Ref. Ares(2024)7679926 - 29/10/2024

ABraytCSPfuture

Air-**Brayt**on Cycle **C**oncentrated **S**olar **P**ower **future** plants via redox oxides-based structured thermochemical heat exchangers / thermal boosters

Deliverable 7.1.

Deliverable: Environmental-friendly materials selection

Dissemination Level: Public

WP7 Environmental & socio-economic sustainability and techno-economic assessment

Date: 29.10.2024

https://www.abraytcspfuture.eu/

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement no 1010845.

Disclaimer

Grant Agreement Number: 101084569	**Acronym: ABraytCSPfuture**			
Full Title	**A**ir-**Brayt**on Cycle **C**oncentrated **S**olar **P**ower **future** plants via redox oxides-based structured thermochemical heat exchangers/thermal boosters			
Call Topic Identifier:	HORIZON-CL5-2021-D3-03			
Funding Scheme	HORIZON EUROPE, RIA – Research and Innovation Actions			
Project start date:	01.11.2022			
Project duration:	48 months			
Project URL	https://www.abraytcspfuture.eu/			
EU Project Officer	Clara Astudillo Llorente			
Project Coordinator	DLR – German Aerospace Center			
Deliverable	7.1 – Environmental-friendly materials selection			
Date of Delivery	Contractual	31.10.2024	Actual	29.10.2024
Nature	Report			
Dissemination Level	Public			
Lead Beneficiary	Fraunhofer IWKS			

Document History

Version	**Issue Date**	**Change History**	**Authors**	**Organisation**
1.0	11.10.2024	Document drafted	Zeller, Vanessa Kinski, Isabel Nazneen, Arooba Ionescu, Emanuel	Fraunhofer IWKS
1.1	22.10.2024	Updated with final partner feedback from CERTH and DLR	Zeller, Vanessa	Fraunhofer IWKS
1.2	28.10.2024	Final Review	Singh, Abhishek	UT

About the Project

ABraytCSPfuture sets forth an innovative, carbon-neutral way for implementing the highly efficient air-Brayton gas turbine power generation cycles into future air-operated Concentrated Solar Power (CSP) plants. Air-Brayton cycles are used in traditional power plants involving fossil fuels combustion via pressurised air. *ABraytCSPfuture*'s carbon-neutral approach aims at achieving higher solar-to-electricity efficiencies, vital for competitiveness of CSP and non-reachable by either PVs or molten salts and thermal oils, increasing in parallel significantly the plants' storage capability. The project will develop and demonstrate a first-of-its-kind compact, dual-bed thermochemical reactor/heat exchanger unit that will transfer heat from a non-pressurised air stream to a pressurised one, while also operating as a thermal booster, raising the temperature of the pressurised stream to the level required for Brayton cycles. Furthermore, the volumetric solar energy storage density of air-operated CSP plants will be significantly increased by rendering their current sensible-only regenerative storage systems to hybrid sensible-thermochemical storage ones within the same storage volume. Both these functionalities will be materialised by thermochemical reactor/heat exchanger units comprised of non-moving, flow-through porous ceramic structures (honeycombs or foams) based on earth-abundant, cost-efficient, non-toxic oxide materials and exploiting reversible reduction/oxidation reactions of such oxides in direct contact with air, accompanied by significant endothermic/exothermic heat effects. The proposed technology is set forth by an interdisciplinary partnership spanning the entire CSP value chain, comprised of leading research centres, universities, innovative SMEs and large enterprises, including ancillary services providers and technology end-users.

Deutsches Zentrum fuer Luft- und Raumfahrt e.V., **DLR**	DE	DLR
Centre for Research & Technology Hellas, **CERTH**	EL	CERTH CENTRE FOR RESEARCH & TECHNOLOGY HELLAS
University of Twente, **UT**	NL	UNIVERSITY OF TWENTE.
Fundación CENER, **CENER**	ES	CENER NATIONAL RENEWABLE ENERGY CENTRE ADItech
Fundación TEKNIKER, **TEKN**	ES	Tekniker
Fraunhofer Gesellschaft zur Foerderung der angewandten Forschung e.V., **FHG (IWKS)**	DE	Fraunhofer IWKS
DESTINUS Energy BV**, DEST**	NL	Destinus
KRAFTBLOCK GmbH, **KB**	DE	KRAFT BLOCK
LANDSON Emission Technologies A/S, **LET**	DK	landson
COBRA Instalaciones y Servicios S.A., **COBRA**	ES	cobra

Table of Contents

This deliverable is part of a project that has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement no 101084569

Executive Summary

The present document constitutes Deliverable D7.1 "*Environmental-friendly materials selection*" developed within *WP7 Environmental & socio-economic sustainability and techno-economic assessment* of the HORIZON EUROPE 2020 ABraytCSPfuture project.

The document describes the Life Cycle Assessment (LCA) that is performed to support the selection of environmentally performant synthesized redox oxide materials applicable for hybrid sensible/thermo-chemical storage (TCS) systems, up to 1100°C.

Deliverable D7.1 defines the functional unit and the studied system which includes (i) the provision of raw materials, (ii) the synthesis of the powder and (iii) the production of the porous structures. Based on the reference composition $CaMnO_3$, two different processing routes for the production of two kinds of porous ceramic structures are described, namely for honeycombs and so-called foams. Both processing routes are modelled in two scenarios: in the *current lab scenario* that represents the current processing routine with averaged experimental batch sizes in the respective partners' laboratories as well as in a *lab scale max* scenario that refers to maximum capacities currently installed in the labs. The chosen impact assessment method ReCiPe and selected impact categories such as the global warming potential, the material resource scarcity and a normalised single point that aggregates 18 impacts into one value are presented.

Deliverable D7.1 presents the life cycle inventory data for these novel processing routes that was compiled from primary data (process in- and outputs or process parameters) from the project partners (mainly DLR and CERTH at this preliminary stage) and combined with LCI data from LCI databases and literature. Impact assessment results are presented for the single, midpoint and endpoint impacts for four scenarios, related to 1kg of synthesized redox oxide materials processed in either honeycombs or foams. The results show most performant environmental profiles for the foam route in the *lab scale max* scenario and identify the most significant process contributors. This outcome though, is primarily due to the higher relevant batch sizes in the milling, calcination, and sintering processes and, indeed, sensitivity analysis shows that the difference between the two processing routes is below 10% when lab conditions are more harmonised.

Changes with respect to the DoA

No changes or Deviations from the Work plan occurred with respect to this Deliverable *D7.1 Environmental-friendly materials selection*.

1. Introduction

1.1 Motivation & previous work

As introduced in the project description, an innovative thermo-chemical storage (TCS) system is developed in *ABraytCSPfuture* for which different combinations of materials, different synthesis routes and shaped product configurations are being researched to identify the most performant ones. In addition to analyses targeting material performances, a Life Cycle Assessment (LCA) is performed to support the selection of environmentally performant synthesized redox oxide materials applicable for TCS systems. LCA is a method to assess environmental impacts of products and services throughout the life cycle of a product. The method is standardised in ISO 14040 and 14044 and is broadly used to compare environmental performances and to determine the most performant material combinations, production routes or product alternatives. It is highly relevant to apply LCA to an innovative system such as the one developed in *ABraytCSPfuture* that is still in its initial development phase, when the design freedom is still high. Through this early integration of environmental considerations into the development process, the material selection can be guided and processes can be optimised from environmental perspective. Further, such early-stage assessments are the starting point for future assessments that use upscaling approaches to anticipate the environmental performance at higher development level in order to benchmark with existing systems.

Thus, the focus of this LCA is to perform such an early-stage assessment and to compare different redox oxide structured materials with different primary production and synthesis routes. More precisely, this report focusses on the investigation of $CaMnO_3$ which was chosen as the reference composition.

As preparatory work for the selection of environmentally performant synthesized redox oxide materials, many different materials compositions have been investigated in the project, resulting in 6 shortlisted compositions synthesised of carbonates ($SrCO_3$, $CaCO_3$, $LaCO_3$) and oxides (Mn_3O_4, Fe_3O_4/Fe_2O_3, NiO, CuO, Co_3O_4) in a solid-state reaction. The environmental impact intensity for the provision of the 13 different raw materials was investigated with a screening LCA (documented in 2024 technical report as well as in the supplement Table S 1). While this previous assessment related impacts on a kg raw material basis, this report includes the synthesis steps and relates environmental impacts to the produced honeycomb and foam structures.

1.2 State of the art

A significant number of LCA studies on Concentrated Solar Power (CSP) systems have been published over the past decade, as summarized by Lamnatou and Chemisana in their 2017 review of the state-of-the-art. A typical functional unit in these studies is the production of electricity fed into the grid, often represented by 1 MWh of electricity

(MWh_{el}). The review found that electricity from CSP plants generally exhibits a Global Warming Potential (GWP) of less than 40 g CO_2eq/kWh_{el} and an Energy Payback Time (EPBT) of less than one year, which is substantially lower than that of conventional power plants.

Various components of thermal energy storage (TES) systems within CSP plants have also been analysed using LCA. TES systems examined include those based on sensible heat storage (primarily molten salts), latent heat storage using Phase Change Materials (PCMs), and thermochemical energy storage (TCES) technology. The first LCA of TCES was conducted in 2019 (Pelay et al. 2020), investigating different TCES integration scenarios for a hypothetical solar power tower plant using calcium hydroxide as the TCES material. This LCA showed a GWP of approximately 11 kg CO_2eq/MWh_{el} and an energy payback time of about four months, with the integration of the TCES system contributing an additional environmental burden of around 30%. In a more recent study, Colelli et al. explored calcium looping as a thermochemical energy storage technology, finding a GWP of 24 kg CO_2eq/MWh. The storage system contributed significantly to various impact categories, ranging from 30% to 85%.

While these previous studies included assessments of the environmental performance of TCES systems, they focused on the overall system performance rather than isolating specific components or materials. LCAs for thermochemical materials and components have been conducted (e.g., Nienborg et al., Hayatina et al.), but primarily for building applications rather than high-temperature TCES systems. Material screening has been carried out for typical TCES systems, including hydrides, metal oxides, and organic systems (Chen et al. 2018), and with a particular emphasis on pure and mixed-metal oxides for CSP applications (Karagiannakis et al. 2014, Dizaji and Hosseini 2018) as well as for perovskite oxides as TCES materials (Cai et al. 2023). However, full life cycle-based assessments of these materials and TCES systems have not been conducted. Environmental aspects such as toxicity are mentioned in some studies, but comprehensive LCAs remain absent.

Without an application-specific focus, LCA studies and Life Cycle Inventory (LCI) data are available for various mixed metal oxides, perovskite-form structures (Ordoñez et al. 2023), and bulk metals and specialty metals (Nuss and Eckelman 2014), but these are not yet tailored to high-temperature TCES systems for CSP applications.

1.3 Report structure

The report follows the structure of the four phases of LCA, starting with the goal and scope description (chapter 2). The data base and modelling are reported in the life cycle inventory (chapter 3), followed by the impact assessment reported in (chapter 4). The report closes with the interpretation of results (chapter 5).

2. Goal and scope description

2.1 Goal definition

The goal of this LCA is to provide decision support regarding the design of the processing routes for the production of synthesized redox oxide materials applicable for TCS systems. The decision at hand is the selection of different materials/combinations and processing route of redox oxide materials at current laboratory scale and a future best-lab performance scenario in a European context. The decisions have to be implemented by the process engineers, so they are the main target audience. Results can also be relevant for the manufacturer of components for CSP plants.

The resulting research questions are:

- What are the environmental performances for the different material combinations and shaping processes (i.e., honeycombs and foams)?
- How can performances be improved when producing at an optimised laboratory scale?

2.2 Functional unit

The function of the produced redox honeycombs and foams is the storage of energy. Thus, the functional unit should be defined as the storage of a certain amount of energy over a certain time period, for example over the life time of the storage unit.

While the storage performance potential can be specified based on the project's relevant target (i.e., ≥300 kWh /$m^3$of structure[1]) for the produced honeycombs and foams, the performance over time is currently under evaluation. Therefore, at the current stage of the project, the material/ product synthesis is in the focus, so a declared unit of **1kg of synthesized redox oxide materials shaped in a honeycomb or foam structure** is defined.

2.3 System boundary

The studied system is structured into three main parts: (i) the provision of raw materials, (ii) the synthesis of the powder and (iii) the production of the porous structures. A so-called "cradle to gate" system boundary is applied, starting with the extraction of resources until the production of the porous structures and, at this relatively early stage of development in the project's course, excluding the use and end of life phase of the developed materials.

[1] or equivalently indicative gravimetric energy density of around 900 kJ/kg for a conservative packing density of the structure on the order of 1.2 kg/L

A generic process chain is presented in Figure 1 illustrating the so-called foreground system, i.e., the system that is developed in the project composed of powder synthesis and the production of the porous structures. The background system describes the processes that have not been particularly developed in the project's context, but still relevant for the environmental performance of the studied system such as the raw material provision and electricity and process additives that serve as an input into the foreground system.

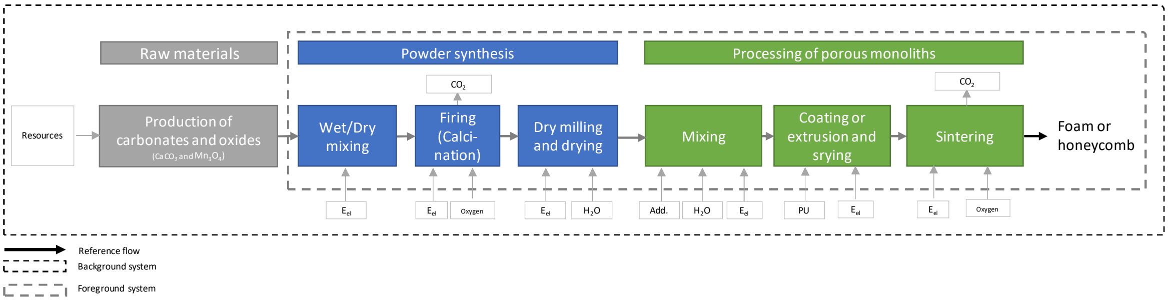

Figure 1: Generic representation of the studied system

Raw material production: As stated in the introduction, many different materials compositions have been investigated in the project, resulting in 6 shortlisted compositions synthesized using carbonates ($SrCO_3$, $CaCO_3$, $LaCO_3$) and oxides (Mn_3O_4, Fe_3O_4/Fe_2O_3, NiO, CuO, Co_3O_4) as precursors. The environmental impact intensity for the provision of the raw materials was investigated with a screening LCA (see supplement 1). At the time of preparation of the present document, several compositions have been qualified as promising (see deliverable D2.4), but LCI process data could be delivered only for one reference material (without the dopants) that can serve as a reference point. Thus, this report focusses on the investigation of $CaMnO_3$ (CMO) and the main targeted raw materials to be investigated in this LCA are $CaCO_3$ and Mn_3O_4.

The environmental impacts of these raw materials are determined based on data from the database ecoinvent (Frischknecht et al. 2005; Weidema et al. 2013). Mn_3O_4 production includes the production of manganese concentrate (mining and beneficiation), the roasting of manganese carbonate in a rotary kiln as well as transport processes. $CaCO_3$ includes production of limestone (mining, beneficiation, transport). Specific datasets are reported in chapter 3.

Powder synthesis: The synthesis of $CaMnO_3$ powder is the second step within the studied system. It includes the processes illustrated in Figure 1 including dry (or wet) mixing of the raw materials, firing (calcination) to synthesize the $CaMnO_3$ phase and a dry milling process; the last one is necessary for further processing of the powder towards shaping of both kinds of monolithic structures. Data for the powder synthesis is provided by the project partners and reported in 3.1.2. The impact of the used equipment, such as mills or furnaces is not considered in this assessment.

Production of the porous structures: The production of the porous structures, i.e., the shaping procedure, includes the processes from the mixing of the milled powder with compounds necessary for the production of the target product which is either a foam or a honeycomb structure. The processes cover mixing of the powders with organic additives that facilitate their shaping, coating of polymeric templates (foams) or extrusion (honeycombs) and sintering. Data for the powder synthesis is provided by the project partners and is reported in 3.1.3. The impact of the used equipment, such as extruder or furnaces is not considered in this assessment.

2.4 Studied scenarios

Table 1 shows the four scenarios that are created to reflect on the one hand the different lab conditions and on the other hand the different structures that are produced (i.e., honeycombs denoted as H and foams denoted as F). The **current lab scale** represents the current use of available lab equipment with averaged experimental batch sizes, while the **lab scale max** scenario represents the maximum capacities currently installed in the labs. This is especially relevant for the use of furnaces that are currently used to sinter smaller charges compared to their max batch size.

Table 1: Scenario overview

	Lab, current	**Lab, max**
Honeycombs (CERTH)	**Lab current H** - <0.5kg scale for powder synthesis - 0.2kg sintering	**Lab max H** - Up to 2kg for powder synthesis - 2kg sintering
Foams (DLR)	**Lab current F** - 0.5kg scale for powder synthesis - 0.25kg sintering	**Lab max F** - Up to 2.5kg scale for powder synthesis - 2.5kg sintering

2.5 System model and selected impact assessment method

An attributional system model is applied to assess environmental impacts of the current lab scale system. The chosen LCI inventory database is ecoinvent (version 3.9.1., cut-off system model).

Due to its scientific credibility and comprehensiveness, the chosen impact assessment method for *ABraytCSPfuture* is ReCiPe (Huijbregts et al. 2017). This impact assessment method provides 18 midpoint and 3 endpoint impact categories, such as damage to human health (HH), damage to ecosystems (ES) and damage to resource availability (Res). As an interesting feature for the project, the endpoints can be converted into one aggregated single point result with the help of normalisation methods. For *ABraytCSPfuture* three impact categories have been selected, that are of particular relevance for the project, these being

1. **Normalised single point** as an indicator for the total environmental damage
2. **Mineral resource scarcity** as an indicator for the impact on resource scarcity

3. **Global warming potential** as an indicator to measure climate change.

Characterisation factors and units are presented in Table 2. The global warming potential is measured in kg CO_2 eq with a time horizon of 100 years based on values from IPPC 2021. The 100-year time horizon GWP expresses the potential of greenhouses gases in relation to the potential of CO_2 over a time horizon of 100 years.

Table 2: Overview on impact categories, characterisation factors and units used in ReCiPe

Normalised categories	**[-] normalized score in points per FU**	
	Endpoint categories	
Endpoint categories	**Characterization factor**	**Unit**
Human health	Disability-adjusted loss of life years (DALY)	$DALY/kg_{emitted}$
Ecosystem damage	Biodiversity loss per year	Species per year
Resource scarcity	Increased costs	USD 2013
	Midpoint categories	
Impact	**Characterization factor**	**Unit**
Fine particulate matter formation	Fine dust formation potential	kg PM2.5 eq.
Fossil resource scarcity	Fossil fuel potential	kg oil eq.
Freshwater ecotoxicity	Freshwater toxicity potential	kg 1,4-DCB
Freshwater eutrophication	Freshwater eutrophication potential	kg P eq.
Global warming	Global warming potential	kg CO2 eq.
Human carcinogenic toxicity	Potential for human toxicity	kg 1,4-DCB
Human non-carcinogenic toxicity	Potential for human toxicity (non-carcinogenic)	kg 1,4-DCB
Ionizing radiation	Ionizing radiation potential	kBq Co-60 eq.
Land use	Agricultural land potential	m^2a crop eq.
Marine ecotoxicity	Marine ecotoxicity potential	kg 1,4-DCB
Marine eutrophication	Marine eutrophication potential	kg N eq.
Mineral resource scarcity	Excess ore potential	kg Cu eq.
Ozone formation, Human health	Photochemical oxidant formation potential: humans	kg NOx eq.
Ozone formation, Terrestrial ecosystems	Photochemical oxidant formation potential: ecosystems	kg NOx eq.
Stratospheric ozone depletion	Ozone depletion potential	kg CFC-11 eq.
Terrestrial acidification	Terrestrial acidification potential	kg SO2 eq.
Terrestrial ecotoxicity	Terrestrial ecotoxicity potential	kg 1,4-DCB
Water consumption	Water use potential	m^3

3. Life cycle inventory

The life cycle inventory (LCI) is the second phase of LCA. It is an inventory of input/output data with regard to the system being studied. Section 3.1 specifies the process data that was used to build the LCI model, for the foreground and background systems.

3.1 Data

3.1.1 Provision of raw materials

The overview of LCI data to model the provision of raw materials is shown in Table 3. Two raw materials are needed for the synthesis of CMO powder: $CaCO_3$ and Mn_3O_4. The amounts of needed reactants are determined based on stoichiometric quantities derived from equation 1, which typically refers to calcination of mixed raw powders in air at high temperature to promote the solid-state reaction for the formation of CMO:

$$3\ CaCO_3 + 1\ Mn_3O_4 + O_2 \rightarrow 3\ CaMnO_3 + 3\ CO_2 \qquad \text{Equation 1}$$

resulting in 700g $CaCO_3$ and 533g Mn_3O_4 for the production of 1 kg of $CaMnO_3$.

The mass balance is maintained by CO_2 emissions that are not included in raw material provision, but in the powder synthesis step.

Table 3: LCI data for raw material provision

		Lab current H	**Lab max H**	**Lab current F**	**Lab max F**	**Data source**	**Dataset**
inputs	***unit***						
$CaCO_3$	g/kg_{out}	700	700	700	700	Stochio-metric calculation	[1]
Mn_3O_4	g/kg_{out}	533	533	533	533		[2]
outputs							
Reactants	kg	1	1	1	1		
[1]	Calcium carbonate, precipitated {RER}| market for calcium carbonate, precipitated | Cut-off, U						
[2]	Manganese(III) oxide {GLO}| market for manganese(III) oxide | Cut-off, U						

3.1.2 Powder synthesis

As illustrated in Figure 1 powder synthesis starts with the **mixing/ process** and refers to the mixing conditions currently applied in the labs of the project partners, with different batch sizes.

For the **firing (calcination) process**, process conditions are reported by the project partners (see Figure 2). A typical furnace runs for 25 hours when isothermal time at the maximum temperature is 16 hours as furnace needs time to heat and cool down. The max. temperature is 1300 °C. The energy consumption to run the furnace at these conditions is estimated as 37kWh.

It is estimated that for the honeycomb route approx. 0.1 kg of reactants are introduced into the furnace at current lab scale and 2kg at max lab scale. Energy consumption is scaled accordingly and specified for 1 kg of reactant powder (Table 4). For the foam route, 0.5kg of reactants are used at current lab scale and 2.5 kg can be used in the max lab scale.

CO_2 process emissions are derived from Equation 1) and correspond to around 308 kg CO_2 for 1kg of products.

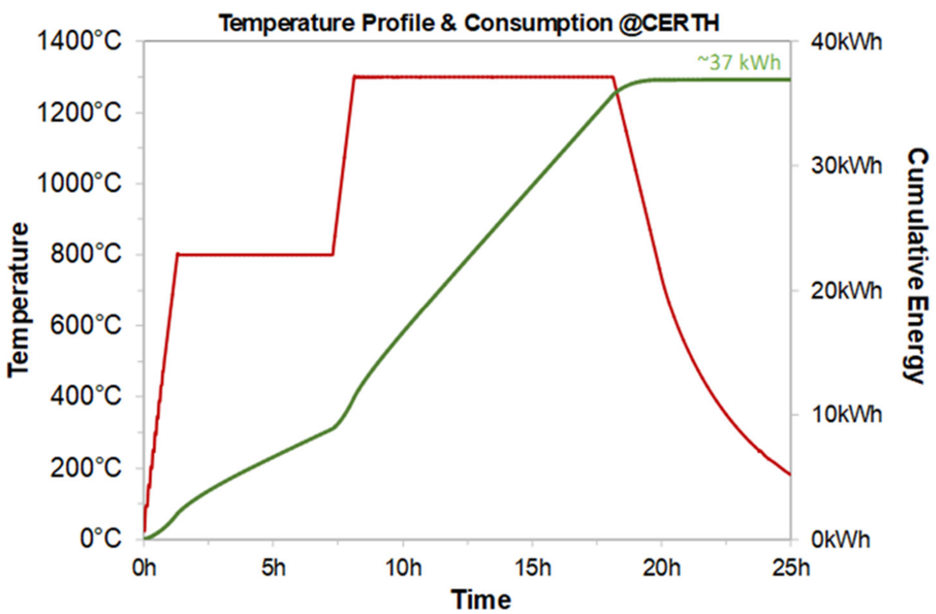

Figure 2: Temperature profile and energy consumption for the calcination during powder synthesis reaction (CERTH 2024)

The powder product after calcination is highly sintered, therefore it has to be milled for its further processing for the production of either honeycombs or foams.

For the honeycomb route, dry milling is currently performed at a 250 g scale and refers to a planetary mill (200 rpm for 180 min, 0.37 kW). For the lab max scenario, the same scale and equipment is assumed.

For the foam route, dry milling is currently performed at a 250 g scale and refers to a ball mill (380 rpm for 180 min, 0.74 kW). For the lab max scenario, a maximum batch size of 12 kg is considered. Electricity consumption is calculated based on these process parameters and scaled to the output 1kg milled reactants to have better comparability. The values are reported in Table 4.

Alternatively, in both routes, wet milling can be also performed; for the foams in the same ball mill above and for the honeycombs in other, in-house ball mill models. For this **wet**

milling process, water is added to the $CaMnO_3$ powder (50 – 62.5 % of solids by weight). Energy consumption is determined equivalent to the dry milling step. The use of wet milling however, necessitates a subsequent drying process to obtain the dried milled powder. For this subsequent **drying** process, with respect to the foams, electricity consumption is calculated based on process parameters provided by the project partner (drying at 100 °C, 450 W furnace for a maximum of 6–12 hours of drying). The drying step is also relevant for the honeycomb route and refers to a batch size of 0.2 kg in the current lab scenario and a batch size of 2 kg in the max lab scenario.

Table 4: LCI data for powder synthesis

		Lab current H	**Lab max H**	**Lab current F**	**Lab max F**	**Data source**	**Dataset**
Milling							
inputs	***unit***						
Reactants	kg/kg_{out}	1	1	1	1		
Electricity	kWh/kg_{out}	4.44	4.44	4.44	0.185	Calculated, based on process parameters	[3]
outputs							
Milled reactants	kg	1	1	1	1		
Firing/ Calcination							
inputs							
Milled reactants	kg/kg_{out}	1	1	1	1		
Furnace electricity consumption	kWh/kg_{out}	370.00	18.50	74.00	14.80	Calculated, based on process parameters	[3]
outputs							
CO_2 (fossil)	kg/kg_{out}	0.308	0.308	0.308	0.308	Stochiometric calculation	
$CaMnO_3$ powder	kg	1	1	1	1		
Wet milling							
inputs							
$CaMnO_3$ powder	kg/kg_{out}	0.5	0.5	0.5	0.5		
Electricity	kWh/kg_{out}	2.22	2.22	2.22	0.09	Calculated, based on process parameters	[3]
Tap water	l/kg_{out}	0.5	0.5	0.5	0.5	Primary data	[4]

outputs							
$CaMnO_{3,\ wet}$	kg	1	1	1	1		
Drying overnight							
inputs							
$CaMnO_3$, wet	kg/kg_{out}	2	2	2	2		
Electricity	kWh/kg_{out}	22.5	2.25	0.00	0.00	Calculated, based on process parameters	[3]
outputs							
$CaMnO_{3,\ dry}$	kg	1	1	1	1		
[3]	market for electricity, medium voltage | electricity, medium voltage | Cutoff, U - DE						
[4]	market for tap water- Europe without Switzerland-tap water | Cutoff, U - RER						

3.1.3 Production of porous monoliths

For the production of **honeycomb structures**, the dry $CaMnO_3$ is mixed with additives listed in Table 5 and a small quantity of water in in a Z-blade mixer to produce a relatively “stiff” paste (“dough”). Electricity consumption during this mixing is calculated based on provided process parameters (1.1 kW, 50/38 rpm, 4h) for a 1.5 l volume in the current lab scenario and for a 3.3 l volume in the max lab scenario.

In the foam route the dry $CaMnO_3$ is mixed with different additives and higher quantity of water in a glass-chamber mixer to produce a slurry. Electricity consumption during this mixing is calculated based on provided process parameters (0.43 kW, 1.5h) for a 0.125 l slurry volume in the current lab scenario and for a 1.5 l volume in the max lab scenario.

Regarding the used additives, significant differences exist between the two processing routes: For the paste for subsequent honeycomb production glycerine and hydroxypropyl methyl cellulose are used as additives, while polyvinylacetate and ethandiol are used in the slurry for the foam route. To relate data to a mass instead of volume flow, a density of 2.22 kg/l was determined for the slurry.

Also, regarding the subsequent process step, significant differences exist between the two routes: honeycomb production uses an extruder, while the foam production uses polyurethane (PU) foams as templates that are coated with the ceramic powder slurry.

For the **extrusion**, electricity consumption is calculated based on process parameters (5min/kg, 5kW). Since the **coating** process is currently performed manually at lab-scale, no energy is required.

After a drying step (air drying), the extruded or coated green body is sintered. Currently, cylindrical segments of approx. 26 mm diameter x 25 mm length are produced. So, the current lab scenario refers to 0.2 kg of final product for the honeycomb, and assumes a 2 kg batch for the max lab scenarios. For the foam processing the current lab scenario refers to 0.25kg and assumes a max batch of 3 kg.

The electricity consumption for the **sintering** including a debinding step is calculated based on the energy profile illustrated in Figure 3 (Sintering at 1200/1300 °C, 2hr). Emissions from the sintering step are calculated stoichiometrically. CO_2 emissions from combustion of methyl cellulose are inventoried as biobased CO_2 emissions. Emissions from the combustion of PU foam are calculated based on an incineration profile for PU foams (64% fossil carbon content, 6.1% nitrogen, 0.02% sulphur).

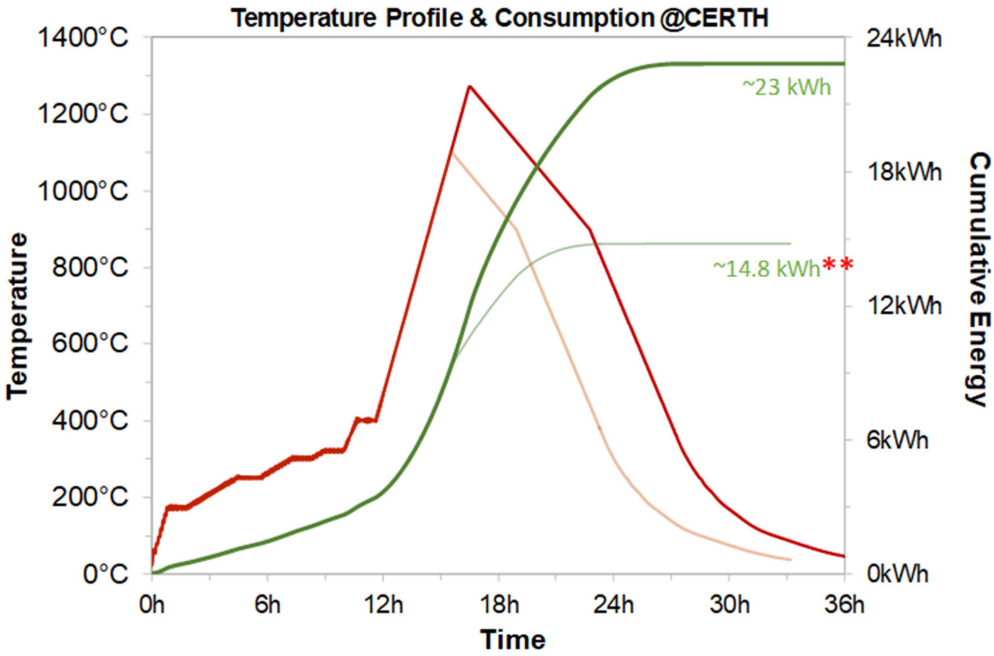

Figure 3: Temperature profile and energy consumption for the sintering (CERTH 2024).

For both process routes material losses of 2-3% are specified by the project partners. The model considers an average loss of 2.5%.

Table 5: LCI data for Processing of porous monoliths

		Lab current H	Lab max H	Lab current F	Lab max F	Data source	Dataset
Mixing							
inputs							
$CaMnO_3$, dry	kg/kg_{out}	0.799	0.799	0.777	0.777		
Tap water	kg/kg_{out}	0.100	0.100	0.200	0.200	Primary data	[4]
Glycerine	kg/kg_{out}	0.001	0.001			Primary data	[5]
Hydroxypropyl methyl cellulose	kg/kg_{out}	0.100	0.100			Primary data	own LCI dataset, based on (EPD International AB 2024)
Polyvinylacetate	kg/kg_{out}			0.015	0.015	Primary data	[6]

Ethandiol	kg/kg_{out}			0.0085	0.0085	Primary data	[7]
Electricity	kWh/kg_{out}	1.44	0.65	2.58	0.22	Calculated, based on process parameters	[3]
outputs							
Paste/slurry	kg	1	1	1	1		
Extrusion/Coating							
inputs							
Paste/slurry	kg/kg_{out}	1	1	1	1		
Electricity	kWh/kg_{out}	0.42	0.42	0.95	0.95	Calculated, based on process parameters	[3]
PU foams	kg/kg			0.05	0.05	Primary data	[8]
outputs							
Green body	kg	1	1	1	1		
Sintering (including debinding)							
Input							
Green body	kg/kg_{out}	1	1	1	1		
Electricity	kWh/kg_{out}	115.00	11.50	92.00	7.67	Calculated, based on process parameters	[3]
output							
CO_2 (biogenic)	kg/kg_{out}	0.350	0.350				
CO_2 (fossil)	kg/kg_{out}			0.117	0.117		[9]
NOx	kg/kg_{out}			1.38E-04	1.38E-04		[9]
SO_2	kg/kg_{out}			1.10E-07	1.38E-04		
Final product, w.o.losses	kg	1	1	1	1		[9]
[5]	market for glycerine | glycerine | Cutoff, U - RER						
[6]	market for ethylene vinyl acetate copolymer | ethylene vinyl acetate copolymer | Cutoff, U - RER						
[7]	market for ethylene glycol | ethylene glycol | Cutoff, U - RER						
[8]	market for polyurethane, flexible foam | polyurethane, flexible foam | Cutoff, U - RER						
[9]	Treatment of waste polyurethan, municipal incineration | Cutoff, U - Switzerland						

4. Impact assessment

4.1 LCI results

LCI results document all resources that are extracted and all emissions to air, soil and water that are emitted, directly and indirectly by all processes necessary for the production of studied structured material, here, 1kg of synthesized redox oxide materials in both honeycombs and foams. For the studied system, a list of around 250 resources and 2000 emissions is compiled that is not presented here in detail. Table 6 shows an extract of selected resources and emissions.

Table 6: Selected LCI data (resources and emissions)

	Unit/kg	Lab current H	Lab max H	Lab current F	Lab max F
Calcium	kg	1.23E-03	1.31E-04	4.67E-04	9.24E-05
Coal, brown	kg	106.535	8.810	38.173	4.939
Coal, hard	kg	24.684	2.302	9.004	1.392
Gas, natural	m3	18.745	1.77	6.86	1.08
Manganese	kg	0.510	0.49	0.49	0.48
Crude oil	kg	1.783	0.23	0.73	0.20
Carbon dioxide, fossil	kg	185.620	16.96	67.63	10.28
Methane, fossil	kg	0.04	0.03	0.13	0.02
Calcium	kg	0.001	0.000	0.000	0.000

4.2 Impact assessment results

4.2.1 Impact overview

Table 7 shows the impact assessment results for the single, midpoint and endpoint impacts for the four scenarios. Results are related to 1kg of synthesized redox oxide materials in either honeycomb (H) or foams (F) form. Single point results, climate change and resource use are highlighted as they are especially relevant indicators for the project.

Table 7: Impact assessment results- overview

	Unit/kg	Lab current H	Lab max H	Lab current F	Lab max F
Single point (normalised endpoint)	**points**	1449.64	131.37	526.53	78.22
Damage to human health	DALY	4.15E-04	3.91E-05	1.52E-04	2.36E-05
Damage to ecosystems	species.yr	9.29E-07	9.56E-08	3.34E-07	5.03E-08
Damage to resource availability	USD2013	7.37	0.73	2.74	0.48
Acidification: terrestrial	kg SO2-Eq	0.28	0.03	0.10	0.02
Climate change	**kg CO_2-Eq**	**204.40**	**18.82**	**74.45**	**11.34**
Ecotoxicity: freshwater	kg 1.4-DCB-Eq	10.93	0.97	3.96	0.58
Ecotoxicity: marine	kg 1.4-DCB-Eq	15.17	1.35	5.50	0.80
Ecotoxicity: terrestrial	kg 1.4-DCB-Eq	795.14	76.24	293.50	49.02
Energy resources: non-renewable. fossil	kg oil-Eq	51.53	4.64	18.74	2.79
Eutrophication: freshwater	kg P-Eq	0.27	0.02	0.10	0.01
Eutrophication: marine	kg N-Eq	0.02	0.00	0.01	0.00
Human toxicity: carcinogenic	kg 1.4-DCB-Eq	27.48	2.53	10.03	1.54
Human toxicity: non-carcinogenic	kg 1.4-DCB-Eq	306.13	26.57	110.65	15.58
Ionising radiation	kBq Co-60-Eq	49.98	4.23	17.97	2.41
Land use	m2*a crop-Eq	5.53	1.58	1.59	0.24
Material resources: metals/minerals	**kg Cu-Eq**	**1.16**	**0.18**	**0.45**	**0.12**
Ozone depletion	kg CFC-11-Eq	0.00	0.00	0.00	0.00
Particulate matter formation	kg PM2.5-Eq	0.10	0.01	0.04	0.01
Photochemical oxidant formation: human health	kg NOx-Eq	0.22	0.02	0.08	0.01
Photochemical oxidant formation: terrestrial ecosystems	kg NOx-Eq	0.23	0.02	0.08	0.01
Water use	m3	0.97	0.10	0.36	0.07

Selected results for the four scenarios at single point and endpoint level are also presented in Figure 4. The trends are very similar for the single point results as well as for the three endpoint categories. Comparing the H and F route in the current lab conditions, shows lower results for the F route which is mainly due to the slightly higher production scales in the lab where F is produced. The comparison of the lab max conditions between both routes shows a lower difference between both routes, still with advantages for the foam route. A comparison of all scenarios shows how significantly impacts can be reduced when production conditions (from current to max lab performance) change.

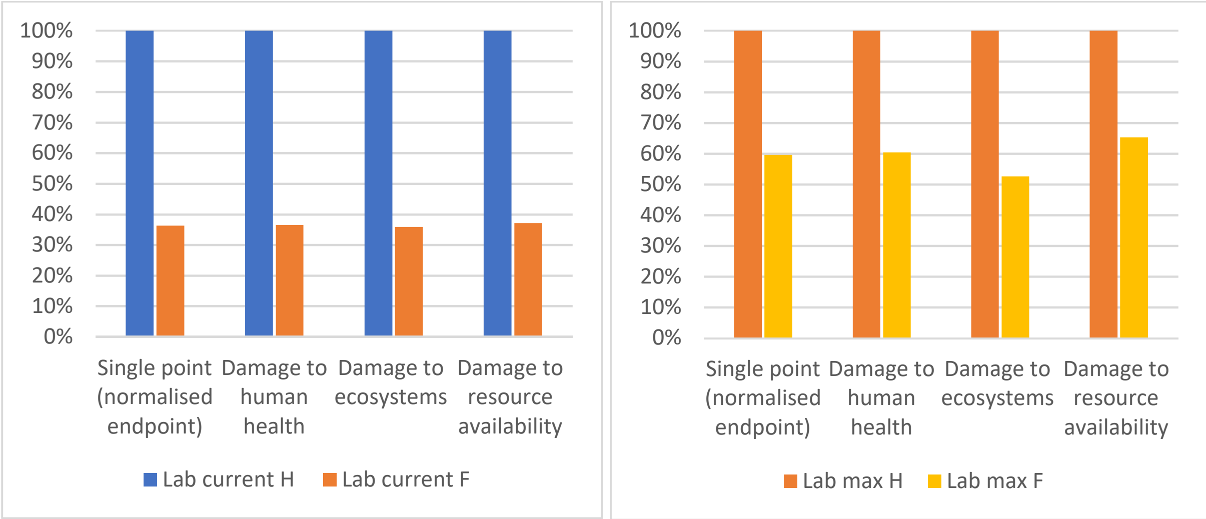

Figure 4: Scenario comparison at single point and endpoint level

4.2.2 Contribution analyses

Figure 5 and Figure 6 show the contribution analyses for climate change and resource use in the four scenarios. Figure 5 displays results, aggregated into raw material provision, powder synthesis and the processing of the porous monoliths. For **honeycomb** production, the results are mainly dominated by the powder synthesis step, while for **foam production** the processing step has the main contribution. The comparison of the two scenarios shows that once the process is more optimised regarding consumption of energy, the impact of raw material selection becomes more relevant. This is given for the impact resource use, which is mainly dominated by the manganese oxide production.

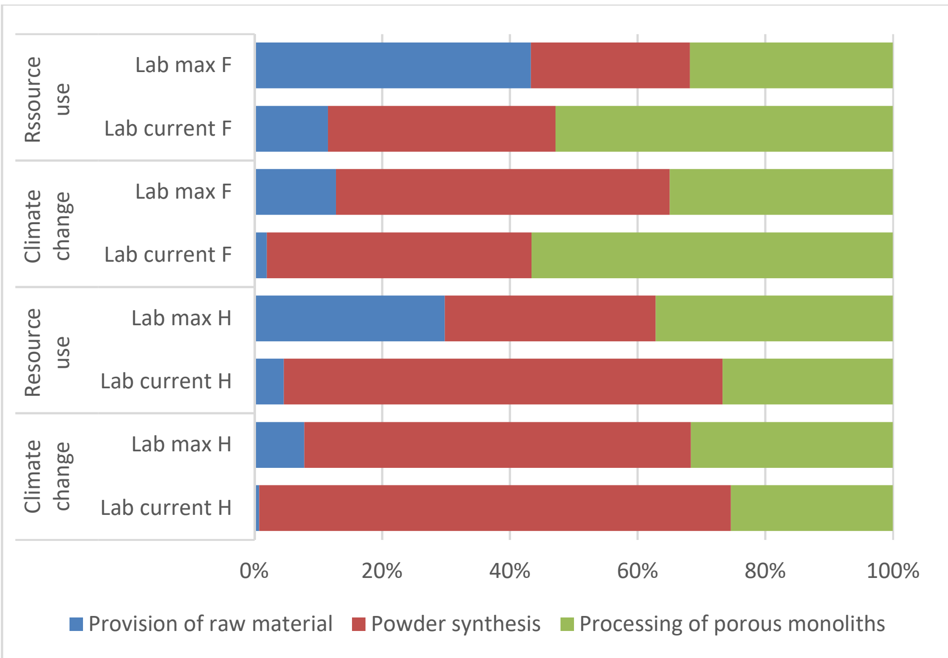

Figure 5: Contribution analysis for climate change and resource use in the four scenarios

Figure 6 shows in more detail the process contribution in the powder synthesis and processing steps.

For **honeycomb** production, the contribution results identify the firing/calcination process as the most contributing. This is due to high energy input needed in the firing process and the comparatively low batch sizes used. The second most relevant process is the sintering process. In the best lab scenario and for resource use, the raw material provision and the mixing process have significant contributions. The impact of the mixing process mainly comes from the use of hydroxypropyl methyl cellulose and more specifically from sodium chloride powder that is used in the production of methyl cellulose.

Foam production is dominated by the firing/calcination and by the sintering process. Climate change impacts are highly dominated by direct process emissions that occur as CO_2 emissions in the firing/calcination as well as in the sintering process, where the PU foams are burned with associated emissions (see 3.1.3). In the max lab scenario, the contribution shifts to more significant contributions from raw material provisions and the coating (use of polyurethane) in the category resource use.

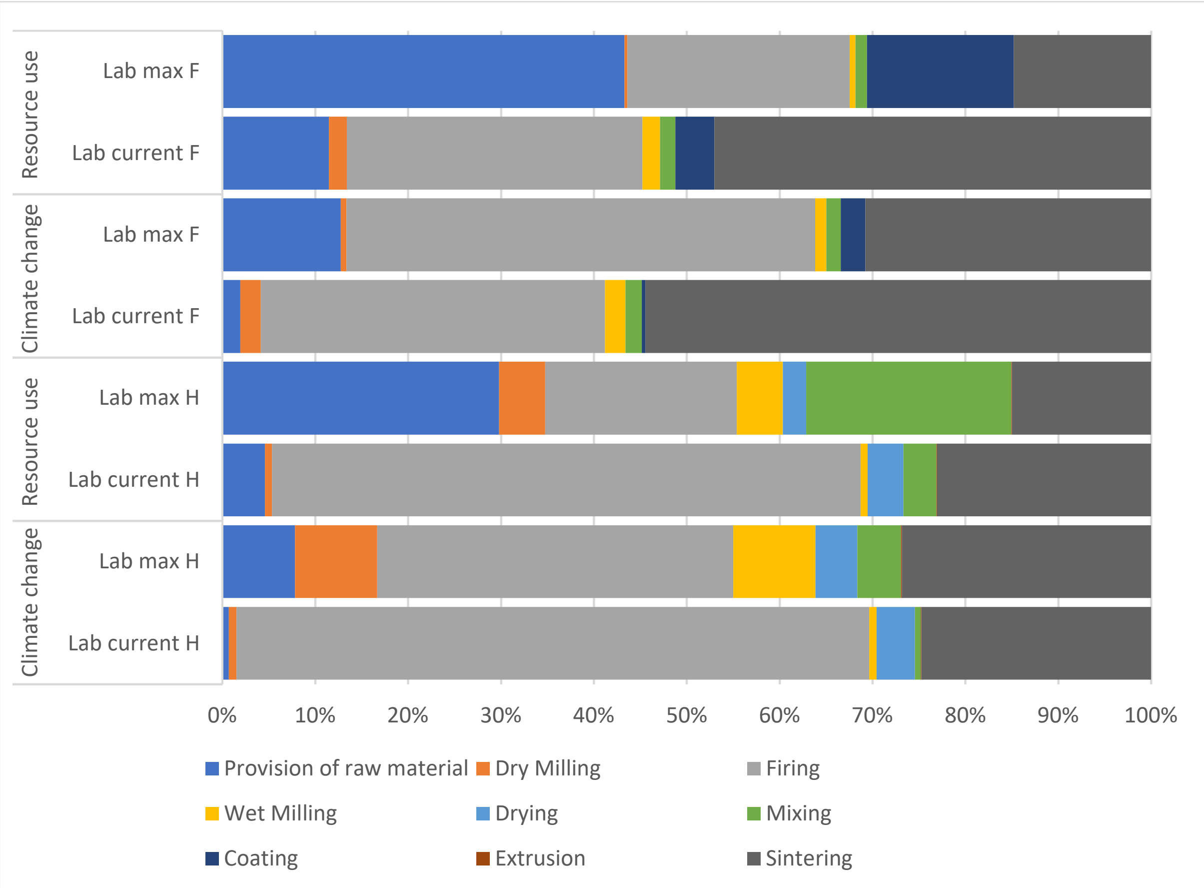

Figure 6: Process contribution analysis for climate change and resource use in the four scenarios

4.3 Sensitivity analysis

The comparison between the two scenarios has identified the used batch sizes and related to that the energy efficiency of the processes as a highly sensitive parameter. Other sensitive parameters are the impact intensity of used electricity, the amount and impact intensity of the used raw materials as well as direct emissions in the calcination and sintering processes. While the latter is calculated based on stochiometric coefficients and can be considered as a fixed value, the other input parameters can be varied in a sensitivity analysis to demonstrate their influence on results.

In the first sensitivity calculation, a harmonised lab max scenario is created where the energy efficiency of the lab equipment is assumed to be equally high in both production routes. The values for the electricity use for the milling as well as for the calcination and sintering processes are taken from the lab max scenario from the foam production route.

In the second sensitivity the harmonised lab max scenario is coupled with a European energy mix instead of the German one.

Figure 7 shows the results of the sensitivity analysis. The results show a significant reduction of the GWP of the honeycomb route, from 18.83 to 12.50 kgCO_2-eq./kg. Thus, in terms of GWP, the difference between the two processing routes is below 10%. The change from German to European market electricity mix has a significant influence on the GWP of both routes. Both results can be improved by around 19%.

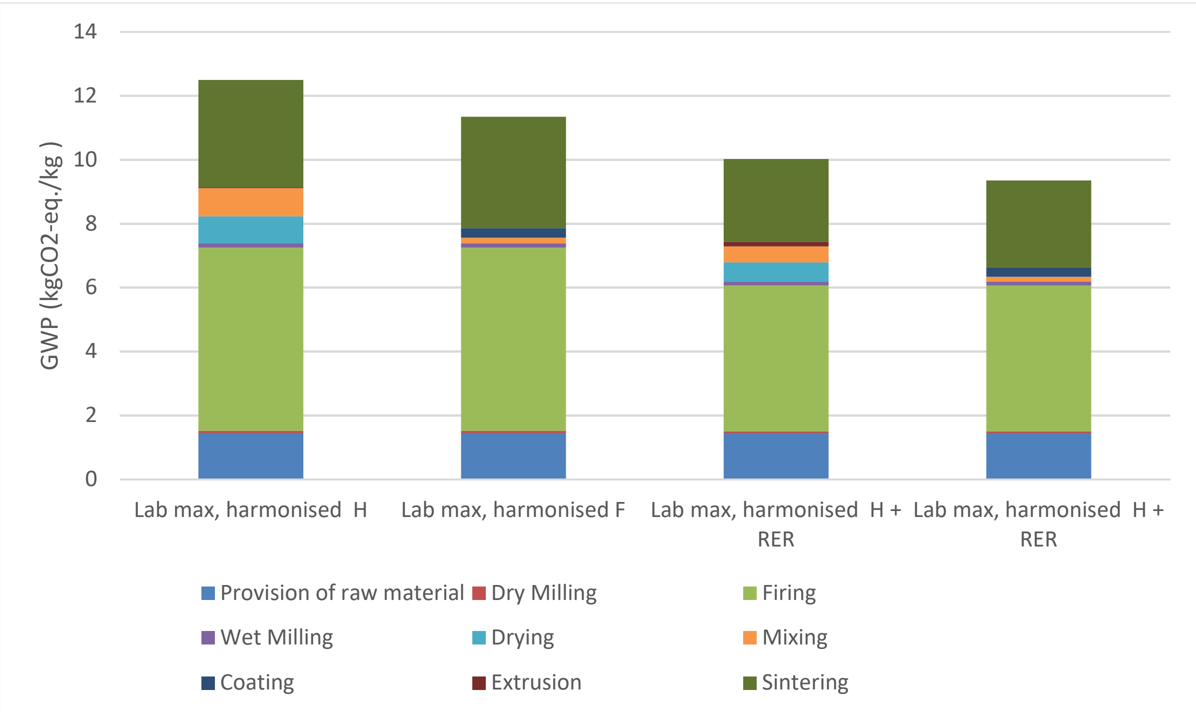

Figure 7: Sensitivity analysis for climate change

5. Interpretation

5.1 Discussion of results

This deliverable has compiled LCA results for innovative synthesised redox oxide-based materials applicable for a TCES systems. As presented in the state of the art, very few studies have so far been performed for such systems and LCA results for redox oxide materials tailored to high-temperature TCES systems for CSP applications are completely missing. Therefore, it is not possible to directly compare results and an additional step is needed to assess environmental performances for the CSP system with integrated TCES. Studies that integrated a TCES system (Pelay et al. 2020; Colelli et al. 2022) show favourable environmental performances, but also identified the TCES system as a contributor to environmental burdens. To be able to assess TCES system, it is needed to have reliable storage performance data over the life time of components which is not yet given for the innovative synthesized redox oxide materials that are currently under development.

For the interpretation of results, it has to be further noticed that the modelled processes and products have not yet been optimised from environmental or economic perspective and it is to be expected that environmental impacts (as well as production cost) are comparatively high, especially when operated at very small batch scale for experimental purpose. This finding has been observed in several prospective LCA studies that assess environmental impacts at low TRL (Thonemann et al. 2020) and is also the case for this study.

The deliverable identified the processes contributing the most to different impact categories for the lab scale production of an innovative synthesized redox oxide materials, shaped into structures suitable to be accommodated in a practical high temperature heat storage reactor/heat exchanger. The focus was set on the three impacts categories: the single score, mineral resource depletion and global warming potential. The results for the current lab scenario are highly dominated by the calcination and sintering step. This is due to the significant electricity consumption in the process and to a much lower extent, due to direct (process) CO_2 emissions. The best lab scenario provides already an indication that energy consumption and associated environmental impacts can be significantly reduced for both the honeycomb and foam route processing routes, especially in terms of resource use. This is not the case for the direct CO_2 emissions that remain equally appreciable in both scenarios. The results also indicate that for more optimised production conditions at higher TRL, the raw material provision will be a relevant parameter in terms of resource use. In this regard it has to be discussed whether raw material consumption can be reduced through a potential remanufacturing (closed loop recycling route).

Regarding consistency of results, it can be stated that results for the four scenarios are highly consistent since they are based on identical system boundaries and they represent an equally high data quality. Most LCI data are provided by project partners and can be qualified as high-quality primary data. However, data represent small scale experiments with associated uncertainty. In order to compile reference values for the scenarios, averaged process condition had to be estimated.

In terms of data completeness, the study represents a cradle to gate LCA, not including the use and EOL phase. The study included energy consumption and other consumables such as additives, but the laboratory equipment itself (the machine, the infrastructure or the buildings) were not included. Regarding direct emissions, the study represents a rather rough estimation, considering CO_2, SO_x and NO_2, based on C, S and N contents of the burned additives.

Scenario and sensitivity analysis were performed that demonstrated high variability and sensitivity of results. However, the trend between scenarios and processing routes was robust and can provide a solid basis and first recommendations regarding environmentally friendly materials selection and processing.

5.2 Recommendations

- The foam route with the lab max scenario demonstrates higher environmental performance mainly due to the higher batch sizes in the milling, calcination, and sintering processes. Results from the sensitivity analysis show that the difference in terms of GWP between the two processing routes is below 10% when lab conditions are more harmonised. Thus, it is recommended to further exploit optimisations potentials in terms of energy efficiency (batch sizes/ equipment) in both routes, to design more harmonised processes that also represent higher TRL and to perform prospective assessment to exploit the future environmental potential.
- The lab max scenario demonstrates the increased importance of the material and additives inputs once energy optimisations are established. It is recommended to further investigate more material combinations as well as the anticipated doping. It is also recommended to further exploit remanufacturing options in terms of the additional impacts through remanufacturing, but also in terms of reduction of resource use through a closed loop processing.
- In order to determine environmental impacts for a CSP plant with the integrated innovative TCES system, it is needed to further exploit the link between material performance and long-term energy storage performance.

6. References

Cai, Runxia; Bektas, Hilal; Wang, Xijun; McClintock, Kyle; Teague, Lauren; Yang, Kunran; Li, Fanxing (2023): Accelerated Perovskite Oxide Development for Thermochemical Energy Storage by a High-Throughput Combinatorial Approach. In *Advanced Energy Materials* 13 (18), Article 2203833. DOI: 10.1002/aenm.202203833.

Chen, Xiaoyi; Zhang, Zhen; Qi, Chonggang; Ling, Xiang; Peng, Hao (2018): State of the art on the high-temperature thermochemical energy storage systems. In *Energy Conversion and Management* 177, pp. 792–815. DOI: 10.1016/j.enconman.2018.10.011.

Colelli, G.; Chacartegui, R.; Ortiz, C.; Carro, A.; Arena, A. P.; Verda, V. (2022): Life cycle and environmental assessment of calcium looping (CaL) in solar thermochemical energy storage. In *Energy Conversion and Management* 257, p. 115428. DOI: 10.1016/j.enconman.2022.115428.

Dizaji, Hossein Beidaghy; Hosseini, Hannaneh (2018): A review of material screening in pure and mixed-metal oxide thermochemical energy storage (TCES) systems for concentrated solar power (CSP) applications. In *Renewable and Sustainable Energy Reviews* 98, pp. 9–26. DOI: 10.1016/j.rser.2018.09.004.

EPD International AB (Ed.) (2024): Environmental Product Declaration for CELLULOSE ETHER. from Shandong Eton New Material Co. Ltd. (S-P-13216). Available online at https://api.environdec.com/api/v1/EPDLibrary/Files/82fb7647-d5e2-447b-cfdd-08dc5acebd38/Data.

Frischknecht, Rolf; Jungbluth, Niels; Althaus, Hans-Jörg; Doka, Gabor; Dones, Roberto; Heck, Thomas et al. (2005): The ecoinvent Database: Overview and Methodological Framework (7 pp). In *Int J Life Cycle Assess* 10 (1), pp. 3–9. DOI: 10.1065/lca2004.10.181.1.

Hayatina, Isye; Auckaili, Amar; Farid, Mohammed (2023): Review on the Life Cycle Assessment of Thermal Energy Storage Used in Building Applications. In *Energies* 16 (3), p. 1170. DOI: 10.3390/en16031170.

Huijbregts, Mark A. J.; Steinmann, Zoran J. N.; Elshout, Pieter M. F.; Stam, Gea; Verones, Francesca; Vieira, Marisa et al. (2017): ReCiPe2016: a harmonised life cycle impact assessment method at midpoint and endpoint level. In *Int J Life Cycle Assess* 22 (2), pp. 138–147. DOI: 10.1007/s11367-016-1246-y.

Karagiannakis, G.; Pagkoura, C.; Zygogianni, A.; Lorentzou, S.; Konstandopoulos, A. G. (2014): Monolithic Ceramic Redox Materials for Thermochemical Heat Storage Applications in CSP Plants. In *Energy Procedia* 49, pp. 820–829. DOI: 10.1016/j.egypro.2014.03.089.

Lamnatou, Chr.; Chemisana, D. (2017): Concentrating solar systems: Life Cycle Assessment (LCA) and environmental issues. In *Renewable and Sustainable Energy Reviews* 78, pp. 916–932. DOI: 10.1016/j.rser.2017.04.065.

Nienborg, Björn; Gschwander, Stefan; Munz, Gunther; Fröhlich, Dominik; Helling, Tobias; Horn, Rafael et al. (2018): Life Cycle Assessment of thermal energy storage materials and components. In *Energy Procedia* 155, pp. 111–120. DOI: 10.1016/j.egypro.2018.11.063.

Nuss, Philip; Eckelman, Matthew J. (2014): Life cycle assessment of metals: a scientific synthesis. In *PloS one* 9 (7), e101298. DOI: 10.1371/journal.pone.0101298.

Ordoñez, Marcela Frias; Biella, Serena; Falletta, Ermelinda; Bianchi, Claudia L. (2023): Paving the green path: A novel approach to material manufacturing using LCA. In *Chemical Engineering Journal Advances* 16, p. 100575. DOI: 10.1016/j.ceja.2023.100575.

Pelay, Ugo; Azzaro-Pantel, Catherine; Fan, Yilin; Luo, Lingai (2020): Life cycle assessment of thermochemical energy storage integration concepts for a concentrating solar power plant. In *Env Prog and Sustain Energy* 39 (4), Article e13388. DOI: 10.1002/ep.13388.

Thonemann, Nils; Schulte, Anna; Maga, Daniel (2020): How to Conduct Prospective Life Cycle Assessment for Emerging Technologies? A Systematic Review and Methodological Guidance. In *Sustainability* 12 (3), p. 1192. DOI: 10.3390/su12031192.

Weidema, Bo; Bauer, Christian; Hischier, Roland (2013): Data quality guideline for the ecoinvent database version 3. [R]. In *Overview and Methodology Ecoinvent. Report 1 St. Gallen: The Ecoinvent Centre*, pp. 76–84.

7. Supplement

Table S 1 Comparison of selected impact categories for the short-listed raw materials.

Raw materials		Single score (1)	Damage to resource availability (2)	Global warming potential (3)
		points/FU	USD 2013/FU	CO2-eq./FU
BaO	Barium oxide	7.86E-04	0.14	2.51
$CaCO_3$	Calcium carbonate	4.14E-05	0.04	0.39
CO_3O_4	Cobalt oxide	1.34E-02	8.58	0.92
CuO	Copper(II) oxide	7.89E-03	0.68	6.12
Fe_3O_4	Iron(II.III) oxide (Iron ore)	5.43E-06	0.01	0.01
	Iron(II.III) oxide (magnetite)	1.16E-04	0.06	0.92
La2(CO3)3	Lanthanite-(La) carbonate	6.65E-03	7.63	41.10
Mn3O4	Manganese(II) oxide	1.40E-02	0.29	5.52
Nd_2O_3	Neodymium oxide	8.68E-03	10.90	54.60
NiO	Nickel(II) oxide (Nickel concentrate)	2.45E-04	0.22	0.66
	Nickel(II) oxide (Electrolyte)	8.64E-04	0.83	3.76
$SrCO_3$	Strontium carbonate	2.95E-04	0.14	2.39
Ti	Titanium	7.63E-03	2.55	48.20