Chemical looping

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Chemical looping technology in general, is the rising star in chemical technologies, which is capable of low CO2 emissions with applications in the production of heat, fuels, chemicals, and electricity. This entry discusses the technology in general, gives an overview of some **pilot scale plants** and the **different chemical looping processes** with focus on the **production of heat and chemicals**, highlights the importance of the development of **oxygen carrier materials** with suitable **properties**,

Keywords: chemical looping ; oxygen carrier ; hydrogen ; CO2 utilization ; metal oxide ; carbon capture and storage ; reforming ; fossil fuels

1. Introduction

Carbon dioxide emissions resulting from combustion and other chemical processes gravely impact the environment. Therefore, alternative processes have been developed in which these CO_2 emissions can be avoided. Among current and emerging technologies for CO_2 capture, chemical looping combustion (CLC) was frequently mentioned as a particularly promising approach to combining CO_2 capture and energy production[1][2][3]. This technology can also be included in the oxyfuel combustion branch of CCS, as in this process also, all diluting components of the air are separated before the combustion of the fuel. The main difference with conventional oxy-fuel capture is the avoidance of a separate costly air separation unit. Pure oxygen is separated from the air inside the chemical looping process itself, by the utilization of metal oxides, which selectively transfer oxygen from the air to the fuel. These oxygen transfer materials are hence commonly called ‘oxygen carriers’ (OC). CLC has gained significant maturity during the last decades, resulting in kW_{th} and MW_{th}-scale operation at various locations throughout the world (see Table 1). Chemical looping technology in general, is the rising star in chemical technologies, which is capable of low CO2 emissions with applications in the production of heat, fuels, chemicals, and electricity. Several aspects are being considered in the current transition and scale-up of the technology to a
level appropriate for industrial implementation. One key critical aspect is the presence of a suitable, sustainable, and cost-e ective oxygen carrier material with the right properties for the specific chemical looping application.

The origin of chemical looping technology is said to start way back in the year 1950 when Lewis and Gilliland filed their patent entitled ”production of pure carbon dioxide”[4]. In this patent, an oxidizable carbonaceous material was oxidized by copper oxide particles to produce carbon dioxide free of inert gases, such as nitrogen. The term ‘oxygen carrier’, which is still used to denote the solid materials that transfer the oxygen from oxidizing agent to fuel, was already introduced then[4]. The term ‘chemical looping’ was derived by Ishida et al. in the second half of the 1980s from the different oxidation and reduction reactions through the oxygen carrier loops, yielding net combustion of the fuel[5]. Since then, the technology has matured significantly. The process has been scaled up to MW_{th}-scale, more than 600 oxygen carrier materials have been developed, and these research activities have resulted in a few thousand publications and several review papers across all domains ranging from reactor design, oxygen carrier design for both combustion and chemicals production to scale up, and operational experience in the units across the world[6][7][8][9][10][11][12][13][14][15][16][17][18][19][20][21][22][23][24][25][26][27]. Five international conferences dedicated specifically to chemical looping have been organized during the past 10 years all over the world.

Figure 1 gives an overview of all relevant aspects of chemical looping. Different types of carbonaceous fuels are used, depending on the focus of the process. While solid fuels are mainly used for energy production, gaseous fuels are interesting for the production of chemicals. In chemical looping processes air is used for replenishing the oxygen in the oxygen carriers. In addition to air, carbon dioxide and steam can also be used. In this way pure CO and H_2 can be generated. As the reactions between the oxygen carrier and CO_2 or H_2O are endothermic, changing the oxidizing agent in the chemical looping process has a large impact on the energy balances of the process. Different reactor systems can be used in chemical looping processes, ranging from packed-bed reactors to interconnected fluidized-bed systems. Each reactor concept has advantages and disadvantages, and their suitability depends also on the reactivity, composition and the properties of the oxygen carrier materials[6].

Table 2. Select fluidized bed chemical looping process development and pilot plants across the world, adapted from [28].

Institution	Location	Year	Capacity (kWth)	Ref
Vienna University of Technology	Vienna, Austria	2009	120	[29]
Hamburg University of Technology	Hamburg, Germany	2012	25	[30]
Chalmers University of Technology	Gothenburg, Sweden	2012	100	[31]
Darmstadt University of Technology	Darmstadt, Germany	2012	1000	[32]
Southeast University	Nanjing, China	2012	50	[33]
University of Utah	Salt Lake City, USA	2012	200	[28]
National Energy Technology Laboratory	Morgantown, USA	2013	50	[34]
Instituto de Carboquímica (ICB-CSIC)	Zaragoza, Spain	2014	50	[35]
Huazhong University of Sci. and Tech.	Wuhan, China	2016	50	[36]
VTT Technical Research Center	Espoo, Finland	2016	50	[37]
Japan Coal Energy Center	Tokyo, Japan	2017	100	[38]
Korean Institute of Energy Research	Daejeon, Korea	2018	500	[39]

Figure 1. A general overview of the parts of the chemical looping process.

2. Oxygen Carrier Materials

A key issue in the further development of this rising star in chemical technologies and its introduction to the industry is the selection and further development of an appropriate oxygen carrier (OC) material [6]. This solid oxygen carrier material supplies the stoichiometric oxygen needed for the various chemical processes. Its reactivity, cost, toxicity, thermal stability, attrition resistance, and chemical stability are critical selection criteria for developing suitable oxygen carrier materials[40]. To develop oxygen carriers with optimal properties and long-term stability, one must consider the employed reactor configuration and the aim of the chemical looping process, as well as the thermodynamic properties of the active phases, their interaction with the used support material [41][42], long-term stability, internal ionic migration[6], and the advantages and limits of the employed synthesis methods[6].

3. Different Focus of Chemical Looping Processes

Commonly, chemical looping is used to denote cyclic processes that use a solid material, which circulates the oxygen required for the conversion of a fuel. This solid material is hence called ‘oxygen carrier’ and consists traditionally of metal oxide particles. To close the chemical loop, the oxygen-depleted solid oxygen carrier must be re-oxidized before starting a new cycle conventionally with the use of air. When the goal of the process is energy production, the fuel is converted to total oxidation products (CO_2 and H_2O), and the oxygen-depleted solid must be regenerated by the O_2 in air. The process is then known as chemical looping combustion (CLC) (See Figure 2 for a generalized process scheme). The large advantage of CLC is the inherent separation of the N_2 from the oxidizing air and the produced CO_2 in the process. The H_2O that is still present in the flue gas can easily be condensed, and a pure CO_2 stream is obtained without additional separation costs, such as needed in post-combustion CCS[18]. There is also no need for an expensive air separation unit, such as used in conventional oxy-fuel combustion[43].

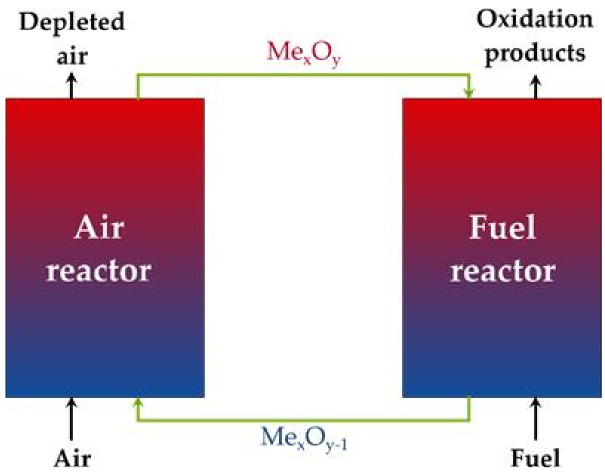

Figure 2. General flowsheet of the chemical looping process.

When chemical looping technology started to gain more attention from researchers, it was predominantly being developed for efficient combustion of fuels, such as coal or natural gas. This technology is commonly called chemical looping combustion, and it is currently, after several decades of research, gaining maturity during the pilot and semi-industrial scale[28].

More recently, however, the focus of chemical looping is shifting more towards the production of hydrogen and other chemicals instead of energy [22][44][45][46][47][47][48]. In this way, products with more economic added value can be generated, which increase the economic viability of the technology in the current context, even at smaller scales. This is expected to facilitate the introduction of the technology into industry. When the focus of the process shifts more towards the production of chemicals, oxygen carrier materials are regenerated by other oxidizing agents instead of air, such as CO_2 [49][50][51][52] or even H_2O [26][49][53][54], with respective productions of CO and H_2. Some of the different chemical looping technologies, which can be found in the literature, are included in Table 2, as well as the used primary fuels, the abbreviation of the CL-branch technology, and the reactor types used. Fuels that are used in chemical looping include coal, liquid fuels, biomass, and natural gas[6]. For characterization purposes, sometimes, syngas or hydrogen is used, but these are not used at an industrial scale[6]. While the initial focus of CLC was the combustion of gaseous fuels, now, gaseous fuels are predominantly converted to chemicals, while combustion for energy production is more frequently executed with solid-fuels (see Table 2). There is, however, some overlap.

Table 2. An overview of some of the different chemical looping technologies found in the literature.

Focus	Primary Fuel	Process		Reactor Type
Combustion	Gas	CLC	Chemical Looping Combustion	f[1], m[2], p[3]
-	Solid	Syngas-CLC	Syngas-Chemical Looping Combustion	f, m, p
-	Solid	*i*G-CLC	In situ Gasification Chemical Looping Combustion	f, m
-	Solid	CLOU	Chemical Looping with Oxygen Uncoupling	f
-	Gas	GSC	Gas Switching Combustion	f

Chemicals	Gas	SR-CLR	Steam Reforming-Chemical Looping Reforming	f
-	Gas, liquid	a-CLR	Autothermal-Chemical Looping Reforming	f
-	Gas	GSR	Gas Switching Reforming	f
-	Gas	CSR	Chemical Switching Reforming	f
-	Gas, liquid	SE-CLSR	Sorption Enhanced-Chemical Looping Steam Reforming	f, m, p
-	Gas	CLHG/TRCH/OSD	Chemical Looping for Hydrogen Generation/Three Reactor Chemical Looping/One Step Decarbonization	f, m, p
-	Solid	SCL	Syngas Chemical Looping	m
-	Solid	CDCL	Coal Direct Chemical Looping	f, m
Oxygen Production	/	CLAS	Chemical Looping Air Separation	f, m, p

[1] f: fluidized bed, [2]m: moving bed and [3]p: packed bed.

4. Outlook

Despite the extensive research in recent years, still, several topics related to chemical looping and oxygen carrier development call for further investigation:

- Further improving the long-term stability and maintaining the high activity of oxygen carriers.
- Developing novel oxygen carriers where optimal materials are combined and where the composition of the active phase is selected and modified in such a way that inherently a high selectivity can be obtained.
- Developing oxygen carriers synthesized with minimal cost, e.g., by starting from impure raw materials or by further developing routes from naturally occurring ores, and checking the effect of the actual use of relevant impure materials on the oxygen carrier performance.
- Developing novel oxygen carrier shapes that can be suitable for use inside pressurized reactor systems, where chemical looping has interesting opportunities, utilized integrated into a system.
- Optimizing oxygen carrier chemistry for use at higher pressures.
- Developing oxygen carriers suitable for catalyst-assisted chemical looping (e.g., see [51][55][56]).

The authors expect that the interaction between catalysis and chemical looping will make significant changes to the chemical looping landscape in the following years and, because of its potential benefits,
deserves extensive attention from the research community.

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