Applied Energy Journal - Special Issue: Chemical Looping: The 3rd International Conference on Chemical Looping 2014

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Chemical-looping combustion (CLC) is a fundamentally new principle of combustion. In conventional combustion the fuel and the combustion air are in direct contact, thus allowing the oxygen of the air to react with the fuel, whereas in chemical-looping combustion air and fuel are kept separate, and the oxygen is transferred from the air to the fuel by use of an oxygen-carrier material. Normally particles containing a suitable metal oxide are used as oxygen carriers, and these particles are circulated between two interconnected fluidized beds, acting as air and fuel reactors. In the fuel reactor the fuel reacts with the oxygen carrier to CO₂ and steam, thus removing oxygen from the oxygen carrier, and in the air reactor new oxygen is added to the oxygen carrier as it is regenerated by the air.

The outlet gas from the fuel reactor consists ideally of CO₂ and H₂O, and the latter is easily removed by condensation, thus yielding a stream of CO₂ without the need of any active gas separation. The possibility of avoiding the large energy penalties and equipment costs of gas separation inherent in other CO₂ capture technologies, has created a large interest in chemical-looping combustion in recent years.

Considerable research has been conducted on CLC in the last ten years with respect to oxygen carrier development, reactor design, modelling and prototype testing. Today more than a thousand oxygen-carrier materials have been investigated in laboratory and the technology has been demonstrated in actual operation in more than twenty chemical-looping combustors in the size range 0.3 kW – 3 MW, using different types of oxygen carriers based on oxides of the metals Ni, Co, Fe, Cu and Mn. The total time of operational experience is more than 7500 hours. From these tests it can be established that 100% CO₂ capture and complete conversion of the fuel is possible using gaseous fuels. Early work focused on gaseous fuels, but now the direct application to solid fuels is rapidly gaining interest as is evident from this issue.

In addition to the direct use of gas, solid fuel or liquid fuel in the chemical-looping combustion, there are also a number of process concepts for using the technology for hydrogen production, chemical-looping reforming technologies. Moreover, reactor systems other than fluidized beds have been proposed.

The application to solid fuel could be important for climate mitigation in view of the large fraction of greenhouse gases coming from coal-fired power plants, as well as for the potential application of CO₂ capture from biomass, which could offer a route for removal of CO₂ from the atmosphere at moderate cost. Ideally, a CLC system for solid fuels could be similar to a commercial boiler based on circulating fluidized-bed combustion, thus having little added equipment costs as well as a minimum energy

penalty for achieving a CO₂ stream. Thus, CLC clearly has a potential for a unique and dramatic reduction of the CO₂ capture cost. But to realize the potential of this process for solid fuels significant development and scale-up remains to be accomplished.

According to the above background and in support of the development of chemical looping technology, this Special Issue, similar to the previous special issue in Applied Energy [1], contains nominated contributions to the "3rd International Conference on Chemical Looping", held in Gothenburg (Sweden). 168 participants from 22 countries took part in the conference, 4 keynotes lectures, 76 oral presentations were held and 36 posters were presented, and totally 85 papers were submitted and distributed, covering all topics related to chemical looping combustion. The series of international conferences on chemical looping started in Lyon (France) in 2010 and it is organized every second year. The second conference was in Darmstadt (Germany) in 2012. Next chemical-looping conference will be held in Nanjing (China) September 26-28, 2016.

In the frame of the 3rd Chemical Looping Conference 2014, a number of subjects relating to chemical-looping were covered and the 76 oral presentations were made in 20 sessions, normally with 5 papers each. Three sessions were given on "Oxygen Carriers", whereas two sessions were given on "Pilot Operation of Solid Fuels", "Hydrogen Production", and "Fluid-dynamical modelling". Furthermore sessions were given on "Pilot Operation of Gaseous and Liquid Fuels", "Packed Bed Processes", "Process Design and Techno-Economic Studies", "Process Modelling", "SO₂ and NO_x", and "Related Processes".

We are pleased to bring the best and recent advancements on chemical looping research to the scientific community in this compact, peer-reviewed Special Issue. The scientific committee and the session chairs of the conference selected the best oral and poster presentations to be invited to submit their contributions to robust critical review in Applied Energy Journal. 48 manuscripts were submitted and reviewed by recognized and experts reviewers. The manuscripts of high-quality that can make an explicit contribution to the technical and scientific knowledge were accepted in the Special Issue, highlighting the main developments and the new findings presented at the conference. Accordingly, 21 papers were accepted and included in this Special Issue.

1. Pilot operation of chemical looping combustion of solid and gaseous fuels

Interesting operational results with coal and ilmenite from the 1 MW_{th} at Technische Universität Darmstadt were reported, [2]. Further, the design and operation of a new 50 kW_{th} chemical-looping combustor for solid fuels in Zaragoza was reported, [3]. Moreover the combustion of coal in a 5 kW CLC using hematite has been studied at Huazhong University, [4]. An iron ore modified with cement/CaO was used in combustion of coal in a 1 kW unit in Nanjing, [5], Vienna University of Technology presented results from operation under varying conditions with four different oxygen carriers in a 120 kW_{th} using natural gas, [6] Two calcium manganite materials were able to fully convert the fuel. Finally, operation of a small 300 W chemical-looping with novel oxygen carrier materials of combined manganese/iron/silicon oxides has been presented, [7].

2. Studies of oxygen carriers

In addition to the pilot studies, which show the performance of the oxygen carriers under varying operating conditions and in different reactor systems, a number papers have studied novel oxygen carrier materials in laboratory. Interestingly, only few of these papers address what was previously the main focus of research, i.e. monometallic oxygen carriers. Instead the focus is on different combined oxide systems. One of the exceptions is thermodynamic/kinetic study of an iron oxide material using a semi-continuous moving bed reactor, [8]. Another study investigates MgO-promoted hematite and a CuO-Fe₂O₃ mixed metal oxide in both fixed bed and fluidized reactor

tests, [9]. Perovskites based on calcium manganites doped with Ba and Sr were studied for CLOU, [10]. Furthermore, natural ilmenite ore used as oxygen carrier in a 100 kW pilot for solid fuels was studied with respect to iron-titanium segregation, [11]. Also, larger granules of ilmenite produced by extrusion for use in the packed-bed CLC process, were studied with respect to thermal and mechanical behaviour [12].

3. Oxygen carriers for hydrogen production

Several oxygen carrier materials were studied for the use in related processes. Thus, combined iron-oxide perovskites were studied for the use in chemical-looping hydrogen production, [13]. Similar materials were studied for the same purpose using perovskites combined with either iron oxide or cobalt oxide, [14]. An iron oxide material supported by alumina was investigated for pressurized hydrogen production using pressures up to 22 bars, [15]. Whereas the previous works addressed hydrogen production from gaseous fuels, autothermal reforming of biomass using a copper-based oxygen carrier was studied in batch fluidized bed experiments, [16].

4. Hydrodynamic studies

The hydrodynamics of fluidized beds and pneumatic valves were studied as effect of temperature and channel size, [17].

5. Design, costing, mathematical modelling and simulation of chemical looping combustion

The modelling of chemical looping combustion plays an important role in the understanding, development and optimization of the entire process. Different ways of desulphurizing the Integrated Gasification Chemical-Looping Combustion (IGCLC) process have been investigated, [18]. A process model for coal chemical-looping combustion, has been validated against test results in a 100 kW_{th} pilot, [19]. Further, a model of a two-stage fuel reactor for Chemical-Looping with Oxygen Uncoupling (CLOU) has been investigated, [20]. A design of a 100 MW_{th} CLC unit for solid fuels has been proposed and investigated with respect to heat and mass balances, hydrodynamics, and performance, [21]. Finally, the design of a 1000 MW_{th} chemical-looping boiler for solid fuels has been investigated in relation to the differences to a circulating fluidized-bed (CFB) boiler of similar size, [22]. The paper provides a detailed and transparent discussion of the costs relating to these differences and indicates that CLC should be able to achieve uniquely low CO₂ capture costs, around 20 €/tonne of CO₂, [22].

6. Conclusion

The papers included in this Special Issue give insights into the some of the latest developments in chemical-looping and as well as idea of the scope and depth in the research. Thus, six papers provide results from pilot operation giving additional evidence of the viability of the technology, another nine papers reflect the development work of oxygen carrier materials, one paper examines hydrodynamics, whereas five papers relate results from design, costing, modelling and simulation of the process. The latter papers clearly highlight the similarity to established CFB technology, and the costing clearly indicates the potential for uniquely low CO₂ capture costs.

We hope the information collected in this Special Issue involving new results in chemical looping technology will benefit the readers of Applied Energy. We also look forward to more submissions to Applied Energy Journal of high-quality, excellence and clarity that stand out and that can make a difference in chemical looping combustion.

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References Yan: Please include all Dols and Volume and issue numbers, and pages....
When they are available.

- [1] Epple, B., Lyngfelt, A., Adanez, J., and Yan, J., (2012), The 2nd International Conference on Chemical Looping, *Applied Energy*, 113, pp. 1827-1829.
- [2] Ströhle, J., Orth, M., and Epple, B., (2015), Chemical Looping Combustion of Hard Coal in a 1 MW_{th} Pilot Plant Using Ilmenite as Oxygen Carrier, Accepted for publication in *Applied Energy: Special Issue*.
- [3] Abad, A., Pérez-Vega, R., De Diego, L., Garcá-Labiano, F., Gayán, P., and Adánez, J., (2015), Design and operation of a 50 kW_{th} Chemical Looping Combustion (CLC) unit for solid fuels, accepted for publication in *Applied Energy: Special Issue*.
- [4] Ma, J., Zhao, H., Tian, X., Wei, Y., Rajendran, S., Zhang, Y., Bhattacharya, S., and Zheng, C., (2015), Chemical looping combustion of coal in a 5 kW_{th} interconnected fluidized bed reactor using hematite as oxygen carrier, accepted for publication in *Applied Energy: Special Issue*.
- [5] Gu, H.M., Shen, L.H., Zhong, Z., Niu, X., Liu, W., Ge, H., Jiang, S., and Wang, L., (2015), Cement/CaO-modified iron ore as oxygen carrier for chemical looping combustion of coal, accepted for publication in *Applied Energy: Special Issue*.
- [6] Mayer, K., Penthor, S., Pröll, T., and Hofbauer, H., (2015), The different demands of oxygen carriers on the reactor system of a CLC plant – Results of oxygen carrier testing in a 120 kW_{th} pilot plant, accepted for publication in *Applied Energy: Special Issue*.
- [7] Källén, M., Rydén, M., Lyngfelt, A., and Mattisson, T., (2015), Chemical-looping combustion using combined iron/manganese/silicon oxygen carriers, accepted for publication in *Applied Energy: Special Issue*.
- [8] Zeng, L., Tong, A., Kathe, M., Bayham, S., and Fan, L.-S., (2015), Iron oxide looping for natural gas conversion in a countercurrent moving bed reactor, accepted for publication in *Applied Energy: Special Issue*.
- [9] Siriwardane, R., Tian, H., Miller, D., and Richards, G., (2015), Fluidized bed testing of commercially prepared MgO-promoted hematite and CuO–Fe₂O₃ mixed metal oxide oxygen carriers for methane and coal chemical looping combustion, accepted for publication in *Applied Energy: Special Issue*.
- [10] Galinsky, N., Mishra, A., Zhang, J., and Li, F., (2015), Ca_{1-x}A_xMnO₃ (A = Sr and Ba) perovskite based oxygen carriers for chemical looping with oxygen uncoupling (CLOU), accepted for publication in *Applied Energy: Special Issue*.
- [11] Knutsson, P. and Linderholm, C., (2015), Characterization of ilmenite used as oxygen carrier in a 100 kW chemical-looping combustor for solid fuels, accepted for publication in *Applied Energy: Special Issue*.
- [12] Jacobs, M., Noyen, J.V., Larring, Y., Mccann, M., Pishahang, M., Amini, S., Ortiz, M., Galluci, F., Sint-Annaland, M.V., Tournigant, D., Louradour, E., and Snijkers, F., (2015), Thermal and mechanical behaviour of oxygen carrier materials for chemical looping combustion in a packed bed reactor, accepted for publication in *Applied Energy: Special Issue*.
- [13] Dueso, C., Thompson, C., and Metcalfe, I., (2015), High-stability, high-capacity oxygen carriers: Iron oxide-perovskite composite materials for hydrogen

- production by chemical looping, accepted for publication in Applied Energy: Special Issue.
- [14] Neal, L., Shafieifarhood, A., and Li, F., (2015), Effect of core and shell compositions on $\text{MeOx}@La_y\text{Sr}_{1-y}\text{FeO}_3$ core-shell redox catalysts for chemical looping reforming of methane, accepted for publication in Applied Energy: Special Issue.
 - [15] Voitic, G., Nestl, S., Lammer, M., Wagner, J., and Hacker, V., (2015), Pressurized hydrogen production by fixed-bed chemical looping, accepted for publication in Applied Energy: Special Issue.
 - [16] Zhao, H., Guo, L., and Zou, X., (2015), Chemical-looping auto-thermal reforming of biomass using Cu-based oxygen carrier, accepted for publication in Applied Energy: Special Issue.
 - [17] Yazdanpanah, M., Forret, A., and Gauthier, T., (2015), Impact of size and temperature on the hydrodynamics of chemical looping combustion, accepted for publication in Applied Energy: Special Issue.
 - [18] Hamers, H.P., Romano, M.C., Spallina, V., Chiesa, P., Gallucci, F., and Annaland, M.v.S., (2015), Boosting the IGCLC process efficiency by optimizing the desulfurization step, accepted for publication in Applied Energy: Special Issue.
 - [19] Ohlemüller, P., Alobaid, F., Gunnarsson, A., Ströhle, J., and Eppe, B., (2015), Development of a process model for coal chemical looping combustion and validation against 100 kW_{th} tests, accepted for publication in Applied Energy: Special Issue.
 - [20] Coppola, A., Solimene, R., Bareschino, P., and Salatino, P., (2015), Mathematical modeling of a two-stage fuel reactor for chemical looping combustion with oxygen uncoupling of solid fuels, accepted for publication in Applied Energy: Special Issue.
 - [21] Abad, A., Adánez, J., Gayán, P., Diego, L.F.D., Garcá-Labiano, F., and Sprachmann, G., (2015), Conceptual design of a 100 MW_{th} CLC unit for solid fuel combustion, Applied Energy - in press.
 - [22] Lyngfelt, A. and Leckner, B., (2015), A 1000 MW_{th} Boiler for Chemical-Looping Combustion of Solid Fuels – Discussion of Design and Costs, Applied Energy - available on line.