Solar Facilities for the European Research Area

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Solar Facilities for the European Research Area

Standardised figures of merit for reactors.

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Executive Summary

In this report we outline figures of merit [1] (or performance indicators) for monitoring and evaluating the performance of solar fuel production reactors. The work was carried out as part of *Task 8.2 Protocols for monitoring and evaluating the performance of solar reactors.* In order to disseminate the protocols to a wider audience, the contents of part A of the report have also been submitted for publication in a special issue of *Frontiers in Energy* titled *Advanced Water Splitting Technologies Development: Best Practices and Protocols* [Bulfin, Miranda & Steinfeld, 2021 *submitted*].

Concentrated solar energy offers a source for renewable high-temperature process heat that can be used to efficiently drive endothermic chemical processes, converting the entire spectrum of solar radiation into chemical energy. In particular, solar-driven thermochemical processes for the production of fuels include reforming of methane and other hydrocarbons, gasification of biomass, coal, and other carbonaceous feedstock, and metal oxide redox cycles. A notable issue in the development of these processes and their associated solar reactors is the lack of consistent reporting methods for experimental demonstrations and modelling studies, which complicates the benchmarking of the technologies. In this report we formulate dimensionless performance indicators based on mass and energy balances of such reacting systems, namely: energy efficiency, conversion extent, selectivity, and yield. These are developed by reviewing the literature (Part B) and by considering standard chemical engineering conventions. Examples are outlined for the processes mentioned above. We then provide guidelines for reporting on such reactors and processes and suggest performance benchmarking on four key criteria: efficiency, conversion extent, selectivity, and performance stability.

[1] The terminology of performance indicators is used in this text as opposed to figures of merit. A figure of merit is usually a unit less parameter that *alone* can be used to benchmark a device, and it is usually a combination of several performance areas. Given the variety of chemical processes covered it is not reasonable to formulate a single figure of merit, so that multiple performance indicators are required.

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Part A: Performance Indicators

Concentrated solar power plants have been established for large-scale renewable power generation in areas with high direct normal irradiance (DNI). These plants convert the entire spectrum of DNI into high-temperature heat, which in turn is used by a heat engine to generate electricity. Alternatively, heat can be used to drive endothermic chemical processes [Romero & Steinfeld, 2012, Yadav & Banerjee 2016], converting solar energy into chemical energy, with the chemical products acting as energy carriers. A promising application in this area is the production of solar fuels. In particular, syngas – a mixture of H_2 and CO – can be produced via a number of routes as illustrated in Figure 1, and further processed to drop-in transportation fuels such as gasoline and kerosene via established gas-to-liquid technologies. Examples of thermochemical processes for solar fuels production include the gasification of biomass, coal, and other carbonaceous feedstock [Nzihou *et al.* 2012, Piatkowski *et al.*, 2012], reforming of hydrocarbons [Agrafiotis *et al.* 2014], and thermochemical redox cycles [Romero & Steinfeld, 2012]. The solar reactor for effecting these processes is the key component and its performance can be the deciding factor in assessing its

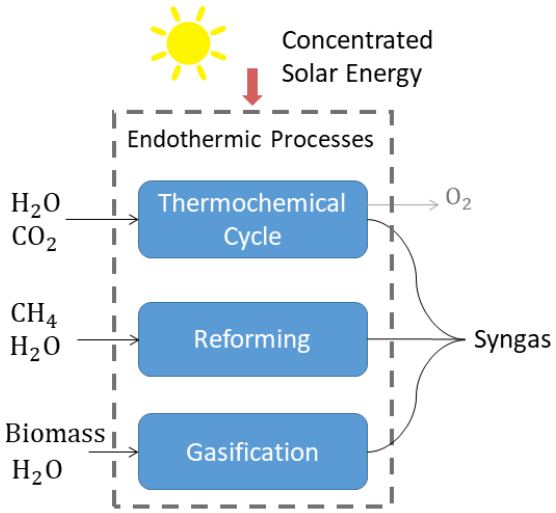

Figure 1. **Main process routes for the solar thermochemical production of syngas – a mixture of H_2 and CO that can be further processed to drop-in transportation fuels.**

technical and economic feasibility. Thus, when reporting on the R&D of such reactors it is beneficial to follow standard conventions and procedures, so that progress can be benchmarked.

A generic solar thermochemical reactor can be defined as a system with both an energy and mass flow input (see Figure 2), with a thermochemical transformation consuming energy and converting chemical species. It is convenient in chemical engineering to define dimensionless parameters to describe the energy and mass balance, which are independent of scale and process and can be used as performance indicators to benchmark the system. These are the energy efficiency for the energy balance, and the conversion extent, selectivity and yield for the mass balance. In addition, we are also interested in the stability of the process, i.e., its performance over time. All of these aspects will affect the capital and operating cost of any scaled-up fuel production process. In an opinion article in *Advanced Science Views,* Ozin highlighted the importance of reporting all these performance indicators to assess the feasibility of renewable fuel production technologies [Ozin 2018], and notes that seldom are all four: conversion, selectivity, efficiency and stability, reported on. When they are reported on, the definitions of these parameters often vary, in particular for the efficiency, but also for standardized chemical process parameters such as selectivity and yield. This article aims to tackle these issues by providing clear protocols and definitions of the dimensionless parameters that can be used as performance indicators for reporting on solar fuel reactors. To do this we propose a standardized efficiency definition and outline the already standardized chemical process parameters of conversion extent, selectivity and yield. Examples for applying them to the solar thermochemical processes shown in Figure 1 are provided.

1. Energy efficiency

Consider a general solar thermochemical reactor as illustrated in Figure 2, which has a feedstock input, a product output, energy inputs in the form of heat and auxiliary work, and waste heat output. We draw a boundary around this system and treat it as our domain. The heat input Q is assumed to be provided by concentrated solar energy, either by direct solar irradiation or by using an indirect method of heat transfer from a solar receiver via heat transfer media. The waste heat can include radiation, conduction, and convection losses, and unrecovered sensible and latent heat in the products and materials of construction. The auxiliary work W_{aux} is the additional work that is required for the operation of the reactor, for example pumping work to overcome pressure drops or to operate at vacuum/high pressures or the energy required to separate the inert gas that is consumed during the process.

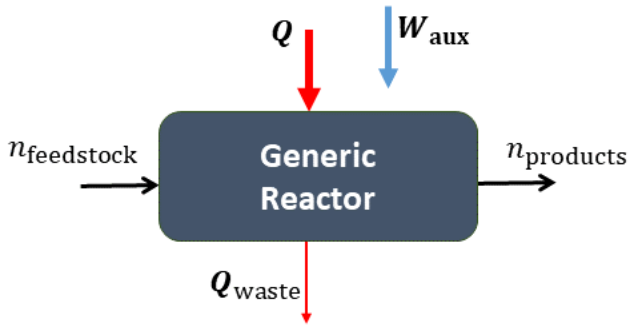

Figure 2. **Generic thermochemical reactor with a feedstock, a product stream, a heat input, waste heat output and some auxiliary work associated with operating the reactor. The heat input is supplied by concentrated solar energy**.

The reactor can perform a continuous or a batch process, or a combination of both. The energy efficiency is defined as the fraction of the energy input that is available as chemical energy in the products. It is expressed as,

$$\eta = 1 - \frac{Q_{\text{waste}}}{E_{\text{total}}} \quad (1)$$

where E_{total} is the total thermal energy supplied to the reactor and Q_{waste} is the heat that leaves the system to the surroundings unused. Evidently, η should always have

a value between 0 and 1. W_{aux} is not necessarily an energy consumption taking place within the reactor (e.g., pumping work for operating at vacuum pressures), however it is work that must be done in order for the reactor to operate. It should therefore be accounted for in E_{total}. Since E_{total} is in the form of heat while W_{aux} is in the form of work, we use instead of W_{aux} the equivalent thermal energy input Q_{aux} to an archetype heat engine with a heat-to-work efficiency,

$$Q_{\text{aux}} = \frac{W_{\text{aux}}}{\eta_{\text{heat-to-work}}}. \tag{2}$$

Typically, $\eta_{\text{heat-to-work}} = 0.4$. The total energy now includes the heat supplied to the reactor and the heat converted to auxiliary work, $E_{\text{total}} = Q + Q_{\text{aux}}$.

Efficiency is rarely defined explicitly as in Eq. (1) because Q_{waste} is not usually a term that can be directly measured. Let us look at some common efficiency definitions seen in the literature and discuss the pros and cons of each. We first consider a definition based around the second law of thermodynamics,

$$\eta = \frac{\sum_i^{\text{products}} \text{n}_\text{i} G_\text{i} - \sum_i^{\text{feedstock}} \text{n}_\text{i} G_\text{i}}{E_{\text{total}}}, \tag{3}$$

where G denotes the Gibbs free energy, i.e., the theoretical maximum work that can be extracted from the chemicals. Thus, the numerator represents the change in Gibbs free energy of the process, which is equivalent to the theoretical maximum *work* that can potentially be performed by the reverse process. This definition was often used in the pioneering work of Fletcher on solar-driven processes [Noring and Fletcher 1982]. Fletcher also derived a theoretical upper bound for this efficiency, given by:

$$\eta_{\text{max}} = \left(1 - \frac{\sigma T_\text{H}^4}{IC}\right)\left(1 - \frac{T_\text{L}}{T_\text{H}}\right), \tag{4}$$

where I is the DNI, C is the solar concentration ratio, T_H and T_L are the temperatures of the upper and lower thermal reservoirs of an equivalent heat engine, and σ is the Stefan-Boltzmann constant [Fletcher and Moen 1977]. This upper bound results from multiplying of the maximum solar absorption efficiency of a perfectly insulated blackbody cavity-receiver (taking into account only radiation losses), and the Carnot efficiency for the maximum conversion of heat to work. However, one issue with equation (3) is that it does not necessarily relate to the fuels energy in a way that is obvious to the reader. The energy available in a fuel is usually quantified in terms of

its heating value, which leads to an alternative efficiency definition, given by:

$$\eta = \frac{\sum_i^{\text{products}} \text{n}_\text{i}\text{HHV}_\text{i} - \sum_i^{\text{feedstock}} n_i \text{HHV}_\text{i}}{E_{\text{total}}} \quad (5)$$

where HHV denotes the higher heating value. Note that lower heating values (LHV) can also be used, as discussed later. The numerator is equivalent to the enthalpy change of the reaction. This efficiency definition has a direct relation to the fuel properties and has been applied in publications on both natural gas reforming and biomass gasification [Jin Jian *et al.* 2018, Z'Graggen & Steinfeld 2008].

It is important to note that Eq. (5) is a *heat-to-heat* efficiency and it can have a larger value than the *heat-to-work* efficiency given by Eq. (3). It is bounded by only the first bracketed term of Eq. (4), which is the maximum absorption efficiency. Thus, while the second law of thermodynamics places a limit on η according to Eq. (3), only the first law of thermodynamics places a limit on η according to Eq. (5).

Finally, the heating value of the feedstock can be included in E_{total} as an additional input of thermal energy. For H_2O and CO_2 splitting cycles, the corresponding heating values are zero. But this is not the case for example for the gasification of biomass, or for the reforming of methane and other hydrocarbons [Piatkowski et al., 2012, Muroyama *et al.* 2018, Müller *et al.* 2017]. The efficiency is then defined as,

$$\eta = \frac{\sum_i^{\text{products}} \text{n}_\text{i}\text{HHV}_\text{i}}{E_{\text{total}}}, \quad (6)$$

where the total energy now includes the solar heat supplied to the reactor, the equivalent heat for auxiliary work, and the heating value in the feedstock, $E_{\text{total}} = Q + Q_{\text{aux}} + \sum_i^{\text{reactants}} \dot{n}_\text{i}\text{HHV}_\text{i}$. Equation (6) is equivalent to equation (5) if the feedstock itself has no heating value (e.g. H_2O or CO_2). This definition is also equivalent to equation (1), in that it tracks the waste heat released in the process as the difference between the denominator and numerator. The heat losses can be radiation, convection, and conduction losses, losses in sensible heat from the products, and waste heat from the generation of W_{aux} (see Eq. (2)). Thus, Eq. (6) can be generally applied to any solar thermochemical fuel production process described in Figure 1.

We should also consider the definitions that other authors have used in the literature, as past work does set some precedent in choosing a definition. A selection of publications with explicitly defined efficiencies are shown in Table 1.

Table 1. **Efficiency definitions in the literature, with the process type, the efficiency equation used, whether they use LHV or HHV, and if they include auxiliary work directly or convert it to heat.**

Reference	**Process**	**Equation***	**LHV/HHV**	W_{aux} /Q_{aux}
Bhosale et. al. 2017	Thermochemical cycle	(5) or (6)	HHV	Q
Bhosale 2019	Thermochemical cycle	(5) or (6)	HHV	Q
Binnoti et. al. 2017	Thermochemical cycle	(5) or (6)	HHV	Q
Bulfin et. al. 2016	Thermochemical cycle	(5) or (6)	HHV	Q
Chuayboon et. al. 2019 July	Gasification	(6)	LHV	-
Chuayboon et. al. 2019	Reforming	other	LHV	-
Falter 2017	Thermochemical cycle	(5) or (6)	HHV	Q
Falter et. al. 2017	Thermochemical cycle	(5) or (6)	HHV	Q
Fletcher & Moen 1977	Thermolysis	(3)	-	-
Gokon et. al. 2014	Gasification	(5)	-	-
Hathaway et. al. 2017	Gasification	(6)	LHV	-
Hathaway et. al. 2016	Thermochemical cycle	(5) or (6)	HHV	Q
Jin et. al. 2015	Reforming	(5)	-	-
Koepf et. al. 2016	Thermochemical cycle	(5) or (6)	-	W
Kong et. al. 2016	Reforming	(5)	HHV	-
Kong et. al. 2018	Thermochemical cycle	(5) or (6)	HHV	Q
Lapp et. al. 2012	Thermochemical cycle	(5) or (6)	HHV	Q
Marxer et. al. 2017	Thermochemical cycle	(5) or (6)	HHV	Q
Müller et. al. 2017	Gasification	(6)	LHV	-
Müller et. al. 2018	Gasification	(6)	LHV	-
Muroyama et. al. 2018	Gasification	(6)	LHV	-

Reference	**Process**	**Equation***	**LHV/HHV**	W_{aux} $/Q_{aux}$
Palumbo et. al. 2015	Reforming/Gasification	(6)	LHV	-
Piatowski et. al. 2011	Gasification	(6)	LHV	-
Yuan et. al. 2015	Thermochemical cycle	(5) or (6)	HHV	W
Z'Graggen et. al. 2006	Gasification	(5)	-	-
Z'Graggen et. al. 2008	Gasification	(5)	-	-
Zheng et. al. 2015	Reforming	(5)	HHV	-
Zhu et. al. 2016	Membrane reactor	other	HHV	-
Zoller et. al. 2019	Thermochemical cycle	(5) or (6)	HHV	Q

* (5) or (6) *indicates that the feedstock has a heating value of zero, so that the definitions are equivalent.*

As can be seen from Table 1, Eq. (6) is the most frequently occurring definition among this sample of literature. It is also the most general formulae, and it can be applied to a reactor or an entire process chain. Consider for example a conventional oil refinery where some of the feed is combusted to provide the heat required for the plant. In this case we cannot apply equation (3) or (5) as they will give a negative efficiency, but equation (6) would be a suitable efficiency definition. Similarly, if we consider a complete solar fuel production process consisting of the endothermic solar gasification of biomass followed by exothermic Fischer-Tropsch synthesis and subsequent refining of the hydrocarbons, the end product will have less heating value than the original biomass. Again, we cannot apply equation (3) or (5) as they would give negative values. Therefore, as the most commonly applied general formula, Eq. (6) is the recommended efficiency definition for future studies.

Other efficiency definitions can be found in the literature, some of which are difficult to interpret and should be avoided. For example, subtracting the auxiliary work term from the numerator rather than including it in the denominator [Zhu et. al. 2016],

$$\eta = \frac{\mathrm{HHV}_{\mathrm{products}} - W}{\mathrm{HHV}_{\mathrm{reactants}} + Q}, \tag{7}$$

as this leads to an equation which can easily have a negative value. Another variation encountered is the inclusion of the conversion extent X in the denominator of the

efficiency definition [Chuayboon *et al.* 2019]

$$\eta = \frac{\mathrm{HHV}_{\mathrm{products}}}{X\ \mathrm{HHV}_{\mathrm{reactants}} + Q + Q_{\mathrm{aux}}}. \quad (8)$$

which mixes up the benchmarks for mass balance with that of the energy balance.

Higher heating value (HHV) vs. lower heating value (LHV) – Some sources use LHV instead of HHV (Table 1). If there is hydrogen or hydrocarbons in the products this will lead to lower efficiency values, and one could argue that HHV is used due to positive bias. HHV does indeed occur more frequently in the literature, and we are also suggesting its use here as it offers a strict upper-bound for the useful energy in the fuel in all utilization cases.

2. Conversion extent, selectivity, and yield

These performance indicators are based on the mass balance and are used to keep track of the chemical reactions taking place, and they are, along with energy efficiency, the most important metrics for assessing the performance of chemical reactors. The conversion extent monitors how much of the feedstock supplied undergoes a chemical change within the reactor, while the selectivity gauges the extent of unwanted side reactions. The yield is the product of conversion and selectivity, and it gives the amount of the desired product formed relative to the stoichiometric maximum product formation, and thus provides information about the purity of the fuel produced. This means that reporting conversion extent and selectivity (or yield) gives the information needed to benchmark the system, while only reporting yield leaves ambiguity about the selectivity and conversion extent. Together, these mass balance parameters have very useful implications for reactor design, including relating the reactor free volume and flow rates to production rates. These metrics can be used to rule out processes as unfeasible for large scale industrial production [Lange 2016]. Although they can be considered the nuts and bolts of chemical engineering research, they are often omitted in solar reactor studies.

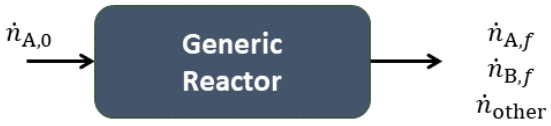

Figure 3. **A continuous-flow thermochemical reactor with a molar feed rate of $\dot{n}_{A,0}$, and a product stream with outflows $\dot{n}_{A,f}$, $\dot{n}_{B,f}$ and $\dot{n}_{other}$.**

Let's first define these indicators for a generic chemical reactor and then give examples for the solar fuel production processes discussed. We follow the definitions formulated in the seminal chemical reactor engineering textbooks by Levenspiel and Scott [Levenspiel 2001, Scott 2006]. The conversion extent is generally formulated in terms

of a limiting reactant. The limiting reactant is the reactant fed to the reactor which can be completely consumed according to the chemical reaction stoichiometry and input flow rates. For a trivial chemical process, such as

$$\mathrm{A} \rightarrow \mathrm{B}, \tag{9}$$

we have only one feedstock so that species *A* is the limiting reactant, and the desired product is *B*. A chemical reactor for this process is illustrated in Figure 3.

The conversion extent for this process can be defined as the relative change in the number of moles of the limiting reactant,

$$X_{\mathrm{A}} = 1 - \frac{\dot{n}_{\mathrm{A},f}}{\dot{n}_{\mathrm{A},0}}. \tag{10}$$

For a batch reactor we can use the same formulae but with the number of initial and final moles, instead of molar flow rates. The *performance equation* of a chemical reactor gives the relation between the free volume, species flow rates and the conversion extent [Levenspiel 2001]. If for example we can only achieve a low conversion extent of say 10 %, and we assume perfect selectivity, then we must have a feedstock flow rate which is 10 times higher than the desired production rate, and a large volume reactor to accommodate the flow, which has obvious implications for the cost and practical feasibility of a process.

As well as the desired reaction there can also be undesired reactions, for example,

$$\mathrm{A} + \mathrm{B} \rightarrow \mathrm{C}, \tag{11}$$

where *A* is the reactant, *B* is the desired product, and *C* is an undesired product. The selectivity of the reactor towards species *B* is defined as the production rate of the desired product *B* relative to consumption rate of the feedstock *A*,

$$S_{\mathrm{B}} = \frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0} - \dot{n}_{\mathrm{A},f}}. \tag{12}$$

If this side reaction does not proceed at all we would have $\dot{n}_{\mathrm{B},f} = \dot{n}_{\mathrm{A},0} - \dot{n}_{\mathrm{A},f}$, and a selectivity of 1. The yield is the amount of desired product formed relative to the maximum amount of desired product that can be formed, i.e., it is the product of conversion extent and selectivity,

$$Y_{\mathrm{B}} = X_{\mathrm{A}} S_{\mathrm{B}} = \frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0}} \ . \tag{13}$$

Note then that reporting conversion extent, selectivity, and feed rates offers a complete description of the chemical transformation in the reactor. However, if the system does not have an ideal selectivity of $S = 1$, it is also recommended to report the yield separately.

Another point worth noting is that the selectivity and yield need to be adjusted by the relative stoichiometry of the product to the limiting reactant. For example, for the reaction $A \rightarrow 2B$, the selectivity would be given by $S_{\mathrm{B}} = \frac{1}{2}\frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0} - \dot{n}_{\mathrm{A},f}}$, where the factor of one half accounts for the 2 moles of *B* formed for every one mole of *A* reacted.

When reporting on the mass balance for reactor demonstrations it is recommend to report,

- The feedstock flow rates in moles/sec (or kg/sec for biomass).
- The conversion extent of the feedstock in terms of the limiting reactant.
- The selectivity towards the desired product, and the yield.

In trivial cases where the selectivity can be assumed to have a value of one, then the conversion extent and yield will be equal. In such cases this should be clearly stated in the results.

3. Stability

The reactor's performance stability can be reported using the same mass and energy balance performance indicators described above, by giving their values as a function of time (or cycle # for cyclic processes). In other words, the efficiency η given by Eq. (6), conversion extent X_i given by Eq. (10), the selectivity S_i given by Eq. (12), and the yield given by Eq. (13), should all be monitored over time to gauge the stability of the performance. Other stability issues such as degradation or complete failure of components are much more difficult to report in a consistent way, as there are no scalable measurements of such faults that can be broadly applied. We therefore restrict our recommendations to recording the performance indicators over time.

4. Examples of the performance indicators

Here we outline the performance indicators for some example processes that have been investigated in the literature, namely solar reforming, two step thermochemical cycles and solar biomass gasification.

Solar methane reforming – Methane can be converted to syngas by reacting it with steam at high temperatures via the endothermic reforming reaction,

$$CH_4 + H_2O \rightarrow CO + 3H_2, \tag{14}$$

The heat for this reaction can be supplied by concentrated solar energy [Agrafiotis *et al.* 2014]. This can be performed with a continuous flow reactor as illustrated Figure 4. There can be numerous side reactions such as the reverse water-gas shift,

$$CO + H_2O \rightarrow CO_2 + H_2. \tag{15}$$

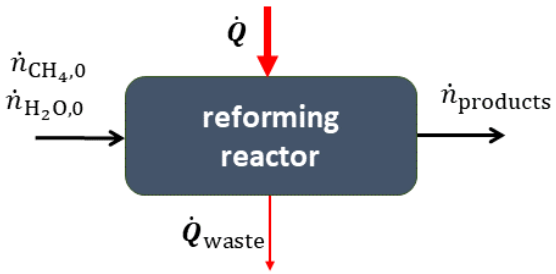

Figure 4. **A thermochemical reactor for the steam reforming of methane. The heat input is supplied by concentrated solar energy.**

For this process, we define the efficiency as (Eq. (6));

$$\eta = \frac{\sum_i^{products} \dot{n}_i HHV_i}{\dot{n}_{CH_4,0} HHV_{CH_4} + \dot{Q}}. \tag{16}$$

where the sum in the numerator is over the species flow rates in the products times their HHVs. To avoid coking, steam is used in excess with $\dot{n}_{H_2O} \approx 2\dot{n}_{CH_4}$ and methane

as the limiting reactant, with conversion extent given by,

$$X_{\mathrm{CH_4}} = 1 - \frac{\dot{n}_{\mathrm{CH_4},f}}{\dot{n}_{\mathrm{CH_4},0}}. \quad (17)$$

CO_2 can also be in the feedstock in mixed and dry reforming processes [Bulfin *et al.* 2021], allowing for more CO production, which can be desirable for gas-to-liquids conversion. An important parameter is then the selectivity towards CO, given by:

$$\mathrm{S_{CO}} = \frac{\dot{n}_{\mathrm{CO},f}}{\dot{n}_{\mathrm{CH_4},0} - \dot{n}_{\mathrm{CH_4},f}} . \quad (18)$$

Solid phase carbon is an unwanted product which can form due to the Boudouard reaction or methane cracking. If present, this can be gauged using the carbon yield [Bulfin *et al.* 2021],

$$\mathrm{Y_C} = \frac{\dot{n}_{\mathrm{C}}}{\dot{n}_{\mathrm{CH_4},0}} , \quad (19)$$

but ideally it should be zero. Note the formulae given can be re-arranged to be in terms of feed rate, and mole fractions in the product stream, as required by the type of measurements taken in the experiment. However, it should be checked that definitions used are equivalent to the standard versions given here.

Thermochemical redox cycles – Two-step metal oxide redox cycles can be used to split H_2O and CO_2, producing H_2 and CO [Romero & Steinfeld, 2012, Bulfin et al. 2017]. A metal oxide first undergoes reduction at high temperature and low oxygen partial pressures,

$$MO_{\mathrm{ox}} \rightarrow MO_{\mathrm{red}} + \frac{\delta}{2}\mathrm{O_2} \quad (20)$$

and is then reacted at lower temperature with H_2O or CO_2 to form H_2 and CO,

$$MO_{\mathrm{red}} + \mathrm{CO_2} \rightarrow MO_{\mathrm{ox}} + \mathrm{CO} \quad (21)\text{a}$$

$$MO_{\mathrm{red}} + \mathrm{H_2O} \rightarrow MO_{\mathrm{ox}} + \mathrm{H_2} \quad (22)\text{b}$$

Both redox reactions can be performed in the same fixed bed reactor but at different times so that the reactor undergoes a cycle [Hathaway *et al.* 2016, Haeussler *et al.* 2020, Marxer *et al.* 2017]. Alternatively, it could be operated continuously using a

particle transport reactor [Ermanoski *et al.* 2013, Singh *et al.* 2017, Welte *et al.*2016]. The process is illustrated in Figure 5, where it is important to note that reduction and oxidation are either taking place at different times, or in separate reaction chambers.

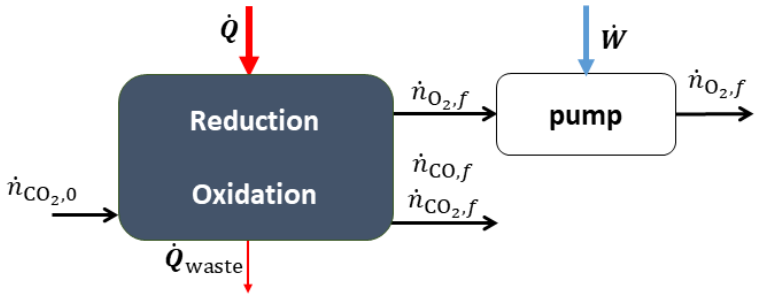

Figure 5. **A thermochemical reactor for the redox splitting of CO_2. The heat input is supplied by concentrated solar energy.**

In the fixed bed case, the system parameters; temperature, pressure, input power, conversion extent, auxiliary work *etc.*, are varying in time. We therefore need to take integrals over an entire cycle to get the desired performance parameters. For CO_2-splitting, we define the efficiency as,

$$\eta = \frac{\mathrm{HHV_{CO}} \int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO}}(t)\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{Q}(\mathrm{t}) + \dot{Q}_{\mathrm{aux}}(t)\;\mathrm{d}t}. \tag{23}$$

Auxiliary work may include vacuum pumping during reduction and the production of sweep inert gas. This definition is the same for water splitting, where CO is replaced by H_2. In this case we only have one feedstock, so that the conversion extent is given by:

$$X_{\mathrm{CO_2}} = 1 - \frac{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},f}\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},0}\,\mathrm{d}t}. \tag{24}$$

The selectivity towards CO is given by:

$$\mathrm{S_{CO}} = \frac{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO},f}\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},0} - \dot{n}_{\mathrm{CO_2},f}\,\mathrm{d}t}. \tag{25}$$

The yield is given by $Y_{CO} = X_{CO_2} S_{CO}$. Alternative formulae which are equivalent to these definitions are possible.

The literature on thermochemical redox cycles is perhaps the most problematic in terms of reporting standards due to the more complex nature of the cyclic process. There are few articles in the literature which address all the performance indicators described here, with the work of Marxer *et al.* a notable exception [Marxer *et al.* 2017]. A few studies report the yield as the moles of H_2 or CO produced per gram of the cycled redox material [Agrafiotis *et al.* 2005, Hathaway *et al.* 2016, Haeussler *et al.* 2020], instead of the dimensionless parameter defined here. This amount of fuel produced relative to the redox material is indeed important, and it provides additional information for scaling up the process, but it should not be referred to as the yield. Of primary importance in the mass balance is to report the conversion extent of the oxidant feedstock and the selectivity to the desired product as defined here.

An interesting example that emphasizes the importance of these performance indicators is that of isothermal redox cycles. In this case both redox steps are conducted at the same temperature by performing a pressure swing. Studies have focused on the energy efficiency [Ermanoski et al. 2014] and experimental demonstrations of the process [Hathaway et al. 2016, Hoskins et al. 2019, Muhich et al. 2013]. The conversion extent in the demonstrations is typically not reported, but it is known to be thermodynamically limited to low values on the order of $X \approx 0.01$, at the operating conditions considered [Bulfin et al. 2016, Ermanoski et al. 2014]. A low conversion extent has practical implications on the scale-up because of the large mass flow rates of H_2O or CO_2 required per mol of redox material. This, in turn, leads to larger reactors and increased capital cost. Classical chemical reactor engineering texts put a large emphasis on the conversion extent and yield [Levenspiel 2001], which highlights the importance of using both the energy and mass balance to benchmark these systems.

Solar Biomass Gasification – Biomass, coal, or carbonaceous materials can be reacted with steam at high temperatures to produce syngas, as illustrated in Figure 6 [Piatkowski et al., 2012]. Since biomass does not have a simple chemical composition, the mass balance is more complex than the previous examples. Proximate and ultimate analysis are required to determine the chemical breakdown of the biomass [Müller *et al.* 2018, Muroyama *et al.* 2018].

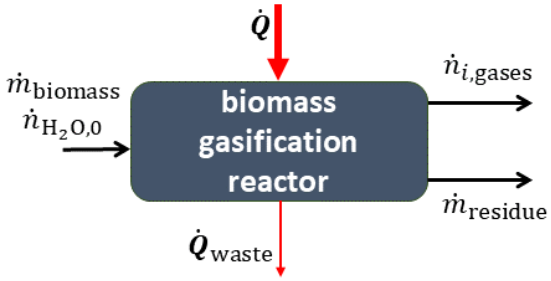

Figure 6. **A reactor for the gasification of biomass. The high-temperature heat input is supplied by concentrated solar energy.**

Biomass gasification is a combination of many independent reactions with the net reaction summarised by the formula,

$$\mathrm{C_nH_mO_k} + \mathrm{yH_2O} \rightarrow x_1\mathrm{CO} + x_2\mathrm{CO_2} + x_3\mathrm{H_2} + x_4\mathrm{CH_4} + \text{other gases} + \text{residue}, \quad (26)$$

where $\mathrm{C_nH_mO_k}$ represents biomass. The biomass geneally also contains sulphur and nitrogen impurities on the order of 1 % by mass. The efficiency can be expressed as

$$\eta = \frac{\sum_i^{\text{gas-products}} \dot{m}_\mathrm{i} \mathrm{HHV_i}}{\dot{m}_{\text{biomass}}\, \mathrm{HHV_{biomass}} + \dot{Q}}. \quad (27)$$

where we only count the heating value of the gasified products, as the residue is an ash like by-product and not a fuel. Note that the HHV are per unit mass and not per mole as in other formulae, which is due to the fact that the feed does not have a well-defined stoichiometric chemical formula. In this case an upgrade factor is also often reported, which is the change in the heating value of the products relative to the feedstock.

$$U = \frac{\sum_i^{\text{gas-products}} \dot{m}_\mathrm{i} \mathrm{HHV_i}}{\dot{m}_{\text{biomass}}\, \mathrm{HHV_{biomass}}}. \quad (28)$$

The value of U depends on the type of feedstock and the syngas yield. This value can offer non-redundant information on the energy balance when reported together with the efficiency.

For the conversion extent, the species which is not fully converted by the gasification is carbon, and so the conversion extent is usually defined in terms of the carbon conversion extent [Müller *et al.* 2018]

$$X_{\mathrm{C}} = 1 - \frac{\dot{m}_{\mathrm{C-residue}}}{\dot{m}_{\mathrm{C},0}}. \quad (29)$$

where $\dot{m}_{\mathrm{C},0}$ is the mass flow rate of carbon in the biomass feed (determined by ultimate analysis), and $\dot{m}_{\mathrm{C-residue}}$ is the mass of carbon in the residue (the unreacted carbon), which can be made up of tar and ash. The carbon to syngas yield can be expressed using molar flow rates of carbon containing species in the gas stream,

$$Y_{\mathrm{syngas}} = \frac{\sum_i^{\mathrm{gases}} \nu_{i,C} \dot{n}_{\mathrm{i,gas}}}{\dot{n}_{\mathrm{C},0}} = \frac{\dot{n}_{\mathrm{CO}} + \dot{n}_{\mathrm{CO_2}} + \dot{n}_{\mathrm{CH_4}} + 2\dot{n}_{\mathrm{C_2H_6}} + \cdots}{\dot{n}_{\mathrm{C},0}}. \quad (30)$$

where $\nu_{i,C}$ is the stoichiometric number of carbons in the gas species. Often this sum is only performed for CO, CO_2, and CH_4, which does offer a good approximation especially at high temperatures where other carbon containing gases are in very low concentration. Given the large number of reactions present there are a number of different selectivities that could be discussed. For downstream gas-to-liquid processes, CO may be favoured over CO_2 and CH_4, in which case the selectivity towards CO can be defined as,

$$S_{\mathrm{CO}} = \frac{\dot{n}_{\mathrm{CO}}}{\sum_i^{\mathrm{gases}} \nu_{i,C} \dot{n}_{\mathrm{i,gas}}} = \frac{\dot{n}_{\mathrm{CO}}}{\dot{n}_{\mathrm{CO}} + \dot{n}_{\mathrm{CO_2}} + \dot{n}_{\mathrm{CH_4}}}. \quad (31)$$

5. Summary of reporting protocols

The dimensioned parameters required to describe the reactor system are:

1. The reactor volume and free volume.
2. Mass loading of any catalyst or cycled redox material.
3. The operating conditions of the reactor (e.g., temperature, pressure, etc.).
4. The molar/mass flow rates of feedstock into the reactor.
5. The total heat supply to the reactor, Q (e.g., solar heat, etc.)
6. Auxiliary work demands, Waux, (e.g., pumping work, inert gas production, etc.)

This information should be sufficient for the study to be reproduced or modelled by other laboratories. The energy and mass balance performance indicators outlined in the previous sections allows for the system to be benchmarked. These are:

1. The energy efficiency $\eta = \frac{\mathrm{HHV_{products}}}{\mathrm{HHV_{reactants}}+Q+Q_{\mathrm{aux}}}$. For solar-upgrading processes such as gasification and reforming, report additionally the upgrade factor $U = \frac{\mathrm{HHV_{products}}}{\mathrm{HHV_{feedstock}}}$.
2. The conversion extent of the feedstock, $X_{\mathrm{A}} = 1 - \frac{\dot{n}_{\mathrm{A},f}}{\dot{n}_{\mathrm{A},0}}$.
3. The selectivity towards the desired product $S_{\mathrm{B}} = \frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0}-\dot{n}_{\mathrm{A},f}}$, and the yield of the desired product $Y_{\mathrm{B}} = \frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0}}$ if the selectivity is not reported.
4. Performance stability, i.e., report the above indicators over time during a test campaign.

Conversion extent, selectivity, and energy efficiency, combined with mass flow rates offer a complete description of the reactor performance, and the performance over time can be used to gauge stability. For cyclic processes, the benchmarks should use integrals over an entire cycle as outlined here. Similarly, for continuous processes,

time averaged parameters will be better indicators of performance.

Table 2 **The performance indicators for the example solar fuel processes discussed in this work.**

Reactor type	**Efficiency** η	**Conversion extent** X_i	**Selectivity (or Yield)** S **(or** Y**)**
Thermochemical redox CO_2 splitting	$\frac{\mathrm{HHV_{CO}} \int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO}}(t)\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{Q}(\mathrm{t}) + \dot{Q}_{\mathrm{aux}}(t)\ \mathrm{d}t}$	$1 - \frac{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},f}\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},0}\,\mathrm{d}t}$	$\frac{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO},f}\,\mathrm{d}t}{\int_0^{t_{\mathrm{cycle}}} \dot{n}_{\mathrm{CO_2},0} - \dot{n}_{\mathrm{CO_2},f}\,\mathrm{d}t}$
Solar methane reforming	$\frac{\sum_i^{\mathrm{products}} \dot{n}_{\mathrm{i}}\mathrm{HHV_i}}{\dot{n}_{\mathrm{CH_4},0}\mathrm{HHV_{CH_4}} + \dot{Q}}$	$1 - \frac{\dot{n}_{\mathrm{CH_4},f}}{\dot{n}_{\mathrm{CH_4},0}}$	$\frac{\dot{n}_{\mathrm{CO},f}}{\dot{n}_{\mathrm{CH_4},0} - \dot{n}_{\mathrm{CH_4},f}}$
Biomass gasification[2]	$\frac{\sum_i^{\mathrm{gas-products}} \dot{m}_{\mathrm{i}}\mathrm{HHV_i}}{\dot{m}_{\mathrm{biomass}}\,\mathrm{HHV_{biomass}} + \dot{Q}}$	$1 - \frac{\dot{m}_{\mathrm{C-residue}}}{\dot{m}_{\mathrm{C},0}}$	$(Y_{\mathrm{syngas}} = \frac{\sum_i^{\mathrm{gases}} \nu_{i,C}\dot{n}_{\mathrm{i,gas}}}{\dot{n}_{\mathrm{C},0}})$
Generic $\mathbf{A} \rightarrow \mathbf{B}$	$\frac{\dot{n}_{\mathrm{B}}\mathrm{HHV_B}}{\dot{n}_{\mathrm{A}}\mathrm{HHV_A} + \dot{Q} + \dot{Q}_{\mathrm{aux}}}$	$1 - \frac{\dot{n}_{A,f}}{\dot{n}_{\mathrm{A},0}}$	$\frac{\dot{n}_{\mathrm{B},f}}{\dot{n}_{\mathrm{A},0} - \dot{n}_{\mathrm{A},f}}$

[2] As there are a large number of possible byproducts and the exact desired composition of the syngas is not always clear, it is common to simply report syngas yield together with conversion extent, instead of selectivity in this case.

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Part B: Literature Review

This literature review was performed as part of Task 8.2, with the goal of guiding the definition of figures of merit for monitoring solar fuel production reactors. The development and application of techniques for the performance evaluation of solar reactors will be established according to the following metrics:

- Fuel quality and process selectivity (fuel composition and purity);
- Long-term cyclic stability for materials and structures;
- Specific fuel output (mass conversion);
- Solar-to-fuel energy efficiency (energy conversion).

The information considered on solar thermochemical processes included the use of different solar technologies and reactors apparatus (such as outdoor and indoor research and demonstration facilities) and material sources (different types of feedstock materials). Concentrated solar technology features high potential to convert solar energy into both thermal and chemical energy with wide-ranging applications.

For that purpose, more than 200 publications were thoroughly analysed on the subject and the data was arranged according to both type of data and parameters (Figure 7) considering the following main research areas:

i) Gasification;

ii) Pyrolysis; and

iii) Redox reactions.

For each research area, the information was then structured taking into account the subgroup of experimental parameters - reaction temperature, pressure, reaction time, solar concentration and number of cycles performed. In the same way, for each research area, a subgroup was considered for the type of work, defined as:

- *Experimental Work (E)*, which correspond to publications reporting work based only in the evaluation of the experimental conditions using either concentrating direct or undirect solar radiation;
- *Modelling (M)*, which correspond to publications reporting work based on the evaluation of the experimental conditions and also developing theoretical models that describe, among others, the effect of experimental conditions, reactor design and/or performance, reaction kinetics, energy or mass transfer conditions and

main products formation;

- *Solar Simulator (SS)*, which correspond to publications reporting work based on the evaluation of the experimental conditions in indoor solar facilities using concentrated light lamps suitable to simulate solar radiation;
- *Modelling and Solar Simulator (M+SS)*, which correspond to publications reporting work based on the evaluation of the experimental conditions, that report or developed theoretical models that describe, for instance, the experimental conditions, reactor design and/or performance, reaction kinetics, energy or mass transfer conditions and main products formation (among others), in indoor solar facilities using concentrated light lamps suitable to simulate solar radiation.

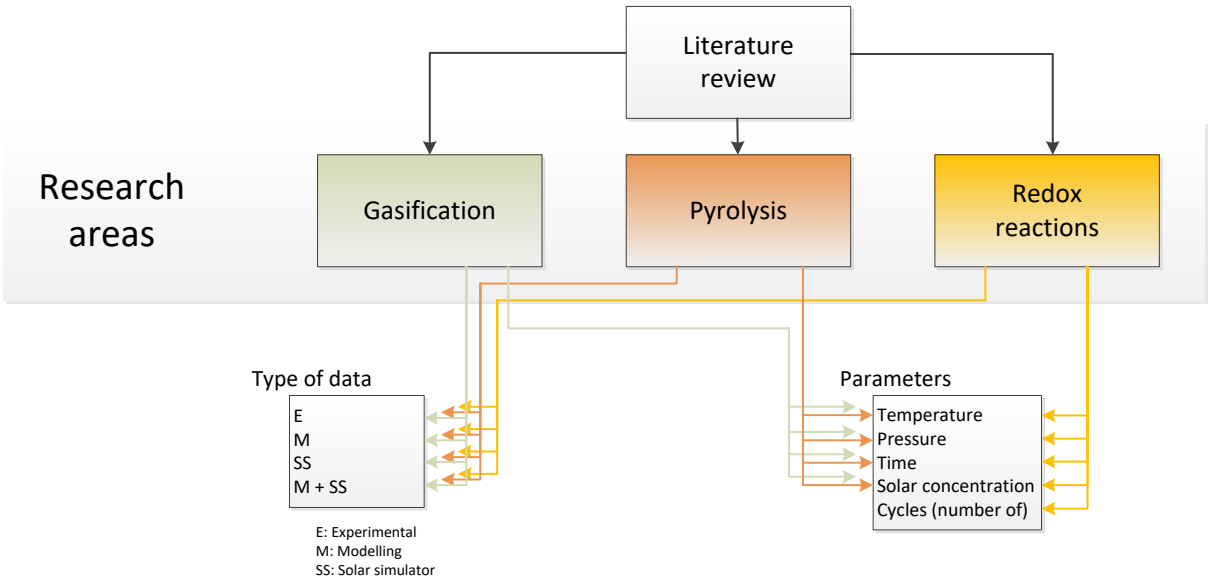


Figure 7. **Literature review conceptual framework on gasification, pyrolysis and redox reactions based on solar concentration energy.**

In "Parameters" subgroup, and also for statistical purposes, it was considered that all information reported in the publications regarding the experimental parameters' temperature, pressure, time, concentration and number of cycles can either refer to a specific value, or to a range of values. This specification was assigned as Total of Entries.

1. Introduction

Fuel production using concentrated solar energy has a significant potential to reduce fossil fuel dependence in current energy economy. The use of solar power to drive thermal or thermochemical processes with different temperature requirements (i.e., medium- to high-temperature) are some of solar power major advantages. Moreover, given the simultaneous demand of carbon-neutral energy, and gaseous and liquid hydrocarbon fuels, the implementation of solar-driven fuel production systems throughout different technologies is likely to expand significantly in the next years.

When considering a research perspective, the first step towards the production of solar fuels using thermochemical reactors usually includes the demonstration in a laboratory environment to better understand a number of critical issues. Among these critical issues are process dynamics and heat and mass transfer limitations associated with reactor design. The performance of these technologies has been under study using high flux solar simulators (HFSS). The HFSS provides an artificial source of concentrated solar energy matching the solar spectral distribution using xenon or argon lamps aligned to the focal point of a highly reflective ellipsoid reflector. This arrangement allows the concentration of radiant energy on the target. Currently, there are a great number of HFSS systems worldwide at well-equipped research centres, available to the research community (with different technologies, configurations and apparatus) and capable of performing several initial studies (Deepak Yadav and Rangan Banerjee, 2016).

The following step usually consists in the use of a solar furnace (SF) with conventional solar input (live sun radiation). This arrangement also features a number of different types of solar furnaces worldwide. A typical solar furnace generally consists of one or more heliostat mirror that reflects the incident solar energy towards a concentrator system, which redirects it to a focal point (Figure 7). One interesting feature is that most of the solar furnaces used are on-axis arrangement for testing solar reactors. For on-axis solar furnaces, the focal point is located between the heliostat arrangement and the concentrator. This arrangement is currently seen as an advantage because it allows the production of a symmetrical beam distribution away from the focal point and the option of attenuating solar radiation by the use of louver shutter (Kuo Zeng$^{\Phi}$ et al., 2017). However, placing the experimental apparatus between the heliostat and

the concentrator system, some of the incoming solar energy is blocked making this configuration a major drawback in its use. Even so, it may be noted that a significant number of solar furnaces featuring this configuration are used for solar reactors testing (E. Koepf et al., 2017).

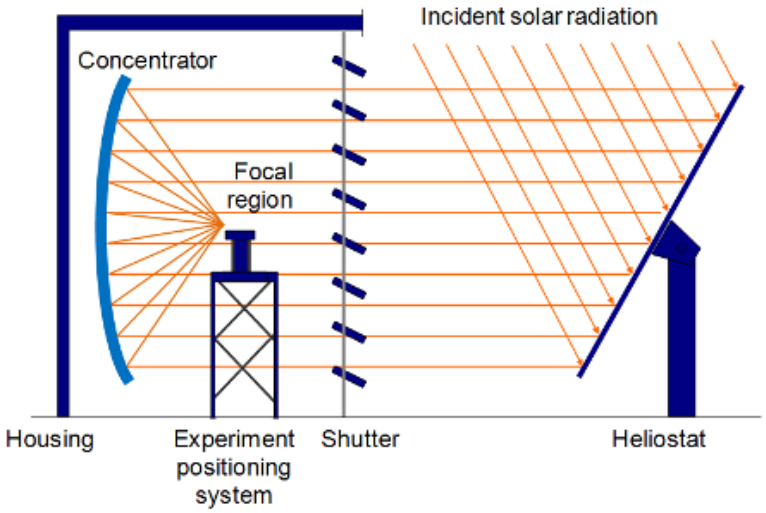


Figure 8. **Horizontal axis solar furnace.**

The solar furnaces used for pilot-scale demonstration present a solar input range from few to hundreds of kW at reactor aperture. For large-scale operations, such as solar thermochemical commercial scale plants and for high concentration, different arrays of heliostat mirrors can be found. The use of a secondary concentrator is normally justified when solar thermochemical processes require operating temperatures above 1,000°C (Deepak Yadav and Rangan Banerjee, 2016).

2. Solar reactors thermochemical processes

On a broad basis, solar thermochemical processes can be classified according to different purposes towards the production of solar fuels and industrial commodities. Considering the range of feedstocks or end-use materials with potential to be used in these processes, reactor configuration significantly impacts on the physical nature of the fuel (i.e., hydrogen, syngas, and other hydrocarbon liquids and solids).

Even with the existing technologies and processes configuration, major oil and gas companies already recognize that it is possible to include synthetic fuels produced in solar reactors (i.e., using hydrogen as a key constituent in the process) as a natural extension of their standard business model. Indeed, synthetic liquid fuels produced from solar-produced synthetic gas through thermochemical processes are considered a promising pathway as a renewable and sustainable energy vector.

Regardless of how to benchmark and evaluate the solar-driven fuel production or the materials processing systems, the design of solar reactors is a key factor that significantly impacts on type and nature of fuel characteristics (both physical and chemical). In general, reactors can be classified as follow (Ronald W. Missen et al., 1998):

a) Batch reactor is a discontinuous reactor in which the operation is inherently unsteady and usually characterized by a cycle of operation (sufficiently to handle a complete set of reactions). This type of reactor is typically composed by a stirred tank that is filled with reactants before the reaction starts and emptied after the operation is finished (or to the extent that is needed). The reaction may be single-phase or multi-phase. A semi-batch reactor is a variation of a batch reactor in which one reactant may be added intermittently or continuously to another reactant contained in a vessel as reaction proceeds (i.e., a fluid can be continuously fed to a biomass feedstock that was previously placed inside the reactor. In order to ensure operation pressure and other parameters, a product or a flue gas must also be extracted simultaneously).

b) Continuous reactor (flow reactors) is a reactor featuring a continuous carry material (flowing stream) in which reactants are continuously fed into the reactor and emerge as continuous stream of product. Depending on the purpose of the studies, these

reactors are also suitable for testing cycle operations.

Although the previous classification is generally used for generic framework, reactors could also be assigned according to fuel production physical nature, as specific processes may lead to both different reactors design. An example that reflects well these differences is related to directly irradiated reactors suitable for dealing with carbonaceous materials and undirected solar irradiated reactors that need a heat transfer fluid to properly distribute the temperature in the reactor. Moreover, when considering different thermochemical processes (i.e., redox reactions, gasification or pyrolysis), the production of gaseous, liquids or even solids compounds may differ greatly from their physical and chemical nature as distinct phenomena occurs within the reactor medium. In fact, experimental conditions and heat and mass transfer conditions within the reactor are critical in the design of solar reactors (E. Koepf et al., 2017). The design and further testing are critical requirements for a scale-up concept as a number of issues must be considered. From those, most important are the operation demand, use of robust construction materials capable to withstand severe operation conditions (i.e., corrosion which could impact on the operation conditions and ultimately the quality of the final product), designed to have both low thermal inertia and high resistance to thermal shock.

2.1. Gasification

In the thermochemical gasification process much R&D have been done over the years to convert biomass to syngas or even to adapt coal gasification technology suitable for dealing with different biomass sources. In general, different types of gasifiers can be found featuring different characteristics. Those characteristics range from fixed gasifiers (counter-flow and concurrent-flow) to both stationary and circulating fluidized bed and entrained-flow. Gasification is an endothermic process by nature, which means that it is necessary to provide heat to sustain the gasification reactions. This process offers the possibility to transfer the chemical energy contained in a solid-fuel into a gaseous energy carrier.

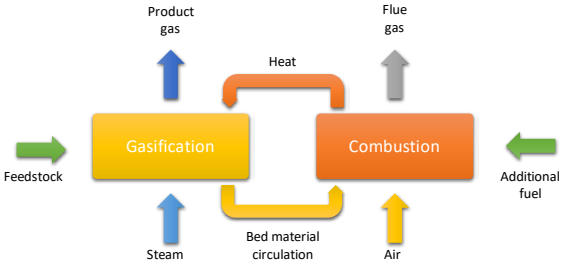
Gasification typically refers to the conversion in an oxygen- or air-deficient environment to produce fuel gases, which are mainly carbon monoxide, hydrogen, methane, and lighter hydrocarbons, but depending on the process used, the products

can contain significant amounts of carbon dioxide and nitrogen, the latter mostly from air. Gasification processes also produce liquids (tars, oils, and other condensates) and solids (char and ash) from solid feedstock. Among the parameters that can influence the production of hydrogen are feedstock composition, gasifier temperature and pressure, moisture content of feedstock, amount of oxidant present, gasifier geometry, and mode of gas-solid contact (Ashok Pandey et al., 2019).

Entrained flow gasifiers (top- and side-fed) are highly efficient, useful for large-scale gasification and suitable for feedstock such as coal, biomass, and refinery residues. For this type of gasifiers, the requirement for highly pulverized fuel particles may be of concern when gasifying biomass is intended. However, at temperatures above 1000°C tar cracking is improved thus becoming suitable to deal with biomass feedstock where tar is a serious issue to consider. Different groups of researchers (Vineet Singh Sikarwar et al., 2016; Binlin Dou et al., 2019) recently published detailed information on reactor configuration of most common gasifiers, output of products, conventional and emerging approaches, challenges and gasification techniques as well.

The process could be either autothermal or allothermal, depending on how the heat is provided. Autothermal gasifiers provide the necessary heat to the reactions through partial oxidation within the gasification reactor. The autothermal gasification process is an inexpensive and sustainable process, which are advantages and the main disadvantage of this process is the lack of inside temperature control, which can be overcome by adding an excess of one of the component or using a diluent (Ashok Pandey et al., 2019). If air is used as oxidizing agent, the syngas contains high amount of nitrogen, which can be a shortcoming to the process due to its high dilution. In order to overcome this drawback, the syngas production could be attained by using an oxidizing agent such as pure oxygen or mixtures of oxygen and steam. One major advantage of autothermal gasification is the direct heating of the reactants, which results in an increased efficient energy utilization while the allothermal is characterized by the separation of the processes of heat production from the heat required for the reactions to occur in the reaction medium (consumption). In the autothermal process, roughly 35% of the feedstock is burned aiming at supplying the activation energy required for the gasification reactions, thus the product gas using autothermal gasification has a low-Lower Heating Value (LHV) of 4-5 MJ/m^3 (Ashok Pandey et al., 2019).

The allothermal gasification process depends on an external source of energy to provide the activation energy for the endothermal reactions. Thus, in allothermal heating, the heat is generated outside the reactor then transferred into the system. Usually, allothermal gasifiers are heated by heat exchangers (Figure 9



) or circulated hot bed material is used to heat between the combustion and gasification zone (Figure 10). A regular allothermal process consists of two reactors (connected by energy flows) in which biomass is gasified in the first reactor (for syngas production) and the remaining solid residue (or even partial syngas) is combusted in the second reactor to produce the heat for the first process. In this process, two gas streams are produced: i) medium calorific syngas resulting from the gasification reactor and ii) flue gas from the combustion reactor and all carbon containing streams from the gas cleaning can be recycled in the combustion zone (converted into heat for further use in gasification reactions). The heat transport is achieved either by circulating the bed material or by heat exchangers (Reinhard Rauch et al., 2013).

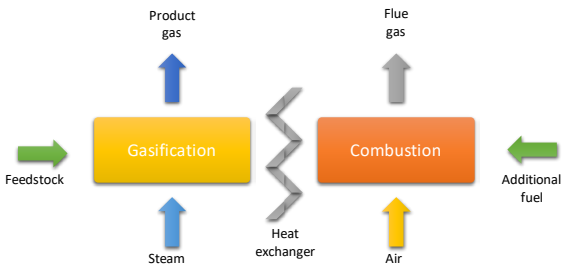


Figure 9. **Allothermal process only with heat exchange.**

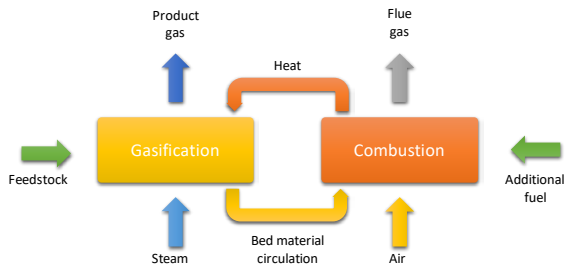


Figure 10. **Allothermal process with mass and heat exchange.**

Typically, an allothermal gasification system produces medium-LHV syngas, with a higher H_2 to CO ratio, with near 15 MJ/m^3, unlike the autothermal gasification

process, using air as the oxidizing agent that produces lower-LHV, as stated above. In the allothermal system there is no need for the incomplete combustion or partial oxidation (volatile products and some of the char reacts with O_2 to form CO_2 and CO) step to take place because the heat required to volatilize the organic (biomass) material is added indirectly. As a result, most of the biomass reacts with CO_2 and water vapour to produce CO and H_2 in the gasification/steam reforming reactions. Afterwards, the water-gas-shift reaction reaches equilibrium in the gasifier, the total resulting H_2 concentration from the allothermal gasifier is typically greater (Ashok Pandey et al., 2019).

Solar-driven gasification is a particular specification of an allothermal gasification process (Figure 11). However, there are additional advantages of considering a solar-driven process, namely:

i) Deliver high syngas output per unit of feedstock as no feedstock amount is combusted for supply the heat gasification requirements for the reactions;
ii) Avoid syngas contamination resulting from combustion by-products which impacts downstream cost reductions (gas cleaning and separations requirements);
iii) To produce high calorific value syngas with lower CO_2 (solar-produced syngas has about two times higher calorific value per feedstock unit when compared with conventional autothermal gasification);
iv) Increase gasification temperatures (exceeding 1100°C) promoting faster reaction kinetics and both higher quality syngas production and low tar content; and
v) More suitable for processing a great number of carbonaceous materials (Christian Wieckert et al., 2013; Alan W. Weimer, 2012).

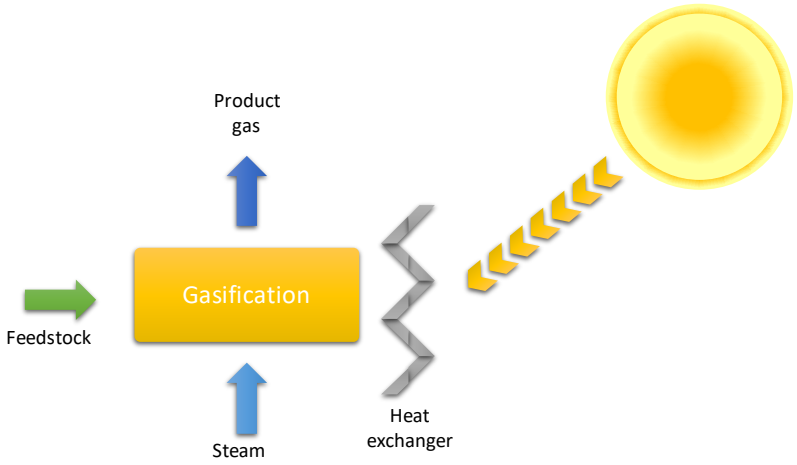


Figure 11. **Solar-driven allothermal gasification process.**

Considering the conceptual framework presented in the Figure 8, the literature review on the main topic of gasification and for the parameters of temperature, pressure, time and solar concentration is presented in Figure 12. This figure depicts the total results for gasification considering the type of work classifications previously assigned as being: a) distribution for both publications and entries and b) entries' year footprint. This analysis reveals 119 total publications (7 publications/year average), of which 57.1% were assigned to type of work E, 22.7% to M, 15.1% to SS and 5.0% to M+SS, ranging mostly between the year of 2003 and 2019. These results also reveal that experimental work has been carried out over the years although an increase in publications were only found between 2013 and 2017. Previously and between the years of 2008 and 2011, only publications on experimental work associated with modelling were performed by the researchers. Studies on solar simulator conditions have been considered occasionally over the years. Publications that report information on the M+SS type of work were only found in the years of 2005, and 2018 to 2019. For this topic, a total of 172 entries were considered.

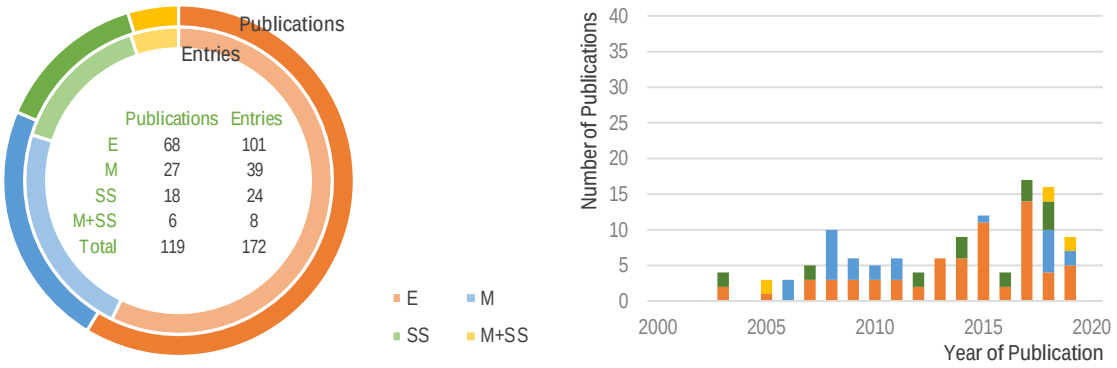


Figure 12. **Publications on gasification grouped according to type of work.**

Figure 13 to Figure 16 depict the data for gasification considering sequentially several reference conditions, such as temperature, pressure, time and solar concentration, and the type of work classifications previously assigned; a) distribution for both publications and entries; and b) entries' year footprint.

The results found for the temperature parameter (Figure 13) reveals 51 total publications being 58.8% assigned to the type of work E, 21.6% to the M, 13.7% to the SS and only 5.9% to the M+SS. For this analysis, 86 total entries were considered most of them assigned to the experimental work (53 entries).

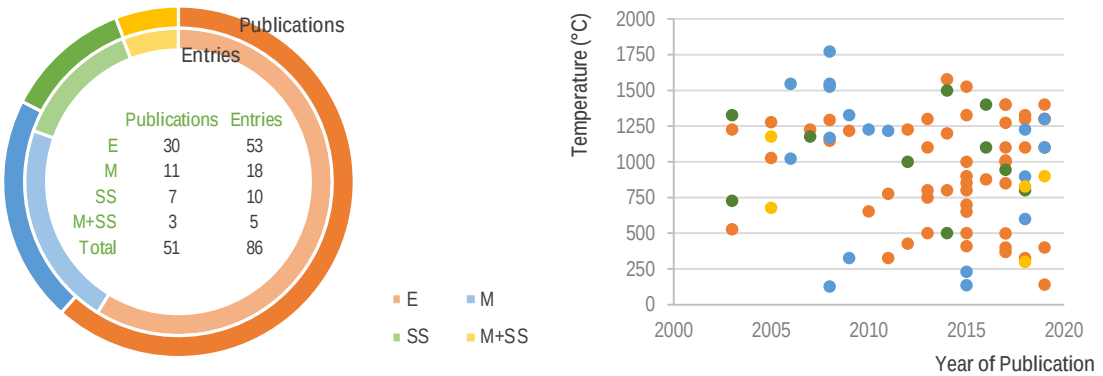


Figure 13. **Publications on gasification grouped according to type of work for the Temperature parameter.**

In Figure 14 is presented the results obtained for the pressure parameter. For this case, less publications were found (16 in total) of which 68.8% were assigned to type of work E. The remaining publications representing 12.5% and 18.8% were respectively assigned to the type of works M and SS. No publications were found for the M+SS type of work. The analysis of the data resulted in 19 entries.

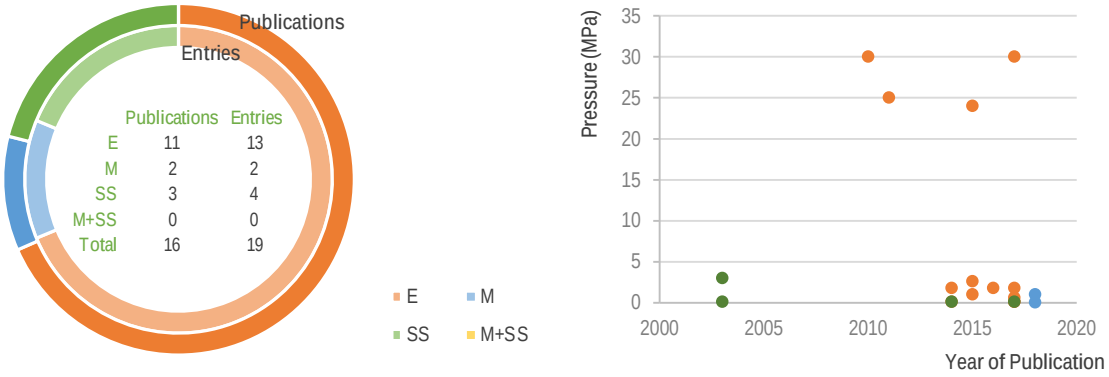


Figure 14. **Publications on gasification grouped according to type of work for the Pressure parameter.**

When considering the time parameter (Figure 15), 33 publications were found between 2005 and 2019. From those 45.5% were assigned to the type of work E, 24.2% to the M, 21.2% to the SS and 9.1% to the M+SS. The information found in the publications resulted in 42 total entries. Regarding the solar concentration parameter, all information is presented in the Figure 16. In this case results reveals 19 total publications being 63.2% assigned to the type of work E, 31.6% to the M and 5.3% to the SS. No publications were found for the M+SS type of work and 25 entries were considered in this analysis.

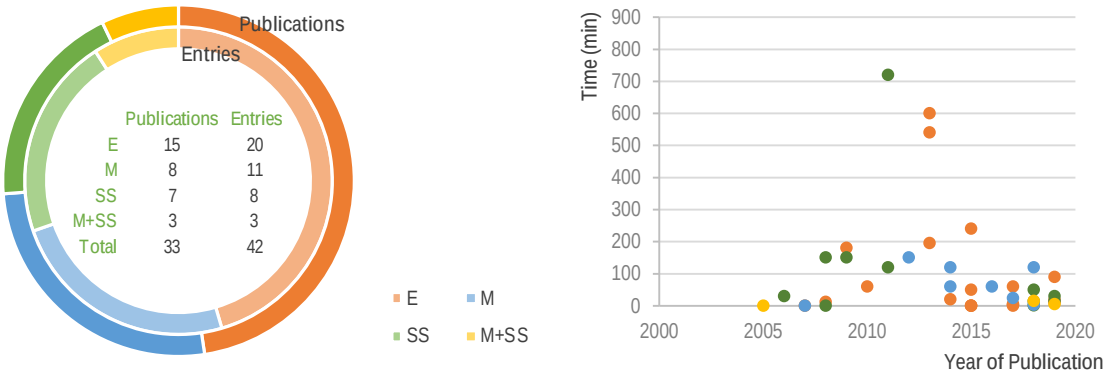


Figure 15. **Publications on gasification grouped according to type of work for the Time parameter.**

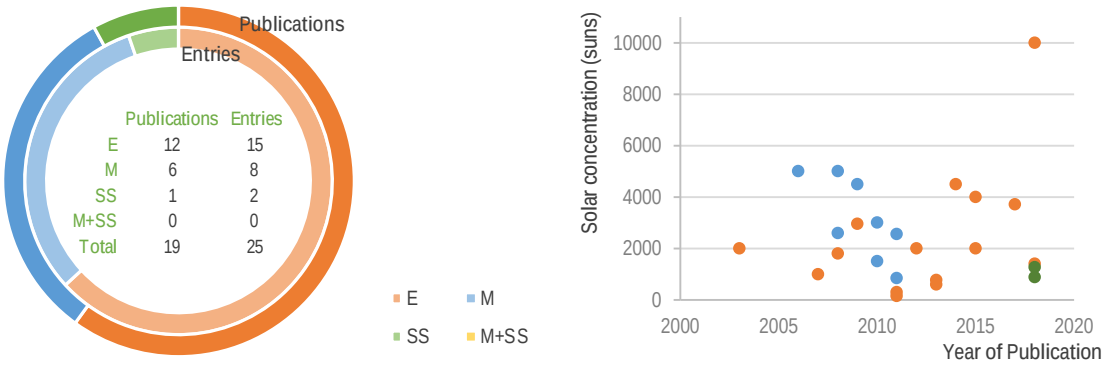


Figure 16. **Publications on gasification grouped according to type of work for the Solar Concentration parameter.**

2.2. Pyrolysis

The thermochemical pyrolysis process has been extensively used in research using a wide range of feedstock materials suitable to produce liquids and gaseous compounds of combustible nature or chemicals. Those extend from biomass to polymeric-based materials, from bituminous coal to oil shale, and to all materials or feedstock that require a reaction medium in absence of oxygen independently of reactor configurations or catalysts types. Pyrolysis offers the possibility to transfer the chemical energy contained in a solid-fuel type feedstock mostly into liquid energy carriers, although some gaseous and solid materials could also be found at room

temperature.

Reactors configuration extend from fixed-bed batch to autoclaves and from fluidized bed to plasma featuring different experimental conditions and purposes (Joseph Zeaiter et al., 2015; R. Edwin Raj et al., 2013; C. Berrueco et al., 2005; L. Tang and H. Huang, 2004; M. F. Laresgoiti et al., 2004; Adrian M. Cunliffe and Paul T. Williams, 1998). When considering large-scale conventional biomass technologies, different types of reactors are currently available extending from fixed-bed (Ozlem Onay, 2007) to fluidized bed (S. Antony Raja et al., 2010) including entrained flow (Gautami Newalkar et al., 2014), free-fall (Adisak Pattiya et al., 2012), vacuum (Manuel Garcìa-Pèrez et al., 2002), ablative (G. V. C. Peacocke and A. V. Bridgwater, 1994) and rotating cone (B. M. Wagenaar et al., 1994).

The operational conditions for pyrolysis are wide-ranging and the process is usually classified into fast, flash, and slow and catalytic pyrolysis. Both type of feedstock and operational conditions can be set in order to favour the production of solid, liquid or gas fractions. For example, fast pyrolysis and flash pyrolysis (which are associated to low residence times) generally favour the production of liquid yields, small amounts of gaseous compounds and, in some situations, almost no solids are formed (as secondary reactions are almost avoided). In contrast, slow pyrolysis produces larger amounts of solid compounds due to the increased conversion time which favour the existence of secondary reactions. Yet, similar experimental conditions associated to reactor and feedstock type could lead to different findings such as higher formation of liquid compounds as those found by other authors (Miguel Miranda et al., 2012; Miguel Miranda et al., 2015; Filomena Pinto et al., 2017). Liquids produced can be used directly (i.e., as boiler fuel and in some stationary engines) or refined into chemical and/or petrochemical industries for higher quality uses.

Taking into account the information presented in the Figure 8, the literature review on the main topic of pyrolysis and for the parameters of temperature, pressure, time and solar concentration is presented in Figure 17. These results also consider the type of work classifications previously assigned as being: a) distribution for both publications and entries and b) entries' year footprint. For this topic, only 28 publications were found (6 publications/year average) of which 67.9% were assigned to type of work of E, 17.9% to M, 10.7% to SS and only 3.6% to M+SS. These publications were all published between 2014 and 2018 and the analysis of data

reported resulted in 44 total entries.

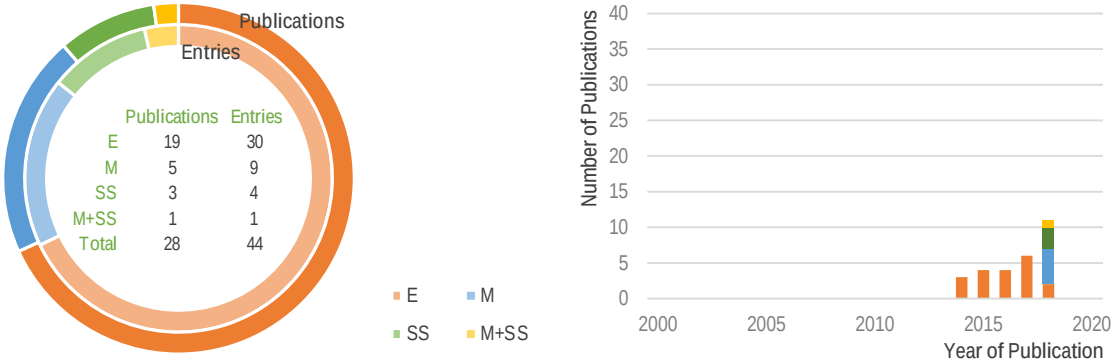


Figure 17. **Publications on pyrolysis grouped according to type of work.**

The information presented in the Figure 18 to Figure 21 depict the data for pyrolysis considering respectively the reference conditions of temperature, pressure, time and solar concentration as well as the type of work classifications previously assigned; a) distribution for both publications and entries; and b) entries' year footprint.

For the temperature parameter (Figure 18) 15 publications were found and most of them performing studies based on experimental work. As a result, 66.7% were assigned to the type of work E, 20.0% to the M and 13.3% the SS. No publications were found for M+SS type of work and all the information considered in this topic resulted in 29 total entries.

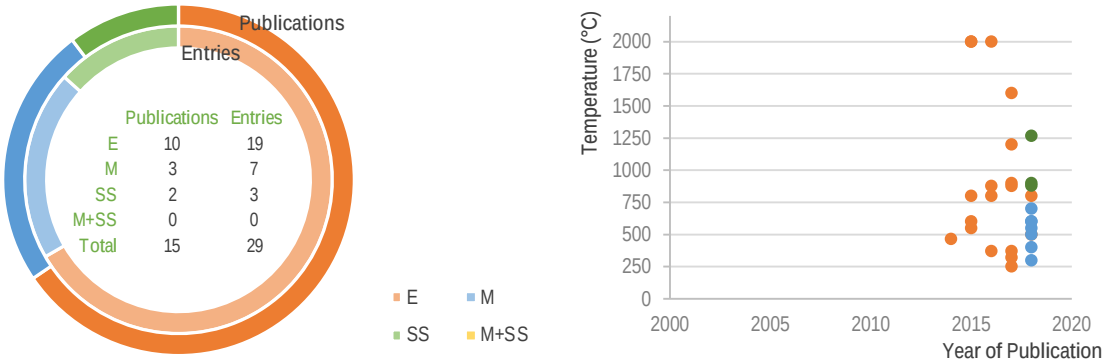


Figure 18. **Publications on pyrolysis grouped according to type of work for the Temperature parameter.**

With respect to pressure parameter, the results are presented in Figure 19. For this parameter 7 publications were found of which 85.7% were assigned to the type of work E and 14.3% to the M. No publications were found for the type of works of SS and M+SS, and the analysis of the reported data resulted in 8 total entries.

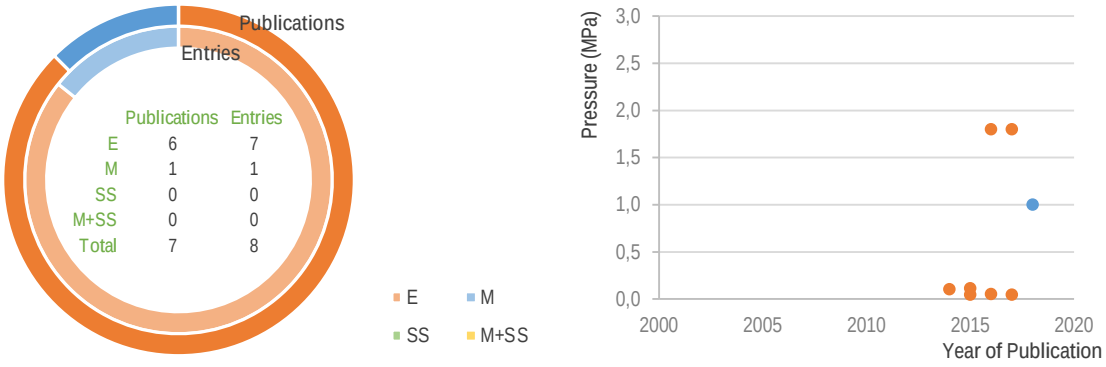


Figure 19. **Publications on pyrolysis grouped according to type of work for the Pressure parameter.**

When considering the results found for the time parameter (Figure 20), less studies were found (4 in total), being most of them assigned for the type of work E (75.0%) and only 1 publication for the M (25.0%) type of work. No publications were found for the SS and M+SS type of works and the information found resulted in 5 total entries.

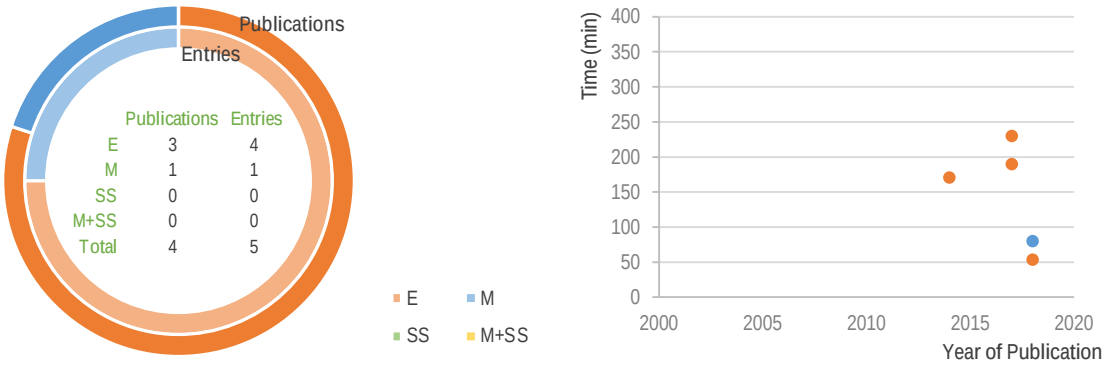


Figure 20. **Publications on pyrolysis grouped according to type of work for the Time parameter.**

For the solar concentration parameter (Figure 21), only 2 publications were found, 1 for each type of work SS and M+SS. The data reported in these publications results in a total entries of 2.

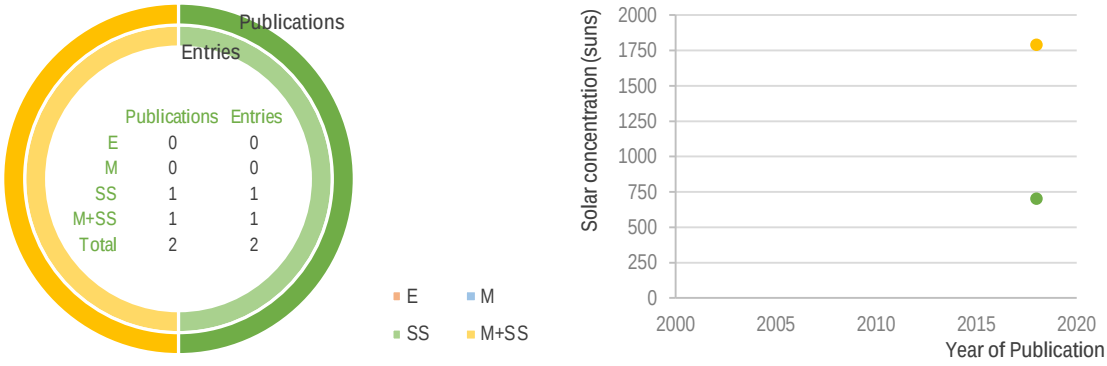


Figure 21. **Publications on pyrolysis grouped according to type of work for the Solar Concentration parameter.**

2.3. Redox reactions

The water splitting thermochemical cycles were initially proposed for the production of hydrogen from the dissociation of water into hydrogen and oxygen. Thermolysis is

conceptually the simplest single-step thermal dissociation of water splitting reaction. Unfortunately, this reaction not only requires very high temperatures (above 2200°C) but also presents the problem of effectively separating H_2 and O_2 avoiding explosive mixtures.

Thermochemical cycles consist in two or more consecutive chemical reactions in which their "net" sum is the H_2O splitting into H_2 and O_2 products. However, the maximum temperature step occurs at a temperature level lower than the single-step water decomposition chemical reaction. As a result, thermochemical cycles avoid the H_2 and the O_2 separation problem, which allow the operation to take place at moderated high temperatures. Considering that the reactions occurring in this step are highly endothermic by nature, the input of an external energy source for the process to occur is mostly required (Christos Agrafiotis et al., 2015).

Among a significant number of thermochemical cycles found on the literature review, the two-step ones based on oxide redox pair systems are of particular interest. These cycles are based on the principle of transition between the oxidized (higher valence MeO oxidized) and reduced (lower-valence MeO reduced) form of an oxide of a metal capable of exhibiting multiple oxidation states (Tatsuya Kodama and Nobuyuki Gokon, 2007). In this concept, in the first step of reaction (high-temperature endothermic reaction) the higher-valence oxide of such a metal is subjected to thermal reduction (TR) in which a quantity of oxygen is release and transforms the metal into a low-valence state (TR, reaction 1). In a subsequent 2nd step, an exothermic reaction at lower-temperature known as water splitting (WS), the reduced oxide is then oxidized back to the higher-valence state by taking oxygen from water and producing hydrogen. When reaction 1 and 2 are completed, a cyclic operation is then established (Christos Agrafiotis et al., 2015). The production of hydrogen (from H_2O) and carbon monoxide (from CO_2) can be combined leading to the production of syngas (also described as "solar syngas" (Robert C. Pullar et al., 2019).

This "solar syngas" is suitable for be used in a Fischer-Tropsch (FT) technology in a number of different product streams (i.e., diesel-like fuel, alcohols and other chemicals). As this approach is conceptually simple, the TR step is commonly used for both WS and carbon dioxide splitting (CDS) reactions allowing particular redox material and the respective thermochemical reactor to be separately used from each other (production of a H_2 stream separately from the CO stream. The process can be

performed simultaneously when steam and CO_2 are co-fed to the redox material, to produce syngas on a single step.

Highest dissociations are usually required, as the subsequent full replenishment of the oxygen released during dissociation of oxygen taken from H_2O/CO_2 during oxidation, results in higher H_2/CO product yields (per mass of redox material) which results in higher cycle efficiencies. These cycle efficiencies correlate the fuel's energy content with the amount of energy that must be supplied to the reaction medium in order to fulfil the entire cycle (i.e., relation between the higher heating value (HHV) of the fuel produced and the thermochemical cycle energy input). Thus, hydrogen or syngas production have been well achieved through both cycles of metal oxide/metal systems (i.e., ZnO/Zn) or metal oxide/metal oxide pairs. Some typical examples are the oxides of a single multivalent metal such as Fe_3O_4/FeO and Mn_3O_4/MnO or the CeO_2/Ce_2O_3.

Metal oxides thermochemical cycles such as zinc oxide (ZnO) is an example of syngas production. A significant number of reactor concepts have been developed by researchers worldwide for ZnO reduction using solar energy at small-scale (from packed beds to entrained and aerosolized flows (Christopher Perkins et al., 2008; Erik Koepf et al., 2012) to quasi-batch arrangements (Marc Chambon et al., 2010; L. O. Schunk et al., 2008; Stéphane Abanades et al., 2007; Reto Müller et al., 2006; P. Haueter et al., 1999). In the solar-driven endothermic reduction of ZnO, metallic Zn is produced at temperatures above 1,700°C followed by an oxidation reaction (exothermic) between Zn and H_2O (and/or CO_2) at lower temperature (< 1,000°C). As a result, a synthetic gas (syngas) is produced capable to be further processed in a FT synthesis towards the production of liquid synthetic hydrocarbon fuels for an increasing number of purposes (Irving Wender, 1996). Other similar example is the zinc produced carbothermally where coal, charcoal and other carbonaceous materials are consumed providing a reduction of process temperature of around 300°C and resulting in the formation of a gas mainly composed of Zn(g) and CO (rather than Zn(g) and O_2).

The technological development, both in materials and reactors capable of achieving and sustaining high temperatures when irradiated with concentrated solar irradiation, are currently in research. Such example is the use of ceria as redox active material for thermochemical cycles by changing the chemistry involved. In the ceria-based cycle,

the physico-chemical properties can be modified by changing its chemical composition, although to date, different operating conditions may be found on literature, during thermal reduction and re-oxidation gas separation is still a critical issue that affects the efficiency of the process.

Some small- and large-scale reactors featuring these applications and developments were recently published by E. Koepf et al., 2017. The materials used for both the window and the cavity receiver must be carefully chosen as they represent the most sensitive components of solar reactors that are commonly based on either direct or indirect heating of the reactants using concentrated solar energy. Direct heating provides efficient and fast heat transfer directly to the reacting matter but such reactor technologies must be designed so that to avoid particle deposition on the optical window (such as entrained particles and particle-laden flow). Alternatively, indirect heating via a heat transfer wall requires using high-temperature resistant refractory materials and may suffer from additional heat losses due to indirect heat transfer. Conventional solar reactor designs usually make use of insulated cavity-type black body receivers that allow obtaining almost isothermal conditions in the cavity volume and high solar energy absorption efficiencies (P. Haueter et al., 1999; Reto Mülleret al., 2006; L. O. Schunk et al., 2008; E. Koepf et al., 2016). High-temperature resistant ceramic materials (refractory) are usually employed for lining the inner reactor volume (including both cavity walls and insulation materials).

Considering once again the information presented in the Figure 8**Error! Reference source not found.**, the literature review on the main topic of redox reactions for the parameters temperature, pressure, time, solar concentration and number of cycles is presented in Figure 22. These results also take into account the type of work classifications previously assigned as being: a) distribution for both publications and entries and b) entries' year footprint. For this main topic, 233 publications were found between 2004 and 2019 (15 publications/year average), being 57.9% of them assigned to type of work E, 30.5% to M, 7.3% to SS and 4.3% to M+SS. These results also reveal that type of work E has been carried out continually over the years although between 2017 and 2018 is more noticeable, while type of work M was almost inexistent until 211 increased significantly its number of publications between 2014 and 2018. For the type of work SS, occasional works have been published over the years while M+SS type of work reveals less publications (10 in total) found specifically in the years of 2007, 2014 and 2015. The analysis of the information on this topic resulted in 507

total entries.

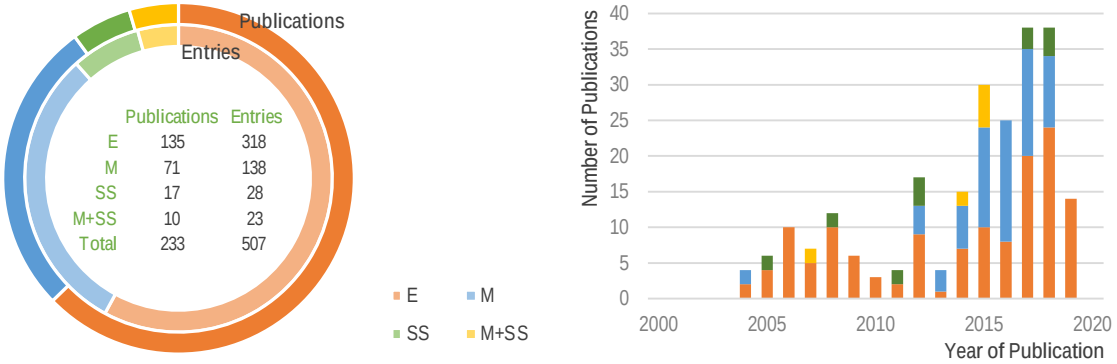


Figure 22. **Publications on redox reactions grouped according to type of work.**

Figure 23 to Figure 27 depict the data for redox reactions considering sequentially several reference conditions, such as temperature, pressure, time, solar concentration and number of cycles and the type of work classifications previously assigned; a) distribution for both publications and entries; and b) entries' year footprint.

The results found for the temperature parameter (Figure 23) reveals 86 total publications of which 57.0% were assigned to the type of work E, 31.4% to the M, 7.0% to the SS and only 4.7% to the M+SS. In the analysis of the published data, 260 entries were considered and the largest slice in the chart represents 162 entries that were assigned to studies that perform only experimental work.

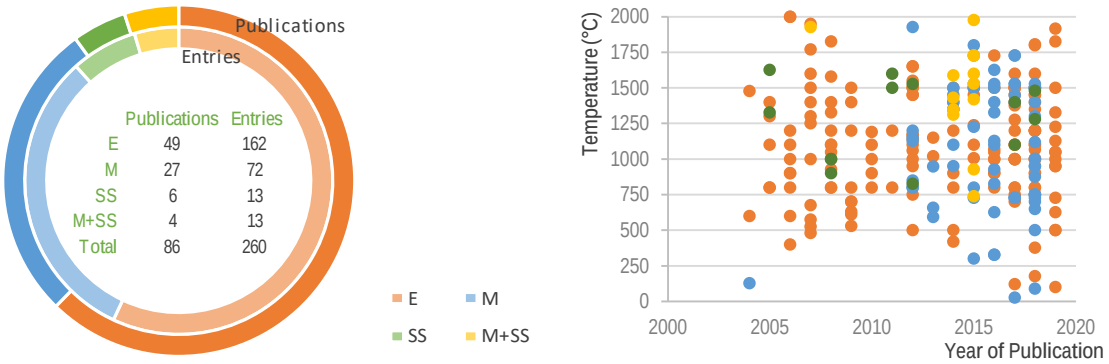


Figure 23. **Publications on redox reactions grouped according to type of work for the Temperature parameter.**

In the Figure 24 is presented all the information obtained for the pressure parameter. In this case, 43 publications were found of which 48.8% were equally distributed for each type of work E and M, and 2.3% to SS type of work. No publications were found for the M+SS type of work and the analysis of the data resulted in 79 total entry being 45 assigned to the type of work E and 33 to the type of work M.

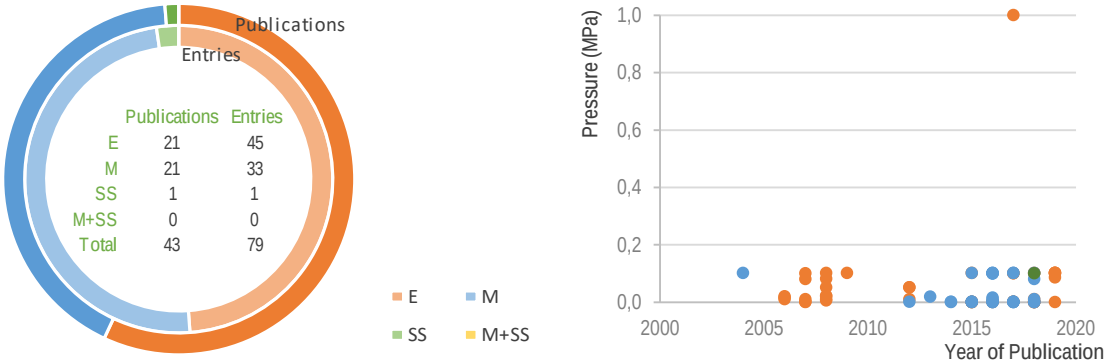


Figure 24. **Publications on redox reactions grouped according to type of work for the Pressure parameter.**

Regarding the time parameter (Figure 25), the results reveal that 48 publications were found, of which 60.4% were assigned to the type of work E, 18.8% to M, 12.5% to SS

and 8.3% to M+SS. The information found in this group resulted in 97 total of entries being most of them assigned to the type of work E (64 entries). A similar outcome was found for the solar concentration parameter (Figure 26). As a result, 31 publication were reported of which 48.4% were assigned to the type of work E, 41.9% to M and 3.2% and 6.5% respectively to type of works SS and M+SS. The analysis of the data yielded a total of 41 entries being most of them assigned to the type of works E (21 entries) and M (16 entries).

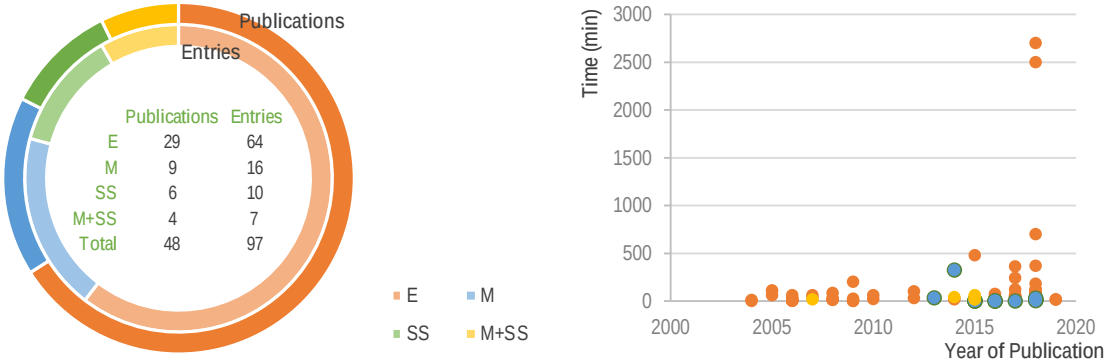


Figure 25. **Publications on redox reactions grouped according to type of work for the Time parameter.**

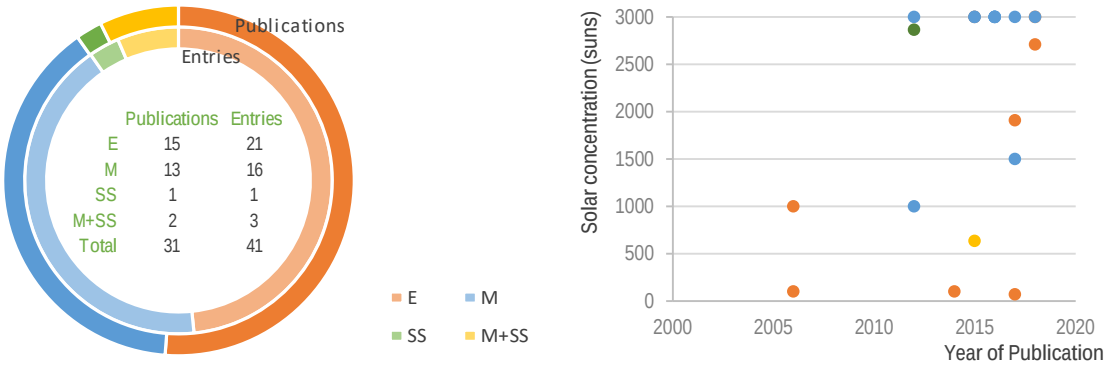


Figure 26. **Publications on redox reactions grouped according to type of work for the Solar Concentration parameter.**

For the number of cycles parameter (Figure 27) the literature review reveals 25 as total publications. From those, the great majority (84.0%) were assigned to type of work E while the remaining publications were only associated to M (4.0%) and SS (12.0%) type of works. The analysis of the data published result in 30 total entries most of them focus on type of work E (26 entries).

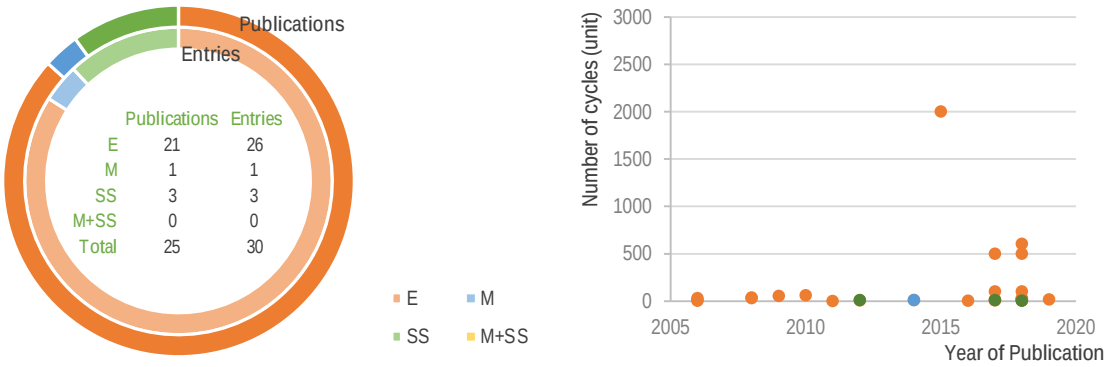


Figure 27. **Publications on redox reactions grouped according to type of work for the Cycles (number of) parameter**.

3. Conclusions

The main purpose of this document is to support a standardised method of assigning a figure of merit to each performance metric previously established through literature review on the topic of solar thermochemical reactors towards fuel production using concentrated solar energy. As such, more than 200 articles were thoroughly analysed on the topic of solar thermochemical processes for fuel production and grouped according to thermal processes: i) gasification, ii) pyrolysis and iii) redox reactions.

Due to the significant amount of information published, the proposed objectives found on each topic and to make feasible the approach, it was developed a database form in Visual Basic for Applications in order to both fine-tune and group accordingly all the information. For each type of work previously established, an additional structure was considered in which the information was organized considering each reference parameter. This structure was defined as being: i) experimental work in real conditions (E), ii) modelling work (M), iii) experimental work on solar simulator (SS) and iv) modelling with solar simulator work (M+SS) considering the reference parameters of temperature, pressure, time, solar concentration and number of cycles (if available). Afterwards, the information reported of each publication was statistically treated and graphically represented considering all parameters in the subject of analysis.

For the main topic of gasification, 119 publications were found between 2003 and 2019 (7 publications/year average) distributed for each type of work as being 57.1% E, 22.7% M, 15.1% SS and 5.0% M+SS. From those publications, 51 were assigned to the temperature parameter being 58.8% E, 21.6% M, 13.7% SS and 5.9% M+SS, while for the pressure parameter 16 publications were found of which 68.8% E, 12.5% M and 18.8% SS as no publications were found for the M+SS type of work. Regarding the time parameter, 33 publications were found being 45.5% E, 24.2% M, 21.2% SS and 9.1% M+SS, whereas for the solar concentration parameter 19 publications were found being 63.2% E, 31.6% M and 5.3% SS as no publications were found for the M+SS type of work.

For the main topic of pyrolysis, 28 publications were found between 2014 and 2019 (6 publications/year average) distributed for each type of work as 67.9% E, 17.9% M, 10.7% SS and 3.6% M+SS. From the total publications, 15 were assigned to the temperature parameter being 66.7% E, 20.0% M and 13.3% SS as no publications

were found for the type of work M+SS, while for the pressure parameter 7 publications were found being 85.7% E and 14.3% M, and no publications were found for the SS and M+SS type of works. When considering the time parameter, a total of 4 publications were found being 75.0% E and 25.0% M as no publications were found for type of works SS and M+SS while for the solar concentration parameter only 2 publications were found (one for each type of work SS and M+SS).

For the main topic of redox reactions, 233 publications were found between 2004 and 2019 (15 publications/year average) distributed for each type of work as 57.9% E, 30.5% M, 7.3% SS and 4.3% M+SS. From that total, 86 were assigned to the temperature parameter being 57.0% E, 31.4% M, 7.0% SS and 4.7% M+SS, while for the pressure parameter 43 publications were found of which 48.8% were equally distributed for each type of work E and M, and 2.3% for SS as no publications were found for the M+SS type of work. For the time parameter, 48 publications were considered being 60.4% E, 18.8% M, 12.5% SS and 8.3% M+SS, while for the solar concentration parameter 31 publications were found distributed as 48.4% E, 41.9% M, 3.2% SS and 6.5% M+SS. When considering the number of cycles parameter, 25 publications were found most of them associated to the type of work E (84.0%) while the remaining publications were only associated to the M (4.0%) and SS (12.0%) type of works. No publications were found addressing the type of work M+SS.

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