



# Chemical looping combustion of biomass for renewable & non- CO<sub>2</sub> emissions energy- status and review

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## Abstract

World depends on fossil fuel combustion for thermal energy generation. Fossil fuel combustion leads to the generation of CO<sub>2</sub> and extinction of non-renewable resources. To meet the future energy demands replacement of existing technologies should take place in the view of large quantities of GHG's emissions from fossil fuels and their extinction. Chemical looping combustion (CLC) is primarily a combustion technique with an inherent separation of CO<sub>2</sub> from the flue gases. Due to its advantage of negative CO<sub>2</sub> emissions, chemical looping combustion got attention of many researchers since last one and half decade. Recent research advancements in the CLC provided a platform for further research and developments in chemical looping combustion of biomass. This paper reviews the CLC of biomass to present the overview of chemical looping combustion technology and its status of biomass utilization as a fuel in CLC reactors.

**Keywords:** Biomass; CO<sub>2</sub> Emissions; Chemical Looping Combustion (CLC); Fossil Fuel Combustion; Renewable Energy.

## 1. Introduction

### 1.1. Energy demand

Throughout the world per capita energy demand is increasing specially in Asian countries. Urbanisation and Population Growth are playing key role in high rate of energy consumption. Since 2000 year electricity utilisation in India almost doubled to 2013 with an average increase of 6.9%. Govt. of India announced programmes like Make in India, 24/7 power supply and Deen Dayal Upadhyaya Gram Jyoti Yojana (DDUGJY) which causes more demand for electricity demand in coming years [1]. India's total power installed capacity increased to 310GW out of which 69.4% is from Thermal power [2]. India every year producing more than 650 million tons of coal and importing more than 200 million tons to meet our demands which is leading govt to spend more on our imports [3]. So, to meet the above energy demands and to reduce the imports of coal it is necessary to use the alternative energy sources.

### 1.2. GHG's emissions

Combustion of fossil fuel leads to increase in atmospheric Greenhouse Gases (GHG's) concentrations. Out of total released GHG's CO<sub>2</sub> is contributing 76%. A total increase of 40% observed in atmospheric CO<sub>2</sub> concentrations after 1750 [4]. India is the third largest country in terms of CO<sub>2</sub> emissions with 6.23% share of global emissions after China and USA [5]. To control rise in global temperatures and GHG's concentrations Paris Climate Change agreement has come into action from 2015. In order to meet the guidelines of Paris Agreement 2015, greenhouse gas emissions need to be controlled and India has committed to reduce GHG

emissions to 33-35% of 2005 emissions by 2030 [6,7,8]. So, it is inevitable to reduce the GHG emissions from anthropogenic activities.

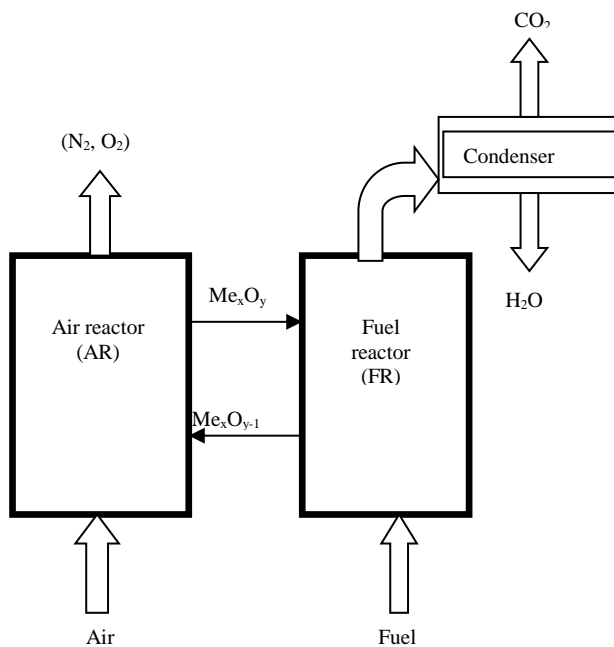
### 1.3. Waste management system

In disposing waste most favored process which will have minimal impact is source reduction. It can be done through modified life style of being aware of waste generated and its consequences on environment. Biomass waste is generated due to essential need of survival of human being i.e food. So, alternative way of providing substitute for biomass is a bit difficult task. Other favored process is Recycling and Reuse. It majorly provides minimization procedure of conversion, increasing efficiency and energy recovery in another form. Biomass waste recycling is done with composting and by making natural fertilizer. It mostly contains organic matter which can be degraded with the help of micro-organism. But the quantity of biomass available and conversion of it in compost takes long time. On the other hand, Energy recovery in the form of waste to energy (WtE) plant is a feasible option for biomass disposal. Biomass can be replaced with coal which is a renewable source of energy. Biomass Waste generated in India is around 500 million metric tons per year [9]. Disposal of such huge Biomass waste by conventional method produce a lot of carbon dioxide gas contributing to Green House Gases (GHGs) affecting atmospheric gas composition and phenomenon responsible for ozone layer depletion. Particularly in case of WtE plant can bring waste residue reduction and maximum energy production where clean energy is being produce through advance reformed technology of Chemical Looping Combustion (CLC).



## 2. Chemical looping combustion

Chemical-looping combustion (CLC) technology have the potential to make the carbon capturing process feasible [10]. A conventional CLC system will consists of two reactors, namely fuel reactor and air reactor. The preheated oxygen carrier will be sent to the fuel reactor along with fuel such as Coal, Syngas, CH<sub>4</sub>, Biomass etc. In fuel reactor the fuel will react with the oxygen carrier, which is generally an endothermic reaction. The products obtained from this will be reduced oxygen carrier and flue gas mixture stream. This flue gas mixture will have the dominant portions of CO<sub>2</sub> and H<sub>2</sub>O. Later this gas stream will be sent to condenser to separate the steam from it. The reduced oxygen carrier and ash mixture will be drive to the air reactor or riser for regeneration of oxygen carrier and is highly exothermic in nature. In air reactor, air will be supplied to the particles for regeneration. As a result of this reaction, regeneration of oxygen carrier will be taken place and heat will be released. This released heat can be used for generation of power etc. The schematic representation of CLC process has mentioned below Fig. 1.



**Fig. 1:** Schematic Representation of Chemical Looping Combustion (CLC) Process.

Initially CLC was patented as a technique to produce pure CO<sub>2</sub> in 1954 by Lewis and Gilliland [11]. At the end of 20<sup>th</sup> century, it was recognized as a potential technique to capture CO<sub>2</sub> from fuel combustion [12]. Later, it was also recognized as a potential technique to control CO<sub>2</sub> emission from thermal power plants. The research works on chemical looping combustion has been summarized in different review articles based on process [13], [14], [15], Solid fuels [16], [17], use of natural ores as oxygen carriers [18] etc. Adanez, J., et al. published a broad review over utilization of 700 oxygen carriers in CLC process [19]. Most of the experiments have been carried out in batch conditions. Since, past few years some organizations are working on continuous CLC systems. This paper is concentrating only on research work carried out on CLC with different biomasses as solid fuels.

## 3. Review of the biomass chemical looping combustion (CLC)

Biomass utilization in CLC as an solid fuel was introduced by Shen L, et al in 2009 [20] until then Chemical Looping Combustion of solid fuels was not given importance. From the beginning

of 21<sup>st</sup> century many experiments have been conducted on CLC of solid fuels with different materials like Victorian brown coal, pine saw dust, wood pallets etc. After getting the importance of renewable energy and solid waste management, researchers started focusing on use of biomass in CLC. With all the available articles from different sources this review has been categorized into different processes CLC and presented below:

### 3.1. Theoretical studies

To demonstrate the performance of both reactors and process simulation a multistage model was developed and simulated by Li et al. using ASPEN plus software for renewable energy using biomass in biomass direct chemical looping (BDCL) and compared the efficiencies with conventional techniques [21]. To reduce the GHG's emission and to encourage utilization of renewable fuels a detailed thermodynamic and environmental aspects analysis has been conducted by Fan J. et al of biomass and coal co-fuelled gasification chemical looping combustion for combined cooling, heating and power (BCCLC-CCHP) generation system [22]. HSC Chemistry 6.0 software was used for thermodynamic simulation of pine saw dust to optimize the carbon conversion and carbon capture efficiency and results were interpreted on the basis of reactivity, thermal stability and catalytic reactivity for tar cracking and reforming [23].

### 3.2 Direct CLC of biomass

Since the production of biomass is in large quantity it can be a substitute for fossil fuels for thermal energy production researchers started using different forms of biomass as a fuel for CLC. To reduce the cost of CLC process Huang, Z., et al. investigated the performance of natural hematite in continuous reaction as an oxygen carrier in Biomass direct looping combustion (BDCL). Due to attrition and structural changes in hematite, carbon conversion rate decreased from 87.63% in 1<sup>st</sup> cycle to 77.18% in 20<sup>th</sup> cycle [24]. Kobayashi et al. has discussed many things like design, potential challenges and feasibility assessments in biomass direct looping combustion process (BDCL) to convert biomass to hydrogen and /or electricity to achieve more conversion efficiency than conventional techniques and to get electricity at less price comparative to fossil fuel processes [25]. Optimizing the CLC process for high fuel conversion without auxiliary fuel experiment was conducted with rice husk as a fuel in annular dual-bed moving bed reactor (ADMBR). The fuel conversion efficiency was reached to a maximum of 92% while the heat demand is just 54% of heat utilized by the gasification of rice husk [26]. Wang, X., et al performed chemical looping combustion using thermo gravimetric analyzer and laboratory scale fluidized bed on pine saw dust with different metal ferrites synthesized by sol-gel method as an oxygen carrier and concluded that CuFe<sub>2</sub>O<sub>4</sub> and CoFe<sub>2</sub>O<sub>4</sub> are suitable oxygen carrier for biomass [23]. To test the effect of naturally available ores sintered manganese ore was used in biomass CLC process. Results of the experiments gave positive sign for full scale application with a life time varying from 100-400hr [27]. Experiments on pine saw dust conducted in a fluidized bed combustor using CuO supported by olivine as oxygen carrier found that carbon conversion rate and carbon capture efficiency are directly proportional to temperature of the reaction and oxygen carrier to fuel ratio. CLC is a low cost carbon capture technology compared to all other Carbon Capturing and Sequestration technologies [28]. Alonso, M., et al conducted experiments of CLC with wood pallet as a solid fuel for in-situ CO<sub>2</sub> capture by using CaO. With several series of tests conducted to check the efficiency of combustor-calciner for different conditions like solid circulating rates, excess air supply etc. resulted in 100% Carbon conversion and 70-95% carbon capturing [29]. It proved that capturing of in-situ CO<sub>2</sub> using CaO is one of the favorable method. Highly volatile components of biomass are affecting the oxygen demand required for stoichiometry condition. In order to analyze the trend in oxygen demand with change in temperature, these volatiles are interrupting the

combustion process. They also concluded that NO<sub>x</sub> and pollutants were not interfering the process [8]. Conversion of volatiles into CO<sub>2</sub> was controlled by gas- solids mixing and char gasification was influenced by fuel feeding rate and recirculation rate of bed material.

### 3.3. CLC of gas obtained from biomass

Chemical looping combustion of solid fuels concept faded due to the interference of ash with oxygen carrier, loss of oxygen carrier with ash separation and the gentle natured gasification of char [16]. Some research works have been concluded that direct CLC of solid fuels lead to the formation of unconverted char in the fuel reactor and burning of fuel in the air reactor [30]. Thus, to reduce the above mentioned difficulties a new technology named In-situ Gasification Chemical Looping combustion (IGCLC) was introduced where the gas released from the gasification of solid fuels will be used as a fuel in CLC process [31]. Adánez-Rubio I., et al demonstrated CLOU concept for the first time with biomass as fuel in a continuous CLC unit using milled pine wood chips with 60% of CuO contained oxygen carrier prepared by spray drying method where full conversion of biomass has been taken place [32]. Results of the Adánez-Rubio, I., et al gave confidence for other researchers to work on the CLC of biomass. Chemical looping combustion of product gas obtained from the gasification of biomass was done using double fluidized bed. All gases except CH<sub>4</sub> in the product gas were converted into flue gases and no performance drop observed during the period of 10hr continuous operation [33]. Cu and Ni supported on ceria are found to be effective oxygen carriers for the combustion of syngas obtained from gasification of biomass [34]. In situ Gasification-Chemical Looping Combustion (iG-CLC) of different biomass residues namely pine sawdust, olive stone and almond shell between temperature ranges 900-980°C with iron ore as oxygen carrier and observed 100% CO<sub>2</sub> capture efficiencies at temperatures above 950°C for all three biomasses [35]. To get the good quality of biochar hyphenation of Biomass torrefaction was experimentally studied by Sarvaramini et al. Iron oxide oxygen carrier was used in CLC process where 99% of volatiles converted into CO<sub>2</sub> between 400 to 600°C because of birch wood torrefaction between 260 and 300°C [36]. Feasibility of iron oxide as an oxygen carrier has been tested by Dong, C., et al in a fluidized bed reactor using CO and biomass gas fuels at different temperatures and reactivity tested during repetition of cycles for CO<sub>2</sub> capture [37].

### 3.4. CLC of biomass in mix with others

Emissions from thermal power plant(TPP)'s and emission standards became a rationale for adopting CCS in TPPs [38]. In this regard feasibility studies of CLC application in TPPs got momentum. To control the over resting on fossil fuels and GHG's emissions from thermal power plants some researchers used renewable fuel biomass with coal [39]. Synergetic effect observed during co-combustion of coal with candlenut wood and K<sub>2</sub>CO<sub>3</sub> and candlenut wood ash mix addition to the fuel helped to decrease gasification temperature and increase of coal conversion [40]. One of the major drawbacks in CLC is cost of oxygen carrier. To overcome this situation research on utilization of natural ores as oxygen carriers got attention. Iron ore from Tierga, Spain was used as a oxygen carrier in Chemical looping combustion of solid fuels (i.e. bituminous coal and wood char) has been studied and reported as the maximum gas conversion efficiencies are 87% and 93% respectively. Also the life time of the oxygen carrier was expected to be 300hrs, for which further studies needed [10]. Increase in temperature of fuel reactor increased the CO<sub>2</sub> concentration in flue gases as well as CO concentration in case of biomass and coal mixture.

### 3.5. Effect of biomass on oxygen carriers

Economically feasible process is necessary for practical application. To know the feasibility of biomass CLC process several researches has been conducted on effect of biomass on oxygen carrier and concluded as efficiency of carbon conversion in CLOU is higher for CaMn<sub>0.9</sub>Mg<sub>0.1</sub>O<sub>3-δ</sub> OC than compared to ilmenite [41]. Ilmenite addition reduced the concentration of CO and total hydrocarbons in CLC of volatile rich biomass [42]. Surface morphology of the CuO particles observed due to the interactions between the metal oxygen carrier and ash [40]. Sintering of Cu particles observed at 800°C during its reaction with syngas obtained from biomass gasification [34]. Fuel conversion rate was significantly influenced by the biomass ash and sintering observed due to the SiO<sub>2</sub> rich wheat straw ash (WSA) [43]. Indicating that ash in the CLC process is causing lot of problems like changes in structural changes, reduction in regeneration of OC, less reactivity with fuel etc. Blending of biomass with coal along with some additives might decrease the negative influence of biomass on iron oxide [44]. Several researches has been done on CLC of Biomass and proved that CLC of biomass has future scope in controlling GHG's emissions, those list of institutes with details given below in Table 1.

**Table 1:** Institute Names and Oxygen Carriers Used for Biomass CLC

Institute Name	Fuel used	Oxygen carrier	Year
Guangzhou Institute of Energy Conversion, China [24]	Biomass	Natural Hematite	2013
National Taiwan University of Science and Technology, Taiwan [26]	Rice Husk	Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub>	2015
Huazhong University of Science & Technology, Henan Normal University, China [23]	Pine Saw dust	Metal Ferrites	2016
Laval University, Canada [36]	Birch wood	Iron oxide	2013
Southeast University, China [44]	Biomass and Coal	Australia Iron Ore	2011
Vienna University of Technology, University of Natural Resources and Life Sciences, Austria [33]	Standard Wood pellets	Synthetic Oxygen Carrier of 9% CuO	2014
Instituto de Carboquímica-ICB-CSIC, Spain [35]	Pine sawdust, Olive stone and almond shell	Iron Ore	2018
Xi'an Jiaotong University, People's Republic of China [40]	Yimin coal, candlenut wood and rice hull	CuO	2013
Chalmers University of Technology, Sweden [10]	Bituminous coal and wood char	Iron ore	2016
Chalmers University of Technology, Sweden [41]	Biochar	Perovskite structure CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3-δ</sub>	2016
Southeast University, China[20]	Biomass	Iron Oxide	2009
Chalmers University of Technology, Sweden [45]	Commercial wood pellets	Ilmenite and Manganese Ore	2017

## 4. Conclusion

Biomass as a solid fuel in Chemical looping combustion process is a feasible option for utilization of renewable energy in more efficient way. Direct use of biomass in CLC process proved that it has 100% potential of carbon conversion. While other processes like CLOU and syngas obtained from biomass are overcoming the problem of sintering of mixture which reduces the performance of CLC. Many experiments carried out on synthesized oxygen

carriers showed better result. CLC of biomass has better scope in controlling GHG emissions.

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