mattisson et al. have investigated the feasibility of using NiO as an oxygen carrier with  $\text{CH}_4$  as fuel and in the temperature range 700–1200°C during chemical-looping combustion and their result showed that the yield of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  decreased with increase in temperature [8]. M.M. Yazdanpanah et al. has presented a reactor model for combustion of  $\text{CH}_4$  with NiO/NiAl<sub>2</sub>O<sub>4</sub> oxygen carriers in the fuel reactor of a 10 kW CLC pilot plant. The model shows good agreement with experimental results considering some kinetic and hydrodynamic modifications, including impact of gas adsorption by falling particles in the bubble phase on the gas exchange coefficient and thermodynamic limitation of CO and  $\text{H}_2$  combustion [9]. C. Saha et al. have carried out an experimental investigation pertaining to CLC of a Victorian brown coal using NiO and CuO as oxygen carriers at 950 °C (NiO) and 800°C (CuO) and have reported the high reactivity of CuO as compared to NiO during cyclic operation [10]. Magnus Rydén et al. have examined the Ce, Ca, or Mg stabilized ZrO<sub>2</sub> oxygen carriers by redox experiments in a batch fluidized-bed reactor at 800–950°C, using  $\text{CH}_4$  as fuel. The experiments showed good reactivity between the particles and  $\text{CH}_4$  [11].

Many studies have also demonstrated the feasibility of using CLC for both gaseous and solid fuels [12]. The chemical looping process was mainly

targeted towards efficient carbon capturing [13, 14] and hydrogen production [15, 16]. Rutuja Bhoje et al. have done theoretical study of chemical looping combustion of methane to consider some key technology development points to help the process engineer choose the right oxygen carrier and process conditions [17]. X. Wang et al. have developed a three-dimensional numerical model to simulate the CLC process in the fuel reactor using a bubbling fluidized bed of oxygen carrier made from 14 wt% of metal oxide CuO and 86% wt of inert material  $\text{Al}_2\text{O}_3$  and a coal gas from coal gasification containing 55 vol. %  $\text{CO}$ , 30 vol. %  $\text{H}_2$  and 15 vol. %  $\text{CO}_2$  [18].

India produces approximately 120 million tons of paddies each year which gives around 24 million tons of rice husk and 4.4 million tons of rice husk ash every year. Rice husk is a high calorific value renewable fuel and can be used for electricity generation in efficient manner [19]. Rice husk can be used as fuel and has oxygen in its structure [20]. The CLC of rice husk has not been studied so far in literature.

## II. Process Design:

The conceptual process design for CLC of rice husk is shown in figure 2. The process scheme consists of a CLC fuel reactor and air reactor. Preheated rice husk,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and NiO from air reactor are fed to the fuel reactor in calculated amount to convert the rice husk to  $\text{CO}_2$ , and  $\text{H}_2\text{O}$ . The fuel reactor products were assumed to be in thermodynamic equilibrium at the exit of the reactor. Complete conversion of rice husk and maximum  $\text{CO}_2$  production are targeted in the fuel reactor. It is assumed that the NiO oxidizes the carbon and hydrogen in rice husk to  $\text{CO}_2$  and  $\text{H}_2\text{O}$  in the CLC fuel reactor. The moles of OC (NiO) for fuel reactor are varied depending on the stoichiometric requirement of reaction between rice husk and NiO. Complete conversion of rice husk to  $\text{CO}_2$  and  $\text{H}_2\text{O}$  is desired; however the conversion is limited by thermodynamic equilibrium.

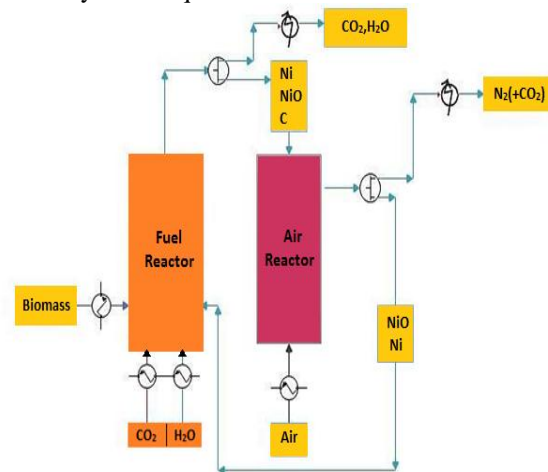


Fig. 2- Process diagram

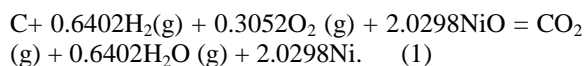
The OC (NiO) is reduced (to Ni) in the CLC fuel reactor. The products of the CLC fuel reactor go through a gas-solid separator and the gaseous products mostly containing CO<sub>2</sub> and H<sub>2</sub>O can be cooled for CO<sub>2</sub> separation and sequestration with heat recovery or recycled back as required. The solid products of the CLC fuel reactor containing OC are transferred to the air reactor, in which preheated air is added to completely oxidize coke to CO<sub>2</sub> and regenerate the reduced OC. Complete conversion is assumed in these oxidation reactions.

The regenerated oxygen carrier (NiO) is separated from the air reactor product stream using a gas-solid separator and is recycled to the fuel reactor. The air reactor is the major source of thermal energy in this process. It is safely assumed that both the air and fuel reactors operate at same temperature and pressure conditions.

Table 1- Rice husk composition [21].

Species	C	H <sub>2</sub>	O <sub>2</sub>
Moles	1	0.6402	0.3052

The base composition of rice husk taken were 1 mole C, 0.6402 moles hydrogen (H<sub>2</sub>), 0.3052 oxygen (O<sub>2</sub>). According to rice husk composition reaction involved in fuel reactor assumed to be



### III. Process Methodology:

Thermodynamic analysis of the system helps us to determine the optimum conditions that can maximize the yield with low energy consumption [22]. HSC Chemistry software version 5.11 is used to generate the thermodynamic equilibrium data for CLC fuel reactor in this process study. The thermodynamic equilibrium calculations in the Gibbs routine of HSC Chemistry are done using the Gibbs free energy minimization algorithm. The Gibbs program searches the best combination of most stable species in which the Gibbs free energy of the system can achieve its minimum at a fixed mass balance (a constraint minimization problem), constant pressure, and temperature. Hence no chemical reaction equations are required in the input.

The chemical species such as C(s), CO<sub>2</sub> (g), H<sub>2</sub> (g), CO (g), H<sub>2</sub>O (g), CH<sub>4</sub> (g), H<sub>2</sub>O (l), O<sub>2</sub> (g), NiO (s) and Ni (s), that are usually found in CLC reaction system considered in this study. The input species fed to the fuel reactor were rice husk with oxygen carrier, H<sub>2</sub>O (g), and CO<sub>2</sub> (g). The material balances are done by Equilibrium Composition module of

HSC Chemistry and the results were used to calculate reaction enthalpy by Reaction Equation module of HSC Chemistry software.

Table 2- Fuel reactor feed condition with NiO in stoichiometric ratio.

Feed condition	Input moles of Rice husk			Input moles of CO <sub>2</sub>	Input moles of H <sub>2</sub> O	GaCR (H <sub>2</sub> O+CO <sub>2</sub> )/C	Input moles of NiO (For 1 stoichiometric ratio)
	C	H <sub>2</sub>	O <sub>2</sub>				
AS1	1	0.6402	0.3052	0	1	1	2.0298
AS2	1	0.6402	0.3052	0.5	0.5	1	2.0298
AS3	1	0.6402	0.3052	1	0	1	2.0298
BS1	1	0.6402	0.3052	0	2	2	2.0298
BS2	1	0.6402	0.3052	0.5	1.5	2	2.0298
BS3	1	0.6402	0.3052	1	1	2	2.0298
BS4	1	0.6402	0.3052	1.5	0.5	2	2.0298
BS5	1	0.6402	0.3052	2	0	2	2.0298
CS1	1	0.6402	0.3052	0	3	3	2.0298
CS2	1	0.6402	0.3052	1	2	3	2.0298
CS3	1	0.6402	0.3052	2	1	3	2.0298
CS4	1	0.6402	0.3052	3	0	3	2.0298

1 mol carbon (in rice husk) is used as basis for all calculations in the temperature range of 500–1200°C for the entire process. The feed gasifying agent-to-carbon ratio (GaCR) ranging from 1 to 3 was used in this study. The CLC of rice husk using both gasifying agent (CO<sub>2</sub> and H<sub>2</sub>O) and oxygen carrier (NiO) is studied with intermediate steps of increase in CO<sub>2</sub> moles (with simultaneous decrease in steam moles) for constant GaCR ratios. These feed conditions for CO<sub>2</sub> and steam input per mole carbon (in rice husk) are shown in Table 2. These inputs were used to calculate the thermodynamic equilibrium composition in the CLC fuel reactor. The oxidation reaction in the air reactor is exothermic.

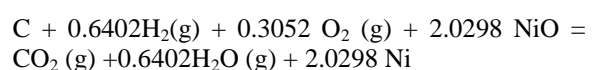
It is presumed that, according to stoichiometric amount given by the reactions the air is supplied to the air reactor:



### IV. Results and Discussions:

#### 4.1. Effect of Amount of oxygen carrier:

The stoichiometric quantity of NiO was varied according to fuel reactor reaction of oxygen carrier:



Stoichiometric amount of NiO reqd. (S) = 2.0298

Assuming S = 2.0298

$$1.25 * S = 2.53725$$

And  $1.5 * S = 3.0447$

The effect of stoichiometric amount of oxygen carrier on CLC of rice husk was investigated for 1, 1.25, and 1.5 stoichiometric of NiO in fuel reactor.

Some side reactions also take place in the fuel reactor and hence the conversions in the fuel reactor are limited by thermodynamic equilibrium constraints. It was necessary to study the equilibrium product composition of the CLC fuel reactor at 1 bar pressure. Since, the OC is generally used in excess of the stoichiometric requirement in CLC processes to enhance the syngas and CH<sub>4</sub> conversion in the fuel reactor. Hence two more cases (1.25S and 1.5S) using higher amounts (1.25 times and 1.5 times the stoichiometric requirement) of OC were also studied.

#### 4.1.1. (CH<sub>4</sub> + CO + H<sub>2</sub>) formation:

The (CH<sub>4</sub> + CO + H<sub>2</sub>) compositions in product gases at 1 bar pressure and at 700°C of the CLC fuel reactor for different inputs of OC and different input feed conditions are shown in Figure 3. It was observed that the H<sub>2</sub>, CO, and CH<sub>4</sub> emissions from the fuel reactor decreased with increase in the amount of OC. At constant GaCR, it was also observed that the (CH<sub>4</sub> + CO + H<sub>2</sub>) moles generally decreased with increase in feed CO<sub>2</sub> moles (with simultaneous decrease in H<sub>2</sub>O moles). The maximum amount of (CH<sub>4</sub> + CO + H<sub>2</sub>) exit moles were found to be 0.19 moles for case CS1, while the minimum quantity of (CH<sub>4</sub> + CO + H<sub>2</sub>) exit moles were observed to be 0.02 moles (AS1).

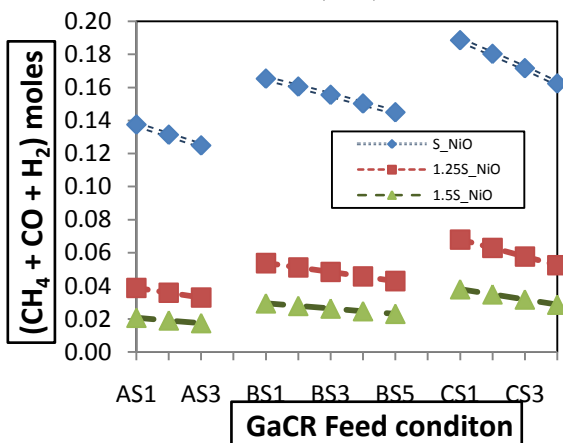


Fig. 3- (CH<sub>4</sub> + CO + H<sub>2</sub>) moles emitted from the fuel reactor at 700°C.

#### 4.1.2. CO<sub>2</sub> Emission:

The CO<sub>2</sub> emissions at 1 bar pressure and 700°C of the CLC fuel reactor for different inputs of OC and different input feed conditions is shown in Fig. 4. It was seen that the CO<sub>2</sub> emission from the fuel reactor increase with increase in the amount of OC. It was also observed that the product CO<sub>2</sub> moles generally decrease with increase in feed CO<sub>2</sub> moles

at constant GaCR. The maximum amount of CO<sub>2</sub> emission was found to be 0.99 moles for case AS1, while the minimum CO<sub>2</sub> emission was observed to be 0.87 moles (CS4).

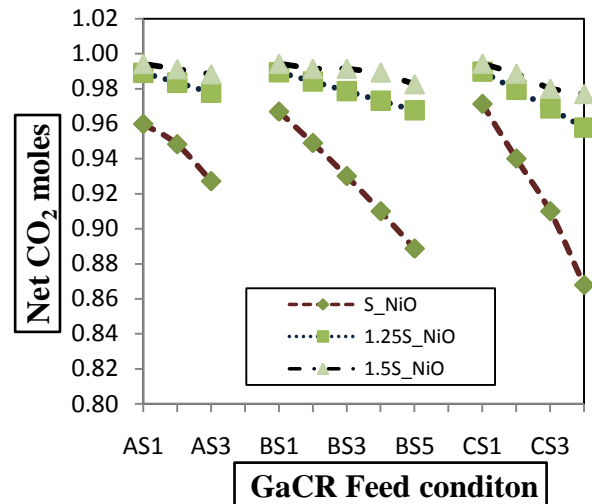


Fig. 4- CO<sub>2</sub> moles emitted from the fuel reactor at 700°C.

#### 4.1.3. Syngas ratio:

The syngas ratio at 1 bar pressure and at 700°C of the CLC fuel reactor for different inputs of OC and different input feed conditions is shown in Fig. 5. It was observed that the syngas ratio remains constant irrespective of increase in the amount of OC. It was also observed that the syngas ratio generally decreased with increase in feed CO<sub>2</sub> moles at constant GaCR. The maximum syngas ratio was found to be 5.73 (CS1), while the minimum of syngas ratio was observed to be 0.25 (CS4). This constant syngas ratio observation is reported for the first time in this paper.

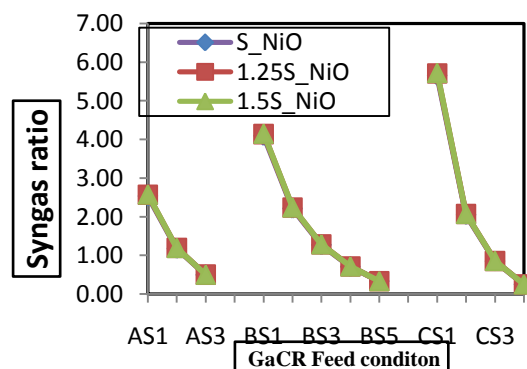


Fig. 5- Syngas ratio at 700°C.

## 4.2 Effect of GaCR Conditions and Temperature.

### 4.2.1. Net CO<sub>2</sub> emission:

The Net CO<sub>2</sub> emissions from the CLC process is used to measure the CO<sub>2</sub> utilization and is shown in Fig. 6. The net CO<sub>2</sub> emission (moles) = CO<sub>2</sub> moles (in output) - CO<sub>2</sub> moles (in input). An increase in



temperature from 500 to 1200°C resulted in decrease in net CO<sub>2</sub> emission. At constant GaCR, increasing the CO<sub>2</sub> moles in the input (with simultaneous decrease in H<sub>2</sub>O moles) decreased the net CO<sub>2</sub> emission in output. The net maximum CO<sub>2</sub> emission found in case CS1 (3 moles H<sub>2</sub>O + 0 moles CO<sub>2</sub>).

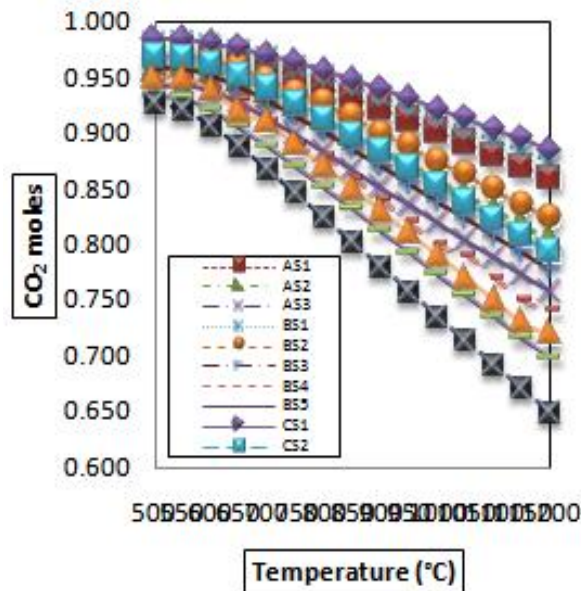


Fig. 6- Net CO<sub>2</sub> emission

#### 4.2.2 H<sub>2</sub> and CO formation:

The H<sub>2</sub> formation increased with increase in temperature from 500 to 1200°C. Increase in the CO<sub>2</sub> moles (with simultaneous decrease in H<sub>2</sub>O moles) in GaCR ratio decreased the H<sub>2</sub> formation in output. The maximum H<sub>2</sub> formation was found in case AS1 (3 moles H<sub>2</sub>O + 0 moles CO<sub>2</sub>) fig. 7a. Similarly, increase in the process temperature led to an increase in CO formation. Increase in the CO<sub>2</sub> in GaCR ratio increased the CO formation in output. The minimum CO formation found in case AS1 (3 moles H<sub>2</sub>O + 0 moles CO<sub>2</sub>) as shown in fig. 7b.

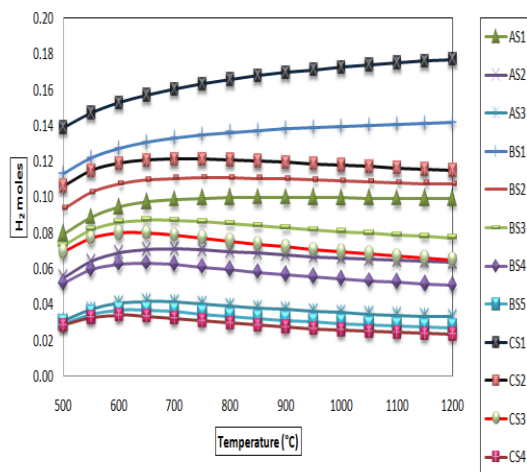


Fig. 7 a- H<sub>2</sub> formation

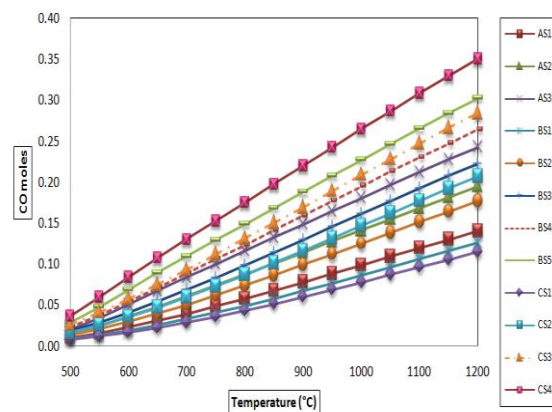


Fig.7 b- CO formation

#### 4.2.3. Syngas Yield:

The syngas yield is calculated by adding the H<sub>2</sub> and CO moles obtained from the process. As seen in Fig. 8, the syngas yield generally increased with increase in temperature from 500 to 1200°C and for constant GaCR. Increase in the feed CO<sub>2</sub> moles led to a decrease in syngas yield, e.g. the syngas yield increased from 0.09 to 0.24 moles for case AS1, and the syngas yeild increased from 0.06 to 0.37 moles for case AS4 and 0.06 to 0.33 for BS5.

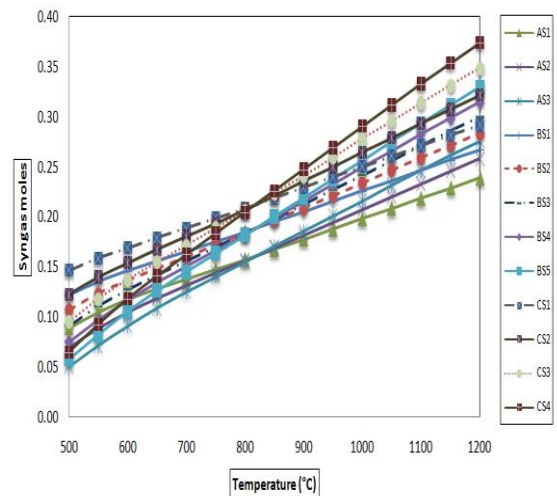


Fig. 8- Syngas Yield.

#### 4.2.4. NiO conversion:

It is assumed that the CH<sub>4</sub>, CO, and H<sub>2</sub> are oxidized to CO<sub>2</sub> and H<sub>2</sub>O by means of NiO in the CLC fuel reactor. The oxygen carrier (NiO) gets reduced in the CLC fuel reactor. The NiO conversion slightly decreased with increase in temperature from 500-1200°C. For e.g. the NiO conversion decreased from 93.75% to 88.23% in case of AS1 while it decreased from 93.21% to 81.57% in case of CS4 as shown in fig. 9.

The carbon conversion at 1 bar pressure is shown in Table 2. It was seen that as the temperature increased from 500–1200°C, the carbon conversion reached its maximum (100%). It was observed that

the 100% carbon conversion was achieved at relatively low temperature for higher GaCR.

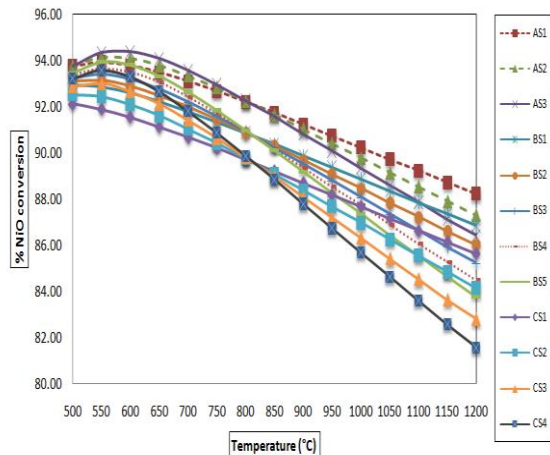


Fig. 9. %NiO Conversion.

#### 4.2.5. 100% carbon conversion in fuel reactor:

It is very important for the CLC process to get 100% carbon conversion in the fuel reactor as the unreacted carbon can go to in air reactor and form CO<sub>2</sub>. The lowest temperature for 100% carbon conversion was found to be 900°C in case CS1, so this point was found to be the best point for process operation.

Table 2- Carbon Conversion

% C conv.	500	550	600	650	700	750	800	850	900	950	1000	1050	1100	1150	1200
AS0	96.38	97.82	98.78	99.36	99.66	99.82	99.90	99.94	99.97	99.98	99.99	99.99	99.99	100.00	100.00
AS1	98.12	99.06	99.54	99.77	99.88	99.93	99.96	99.98	99.99	99.99	99.99	100.00	100.00	100.00	100.00
AS2	97.16	98.48	99.23	99.62	99.80	99.90	99.94	99.97	99.98	99.99	99.99	99.99	100.00	100.00	100.00
AS3	96.19	97.80	98.85	99.43	99.72	99.86	99.92	99.96	99.98	99.98	99.99	99.99	100.00	100.00	100.00
BS1	98.95	99.52	99.77	99.89	99.94	99.97	99.98	99.99	99.99	100.00	100.00	100.00	100.00	100.00	100.00
BS2	98.34	99.21	99.62	99.81	99.90	99.95	99.97	99.98	99.99	99.99	100.00	100.00	100.00	100.00	100.00
BS3	97.69	98.85	99.45	99.73	99.86	99.93	99.96	99.98	99.99	99.99	99.99	100.00	100.00	100.00	100.00
BS4	96.99	98.43	99.24	99.64	99.82	99.91	99.95	99.97	99.98	99.99	99.99	100.00	100.00	100.00	100.00
BS5	96.24	97.94	98.98	99.52	99.77	99.89	99.94	99.97	99.98	99.99	99.99	100.00	100.00	100.00	100.00
CS1	99.35	99.71	99.86	99.93	99.96	99.98	99.99	99.99	100.00	100.00	100.00	100.00	100.00	100.00	100.00
CS2	98.54	99.33	99.69	99.85	99.92	99.96	99.98	99.99	99.99	99.99	100.00	100.00	100.00	100.00	100.00
CS3	97.55	98.81	99.44	99.74	99.87	99.93	99.96	99.98	99.99	99.99	100.00	100.00	100.00	100.00	100.00
CS4	96.35	98.09	99.10	99.59	99.81	99.91	99.95	99.97	99.98	99.99	99.99	100.00	100.00	100.00	100.00

## V. Conclusion

This theoretical study was done to understand the material balances for process design of direct CLC of rice husk. The study considered the effect of temperature and effect of amount of oxygen carrier in the fuel reactor using steam and CO<sub>2</sub> as gasifying agents in steps. It was concluded that rice husk CLC using 3 moles H<sub>2</sub>O and 0 moles CO<sub>2</sub> per mole carbon at 1 bar pressure and 900°C is the best point to operate the process as hundred percent carbon conversion in the rice husk occurs in the process. The results obtained in this detailed study can be used for

scale up of the process. Further Experimentation is required for the commercialization of the technology.

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