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Energy analysis and life cycle assessment of a thermal energy storage unit involving conventional or recycled storage materials and devoted to industrial waste heat valorisation

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A B S T R A C T

Any system intending to improve the environmental performances of a process should be assessed by a Life Cycle Assessment. This work draws up the environmental profile of the heat provided by a storage system recovering industrial waste heat at high temperature (500 °C) through 5 selected indicators: Cumulative Energy Demand, Global Warming Potential, abiotic depletion potential, particle matter and freshwater eutrophication. The calculated indicators were compared to those of the fossil fuel substituted by the recovered heat, that is to say natural gas, and proved to be reduced. Then, the environmental payback times were calculated; and an energetic profitability evaluation by the Energy Returned on Investment expanded on the profile. Besides, the influence of operating conditions variations was investigated through a parametric study. An optimal number of cycles to provide the same amount of energy may be defined, as a compromise between the abiotic resource depletion potential reduction with smaller tanks performing more cycles and the energy returned on investment deteriorated with the operational energy consumption increased due to higher pressure drop with smaller tank diameter. The most rewarding environmental performances concerned the Cumulative Energy Demand, Global Warming Potential indicators of the provided heat, about 1.2 kgCO₂-eq and 65 MJ-eq per kWh (payback time lower than 3 months), and the Energy Returned on Investment that doubled compared to natural gas, e.g. reached a value of 55. The positive effect of using recycled storage materials on the resource depletion indicator were enlightened, notably when the operating conditions strayed from the reference case.

Keywords:

Waste heat valorisation

Thermal energy storage (TES)

Life cycle assessment (LCA)

Energy returned on investment (EROI)

Recycled thermal energy storage materials

1. 1. Introduction

The European industry consumes about 23% of the final energy demand, that is to say ~3000 TWh.year⁻¹, mostly as fossil fuels coming from importation at 70% (Eurostat, 2019). More than half of this energy is consumed as heat (70% worldwide) and lost in processes up to 50%: globally, this industrial energy use emits 33% of greenhouse gases (Jouhara and Olabi, 2018). A low-impact and low-cost heat could be supplied by recovering that waste heat (E and Excess Heat. 2015., 2015), responding jointly to the increasingly stringent legislation targeting a massive greenhouse gas emissions reduction; to the sector competitiveness issue; and to the energetic security improvement. A waste heat

stream is firstly characterized by its temperature level: the 200–600 °C rank represents about one third of the 300 TWh.year⁻¹ potential estimated in EU28, mainly loose in Germany, France and Italy (Papapetrou et al., 2018); it is thus a relevant rank to address. That waste heat is mainly lost through the exhaust fumes, which recovering have been notably identified as a best available technique to improve sustainability in the ceramic industry sector (Ibáñez-Forés et al., 2013).

According to the US Department of Energy (DOE), a barrier to a massive heat recovery is the lack of end uses that should be enlarged by introducing efficient heat storage solutions (Department of Energy, 2008): the thermal energy storage systems solves the issue in coinciding the energy supply and demand. Their wide applications have been reviewed by Miro et al. (Miró et al., 2016), who pointed the lack of pilot

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Nomenclature

TES	Thermal Energy Storage
PCM	Phase Change Material
MFA	Material Flow Analysis
CFA	Coal Fly Ashes
EROI	Energy Returned On (energy) Invested
PBT	Pay Back Time years
CED	Cumulative Energy Demand MJ-eq
GWP	Global Warming Potential kgCO ₂ -eq
ADP	Abiotic Depletion Potential kgSb-eq
PM	Particle Matter kgPM _{2.5} -eq
FE	Freshwater eutrophication kgPO ₄ ²⁻ -eq
L	Length m
D	Diameter m
H _{slot}	Slot duration (charge and discharge) Hour per day
N _{cycles}	Number of cycles Cycle per year
T _{max}	Storage temperature °C
ε	Storage bed porosity

and real scales solutions and detailed environmental profiles. Indeed, when it comes to propose new renewable energy or energy efficiency systems, one should quantify and verify the carried environmental benefits, as they are not always consistent with first intuitions (Suh et al., 2016). That may be done by a Life Cycle Assessment (LCA), a normalized method (ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b) evaluating the quantifiable effects of products or services on the environment through their whole life cycle (from cradle to grave or cradle to cradle in circular economy) (Azapagic, 1999).

Such studies on storage system have been more widely undertaken in the field of concentrated solar power (CSP) than waste heat recovery (Koçak et al., 2020), while those two domains lies on similar storage technologies in the focused temperature range (200–600 °C), namely sensible heat storage on rocks or ceramics (Ortega-Fernández and Rodríguez-Aseguinolaza, 2019). The LCA framework and data inventory on CSP systems were firstly detailed by Heath et al. (2009) and Oró et al. (2012) brought a global approach on TES systems LCA comparison. Further work detailed studies on heat storage systems containing waste (Lalau et al., 2016) or natural (Nahhas et al., 2020) TES material. Concerning the industrial waste heat domain, the previous work is scarce and mainly focused on latent heat storage, such as López-Sabirón et al. who found that up to half the life cycle of a heat exchanger filled with phase change materials (PCM) was needed to payback its life cycle impacts by avoiding fossil fuel consumption (López-Sabirón et al., 2014a) or that some PCM may not be able to balance the TES system carbon footprint (López-Sabirón et al., 2014b).

Besides, most of those studies focused on two main indicators – the global warming potential, GWP, and the cumulative energy demand, CED – that may lead to missing an important environmental burden in another category such as ozone depletion or particle matter (Hiremath et al., 2015). For instance, Miro et al. compared the embodied energy of three types of thermal storage based on concrete, molten salts or phase change materials and concluded from that information that the first one was the most environmentally efficient (Miró et al., 2014). Another work including eleven impact indicators founded that molten salts storage was less detrimental than concrete one (Adeoye et al., 2014). Hence, various relevant indicators should be considered, notably the resource depletion that is often pointed to be an issue when trying to substitute conventional fossil fuels by renewable or recovered energy: Davidsson studied the potential conflict between some wind and solar photovoltaic targeted growth rates and the resource flow potential of some materials notably in lithium-ion batteries (Davidsson, 2016); Phil et al. founded that nitrate salts and silver could face supply shortage and

increased prices in a CSP deployment following the International Energy Agency (IEA) roadmap (Pihl et al., 2012); Vidal et al. underlined the scarcity risk of basic element such as cement, steel, aluminium and copper to meet the carbon neutral target in 2050 (Vidal et al., 2018). A recent study proposed a combined methodology mixing Material Flow Analysis (MFA) and Life Cycle Assessment (LCA) that recommended the replacement of the molten salts by a more environmentally friendly TES material to enhance the system compliance with circular economy (Abokershet et al., 2021).

As well, those systems may be charged to be less energy-efficient than fossil fuels in terms of net energy supplied to society (Capellán-Pérez et al., 2019), so that should be quantified by the Energy Return on (energy) Investment (EROI) ratio (Walmsley et al., 2018). The combination of LCA and net energy analysis should bring a new exhaustive vision on energy systems sustainability, such as in Raugei et al. work on the electricity supply system in Chile (Raugei et al., 2018) and in UK (Raugei et al., 2020).

This work aims at evaluating the environmental impacts of the heat supplied by thermocline thermal storage tank using air as heat transfer fluid and ceramics as heat storage media. This system recovers the heat wasted by industrial furnaces (ceramic production, foundries, etc.) and reuses it on demand: the valorised heat substitutes a fraction of fossil fuel consumption (generally natural gas), avoiding the associated impacts. Besides, the thermal storage ceramics may be elaborated from industrial wastes such as the coal fly ashes (CFA) coming from coal fired power plants that are also energetic processes involving heat. The relevance of using wastes as secondary raw material for sensible storage have been established in previous study, studying the TES material elaboration from asbestos containing wastes (Pyet et al., 2011), fly ashes (Lin et al., 2019), bottom ashes from municipal waste incinerator (Lopez Ferberet et al., 2020), or other industrial wastes (Gutierrez et al., 2016) and their durability in use conditions (Xu et al., 2018). Coal fly ashes is an abundant waste that still not equally valorised across countries: for instance, only 67% are reused in China (Tang et al., 2013), where the industry sector stands for 86% of the final energy demand (Wang et al., 2020) – in such context, the proposed system could advantageously bring a double waste material and heat valorisation in a circular economy thinking.

Thereby, the following analysis will evaluate the environmental benefits provided through the substitution of a fossil energy by an energy recovered with an industrial heat storage system named Eco-Stock®, containing conventional or waste-based ceramics. This evaluation will be based on a double approach of life cycle assessment and net energy analysis.

2. Material and method

2.1. LCA, PBT & EROI

2.1.1. Life cycle assessment (LCA)

The Life Cycle Assessment (LCA) consists in establishing a balance between production of emissions and wastes and consumption of resources and energy (ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b)). These fluxes are aggregated to quantify the system effects by impact indicators. This step relies on referenced methods; the one used in this study was the ILCD 2016 mid-point method as recommended by the European Commission (European Commission - Joi, 2010). The database was EcoInvent v3.5 (Ecoinvent 3.5 database, 2021), and the software OpenLCA v1.8 (OpenLCA software, 2021).

When studying a system providing energy, the analysis should include a calculation of its energy payback time (EPBT) and its energy returned on investment (EROI), as defined below.

2.1.2. Payback time (PBT)

Along its lifetime, a system acquires an energetic debt calculated by its cumulative energy demand (CED), also called a primary energy

content. If the system saves energy by decreasing a fossil energy consumption, it should payback this energy debt in a time defined by Equation (1). The CED_{Total} is the cumulative energy demand from the whole system lifetime, while the $CED_{Utilization}$ only concerns the operating period.

$$EPBT = \frac{CED_{Total} - CED_{Utilization}}{CED_{MWh\ Gas} \times MWh_{saved/year} - \frac{CED_{Utilization}}{Lifetime}} \quad (1)$$

In this study, the produced heat substitutes natural gas, thus the conventional CED is the one of gas burned in an industrial furnace. The payback time may be calculated for any avoided impact, such as global warming potential (GWP): the CED indicator in Equation (1) is replaced by the suitable indicator, as described by Equation (2) where X stands for the considered indicator.

$$X - PBT = \frac{X_{Total} - X_{Utilization}}{X_{MWh\ Gas} \times MWh_{saved/year} - \frac{X_{Utilization}}{Lifetime}} \quad (2)$$

2.1.3. Energy returned on (energy) investment (EROI)

This EROI describes the effort developed to bring the energy to the consumer: it is the ratio between the produced energy and the energy consumed in delivery (Murphy and Hall, 2010).

$$EROI = \frac{MWh\ provided}{CED_{MWh} - CED_{Resource}} \quad (3)$$

The energy consumed does not include the resource energy content, unlike the CED indicator in LCA: different boundaries are considered (Fig. 1) (Arvesen and Hertwich, 2015), but the LCA methodology within that perimeter is still followed (Murphy et al., 2016). That indicator is at the boarder of energy, economy and social science (Fischer-Kowalski et al., 2019): it quantifies the effort needed by a society to capture and deliver the available energetic resources. That counting makes no difference between stock (fossil) and flux (renewable) resources, giving no information on resource depletion nor pollution: it should be considered jointly with LCA indicators. Its benefit lies in the system viability evaluation: if the delivered energy is too small compared to the invested energy for delivery, then the system is not profitable.

A simple example may illustrate that principle (schematized on Fig. 2): “the oil has an EROI of 100” could be traduced by “one oil barrel must be spent to bring 100 barrels to the consumers”. Likewise, an EROI of 2 means that one barrel is spend to deliver 2 to the consumers. In the first case, 99% of the energy extracted is returned and only 50% in the second case.

The share of energy returned does not decrease linearly with the EROI: above ~ 20 an EROI variation will barely be noticeable on energy returned, but will be critical below ~ 8 – that is the “energy cliff” under which a system will not be viable (Murphy and Hall, 2010). Indeed, a margin should be taken to cover the energy needs not enclosed in the perimeter (transport or cooking for the factory workers for instance).

2.2. Studied system & functional unit

The studied system is a couple of 950 kWh thermal energy storage

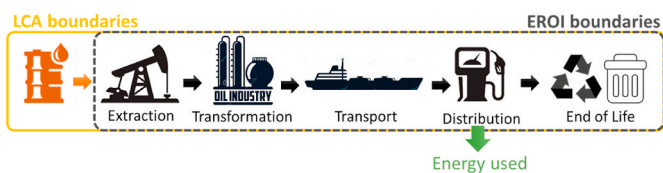


Fig. 1. Difference in LCA and EROI boundaries – Oil example (from (Lambert et al., 2012)).

thermocline tanks, filled with granular ceramic as storage material. The two tanks may charge and discharge simultaneously or not, allowing more monitoring flexibility than a single tank. They recover heat from industrial furnace by directly flowing hot fumes through the packed bed, store the heat in the filler ceramics, and return 500 °C heat to a drying process on demand at a 475 kW power. Considering a small-medium factory producing 11 500 t of tiles per year at a mean firing consumption of 2500 kJ kg⁻¹ (European Commission, 2007), the yearly energy consumption would be around 8 GWh. Thus, considering 35% heat losses in the flue gases (E and Excess Heat. 2015., 2015), the valorisation potential would be about 2.8 GWh per year. The storage system has an 80% charging efficiency (accounting for the ducts thermal losses and the low-temperature waste heat fraction that is not recovered) and 92% discharging (accounting for the ducts thermal losses) (Touzo et al., 2020). The two tanks operate 3 charge/discharge cycles per 24 h h 7/7days with a two weeks plant closure, totalizing 1050 cycles per year that is to say 2 GWh year⁻¹ valorised. Along its 25 years lifetime, 50 000 MWh will be provided from waste heat.

That study aims at quantifying the environmental benefit brought by the Eco-Stock® storage system integration. Thus, the first step is to quantify the solution environmental impacts, and then compare them with a “business as usual” scenario that would be satisfying to the plant heat demand with fossil fuel only (gas burner). A fair comparison requires a same functional unit to which will refer all calculated impacts. The functional unit describes the service provided during a given period, in the present case that could be:

« Provide 1 MW_{th} at the rate of 2 GWh year⁻¹ during 25 years »

The reference flux is thus the provided MWh of heat. One MWh from the storage system will substitutes to one MWh of natural gas burned in industrial furnace.

The results will be enlarged by looking at the following points:

- the error margin from technical variables, as efficiency or lifetime, will be assessed by a sensitivity study;
- two types of filler ceramic material (conventional or made from wastes) will be compared;
- the influence of operating conditions such as the number of cycles or the valorisation temperature will be studied through different scenarios.

2.3. Boundaries

This LCA includes all life cycle steps, from raw material extraction to end-of-life management (Fig. 3). Some materials such as steel contains a part of recycled material: the recycling process is included in the perimeter, but the wastes are considered at the recycling plant gate.

The production step (raw materials + elaboration) includes the materials and systems composition, their transportation to production site, the equipment and energy used for the elaboration. The storage solution is then transported to the industrial site where it will be used. During the utilization step, the storage provide heat while it consumes electricity to run the auxiliaries and waste heat to valorise. Besides, a replacement of a part of the equipment is considered in this step. The system boundaries end with the landfilling of unrecyclable wastes or the transportation of recycled wastes to recycling plant (the recycling process is not included) that is consistent with the boundaries beginning (avoid double counting). The same system boundaries definition is considered for the gas burner providing the conventional heat.

2.4. Sensitivity study

The parameters in Table 1 could vary in an acknowledged range of values: the storage charge and discharge efficiency, the fans efficiency, the storage porosity and the system lifetime. Their possible variations

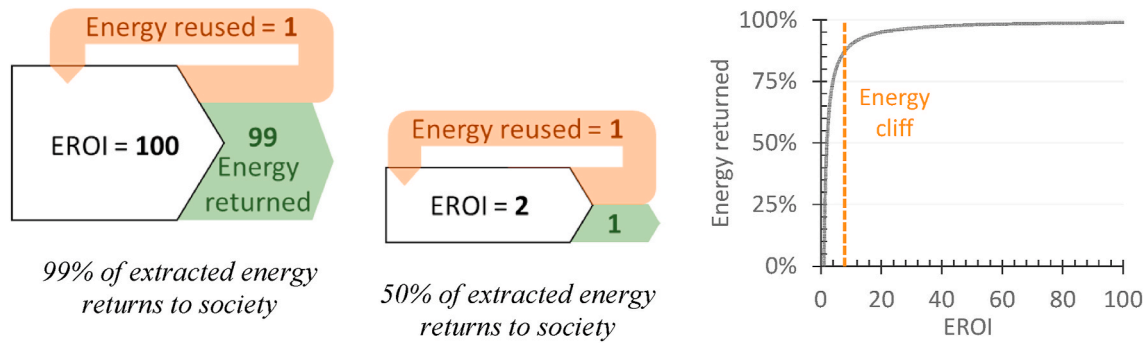


Fig. 2. EROI – Principle of energy returned to society (left) & energy cliff (right) (from (Murphy and Hall, 2010)).

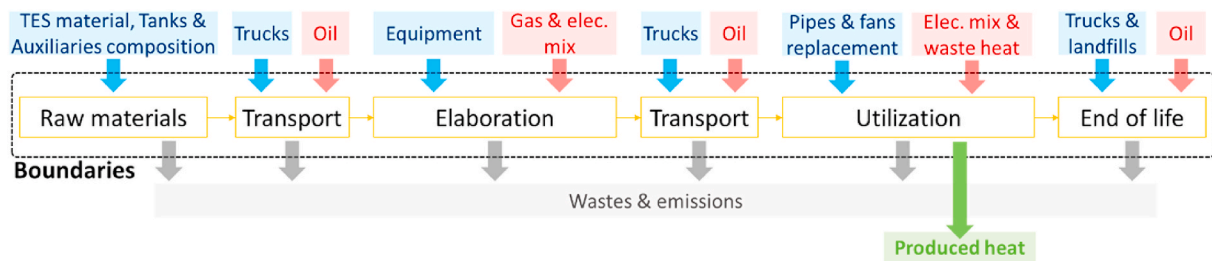


Fig. 3. Cradle-to-grave boundaries – Energy (red) and materials (blue) inputs, wastes and emissions (grey) and function (green) through the lifecycle steps.

Table 1
Variability range on operating parameters.

Parameter	Efficiency			Storage porosity	System lifetime
	Charge	Discharge	Fans		
reference value	80%	92%	77%	40%	25 years
Variation	±2%	±4%	±2%	±2%	±2 years

were used to define a specific error on each environmental impact indicator. The parameters reference value and range of variation were set according to the Eco-Stock® industrial manufacturer, except for the fans efficiency that is defined by commercial specifications. The charge efficiency includes the thermal losses due to the piping and mostly to the non-valorisation of low-temperature waste heat (a threshold is set at 150 °C), while the discharge efficiency is only affected by the piping thermal losses and potentially by a self-discharge depending on the use conditions (the whole heat is valorised up to ambient temperature). Those efficiencies are consistent with similar systems in the literature (Touzo et al., 2020), adjusted by the specific temperature threshold corresponding to the industrial application, as this parameter is known to be of major influence (Fasquelle et al., 2018). The storage porosity corresponds to a homogeneous and disordered granular media porosity that is about 30%–40% (Nellis and Klein, 2008), usually set closely to 40% in real systems (Ortega-Fernández and Rodríguez-Aseguinolaza, 2019) and even increased by the use of internal separators (Fasquelle et al., 2018). The Eco-Stock® storage porosity is evaluated at 40% and may be impacted by the bed settling.

The sensitivity study was conducted by varying one parameter at once, keeping the other at their reference value. The parameter variation induces a variation on the calculated environmental impacts indicators: the range of values reached by the indicator defines its uncertainty as the square of the maximum difference to the indicator reference value. The global error on each indicator is calculated by a propagation of the uncertainty due to each parameter variation, defined as the square of the sum of each uncertainty. This error is then reported on the results as error bars on the graphs presented on the result section.

3. Life cycle inventory

3.1. Storage materials elaboration

Two types of storage materials are studied: a conventional one, bauxite provided by an industrial material provider, and an innovative made at 100% from industrial wastes (coal fly ashes (CFA) and SD clay) provided by Eco-Tech Ceram (ETC). The materials composition is detailed in Table 2, which also contains the elaboration steps: grinding (in case of dispersed granulometry), blending, extrusion and firing. These processes are described by their most impacting input, the energy consumption, and the last step is the more energy consuming. The materials wastes (dust, shaping, etc.) are directly reused in processes, they are consequently not considered as outputs.

The CFA ceramic production being not industrialized yet, their drying and firing energy needs were defined as similar as bricks cooked at similar temperature level and residence time (European Commission, 2007). The bauxite is dried in atomization tower and fired in tunnel kiln, whose consumption were provided by the supplier (factory in Spain). They are higher than those for CFA ceramic because of a higher temperature cooking (~300 °C above) and the densification plateau is longer. After drying, the materials lose the water they contained to facilitate the shaping (humidity mass lost) and the remaining water during firing (ignition mass lost).

The Bauxite elaboration data from Table 2 is presented on a chain diagram (Fig. 4) to illustrate the process, the CFA ceramic elaboration being similar.

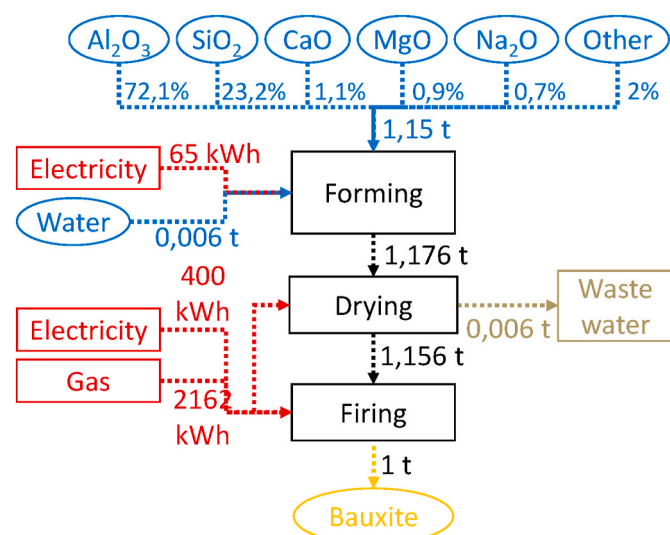
The industrial wastes are considered virgin of any environmental burden. However, the bauxite presents superior thermal performances, especially concerning the thermal conductivity (Table 3). That will induce some dimensioning consequences of the TES unit to guarantee a same service provided by the CFA ceramic:

- The lower thermal capacity (~10%) requires a higher mass;
- The lower density (~25%) involves a higher storage volume;
- The lower conductivity (~75%) requires a smaller diameter preventing internal thermal transfer limitation: 4 cm for bauxite and 3 cm for CFA. The diameter reduction increases the pressure drop.

Table 2

Raw materials and energy for 1 t of ceramic. Energy from French mix – Transport included ETC: Eco-Tech Ceram; MP: TES material provider; BREF: Reference Document on Best Available Techniques (European Commission, 2007).

	EcoInvent process	CFA		Bauxite		Source
		Mass	Conso.	Mass	Conso.	
		t	MWh t ⁻¹	t	MWh t ⁻¹	
Composition						
Ashes&clay	Wastes	1.05	–	0	–	ETC
Al ₂ O ₃	Aluminium oxide	–	–	0.829	–	MP
SiO ₂	Silica sand	–	–	0.267	–	
CaO	Quicklime, milled	–	–	0.0127	–	
MgO	Magnesium oxide	–	–	0.0104	–	
Na ₂ O	Sodium oxide	–	–	0.0081	–	
Other	Chemical inorganics	–	–	0.023	–	
Elaboration						
Water	Tap water	0.026	–	0.006	–	ETC
Waste water	Treatment of wastewater from ceramic production	0.026	–	0.006	–	
Grinding	Electricity, medium voltage	–	–	1.156	0.02	
Blending		1.176	0.015	1.156	0.015	
Extrusion		1.176	0.032	1.156	0.03	
Drying	Natural gas, burned in industrial furnace >100 kW	1.176	0.15	1.15	2.162	BREF
Firing		1.05	1.1	1.15	2.162	MP
Auxiliaries	Electricity, medium voltage	–	0.1	–	0.4	
TOTAL		1	1.397	1	2.627	

**Fig. 4.** Bauxite elaboration main steps and data.**Table 3**

Thermophysical properties of the two studied ceramics at 500 °C.

Ceramic	Density (g cm ⁻³)	Specific heat (J kg ⁻¹ K ⁻¹)	Thermal conductivity (W m ⁻¹ K ⁻¹)
Coal fly ash (CFA)	2.2	1050	<1
Bauxite	2.98	1150	3.6

3.2. Tanks manufacturing

The storage tanks are made of a 6 mm thick cylinder of stainless steel insulated with 300 mm of rock wool maintained with a 1 mm steel sheet (Fig. 5). The tanks are placed on a steel frame and connected to the industrial kiln with insulated ducts to recover the hot fumes. The cooled fumes are released through a chimney. During charging step, these fumes are blown by fans that also blow the cold air during discharge. The tank is slightly larger in the CFA ceramic system to get a same storage capacity than the bauxite ceramic system despite the lower density. The two tanks have a same ratio between length and diameter ($L/D = 1.5$), but different diameter of respectively 1.94 m and 1.72 m.

The tank and auxiliaries' components are listed in Table 4. The fans and control cabinet EcoInvent processes have been adapted to fit the studied system dimensioning according to industrial available data. It includes the materials transport and assembling energy. The energy to assembly the remaining materials is neglected and they are transported by 30 t lorry over 500 km token as mean value.

3.3. Storage utilization

The storage system includes two auxiliaries: the control cabinet has a constant consumption of 0.2 kWh cycle⁻¹ during any storage cycling, and the fans have a consumption defined by the flowing fumes specific to the charge or discharge (see Annex B for calculation details). The EcoInvent process used is the medium voltage electricity from French grid.

The energy to valorise during the defined cycles number and duration set the charging flow and power as well as the corresponding pressure drop. The pressure drop is proportionate to the ratio between the tank length and its square diameter and to the inverse of the ceramic balls' cubic diameter. Thereby, the tank diameter and the ceramic balls diameter drives the pressure drop:

- The smaller CFA ceramic balls increase the pressure drop,
- The higher tank diameter in the CFA ceramic storage decreases the pressure drop.

These two phenomena almost compensate in the studied case, leading to fans consumption of 7.6 and 7.7 kWh cycle⁻¹ respectively for CFA and bauxite ceramic systems. Besides these consumptions, the utilization step includes a complete replacement of the ducts, the chimney and the fans: this is defined as preventive maintenance de-risking the lifetime hypothesis.

**Fig. 5.** Industrial installation of the Eco-Stock® thermal storage system.

Table 4

Thermocline 2-tanks composition. *the output fan is 2.5 time smaller than the input one**adaptated from EcoInvent process 'heat and power co-generation unit, 160kWe'.

EcoInvent process		CFA	Bauxite
		Mass (t)	Mass (t)
Frame	Steel section	0.384	
Tank	Chromium Steel 18/8 sheet	2.750	2.294
Stiffener & nozzle	Chromium Steel 18/8 sheet	0.340	
Insulation	Rock wool	1.536	1.241
Cover	Steel sheet	0.126	0.149
Ducts & Chimney	Chromium Steel 18/8 sheet	1.3	
	Rock wool	0.473	
	Aluminium sheet	0.031	
Fans	Air input/output unit**	1.4*	
Control cabinet	Control cabinet**	1	

3.4. End of life management

At the end of the storage system life, the fraction of recycled materials is carried to the recycling facility (500 km) where the system perimeter ends: 98%, 93% and 90% respectively for stainless steel, steel and aluminium (International Resource Pa, 2011). The remaining materials are transported to waste treatment centre (500 km) and landfilled (EcoInvent process: process-specific burdens production, inert material landfill). The insulation material is fully landfilled (EcoInvent process: treatment of waste mineral wool, inert material landfill). The storage ceramics (CFA or bauxite) are reused, thus only a transport step is considered.

4. Results

4.1. Indicators selection

There are four impact categories in the International Reference Life Cycle Data (ILCD) method (European Commission - Joi, 2011) and various indicators per category (Table 5) (European Commission - Joi, 2010). One indicator per category should be studied to simplify the analysis. In addition, an energetic indicator is considered: the cumulative energy demand (CED). The indicators have been selected according

Table 5

Indicators selection and normalization.

Category	Indicator	Unit	Normalization	Robustness
Energy	Cumulative energy demand	MJ-Eq	27	–
Climate change	GWP 100a	kg CO ₂ -Eq	10	Very High
	Ecosystem quality	Freshwater & terrestrial acidification	mol H ⁺ -Eq	11
Freshwater eutrophication		kg P-Eq	20	Medium/Low
Marine eutrophication		kg N-Eq	5	Medium/Low
Terrestrial eutrophication		mol N-Eq	5	Medium
Human health	Ionising radiation	kg U ²³⁵ -Eq	115	Medium
	Ozone layer depletion	kg CFC-11-Eq	1.2	Medium
	Photochemical ozone creation	kg C ₂ H ₄ -Eq	7.5	Medium
	Respiratory effects, inorganics	kg PM2.5-Eq	22	Very High
Resources	Land use	kg SOC-Eq	0.8	Medium
	Mineral, fossils and renewables	kg Sb-Eq	97	Medium

to the following criteria:

- Their **relative weight**: the values calculated for the reference case (with bauxite) were normalized by the annual impacts of a European resident (Aymard and Botta-Genoulaz, 2017). The indicator with the higher normalized value in each impact category represents the impact with the higher relative weight on environment;
- Their **robustness** (established by (Aymard and Botta-Genoulaz, 2017)): if the normalized indicators had similar values ($\pm 20\%$), the most robust one was selected.

The normalization coefficients are provided in the Annex A.

Besides, the ionising radiation indicator was ruled out, as its high relative value is due to the fans' electric consumption from French grid mix that have a particularly high nuclear energy proportion (>70%): it is thus not specific to the studied storage solution. Finally, the following indicators have been selected:

- **Energy**: Cumulative Energy Demand (CED)
- **Climate change**: Global Warming Potential (GWP)
- **Ecosystem quality**: Freshwater eutrophication (FE)
- **Human health**: Respiratory effects or particles emission matter (PM)
- **Resources**: Mineral, fossil and renewable resources depletion or abiotic depletion potential (ADP)

4.2. Thermal energy storage (TES) materials cradle to gate impacts

Fig. 6 depicts the composition and elaboration normalized impact indicators of CFA and bauxite ceramics. The CFA ceramics indicators (left) have lower values in each category: the reduction factor is about ~50 for freshwater eutrophication, ~30 for particles emission and resource depletion, and ~4 for GWP and CED. The following observation may be done by looking in detail at the two manufacturing steps:

- **Composition**: the higher impact is on resource depletion, reduced by 97% when ceramics are made from waste. The aluminium oxide is responsible on more than 90% of all bauxite composition studied impacts;
- **Elaboration**: the higher impact is on energy consumption (CED), followed by global warming potential; and is due to the firing step. As the CFA ceramics are sintered at lower temperature than bauxite and require shorter firing plateau, thus their impacts are reduced.

Hence, the advantageous impacts reduction provided by the use of industrial wastes as storage materials mineral resources could be improved by using a waste that would not need to be fired, decreasing specifically the CED and GWP indicators. Some other wastes suitable for thermal storage (Gutierrez et al., 2016) present this feature, such as vitrified asbestos or steel slag: as the waste melting is part of another

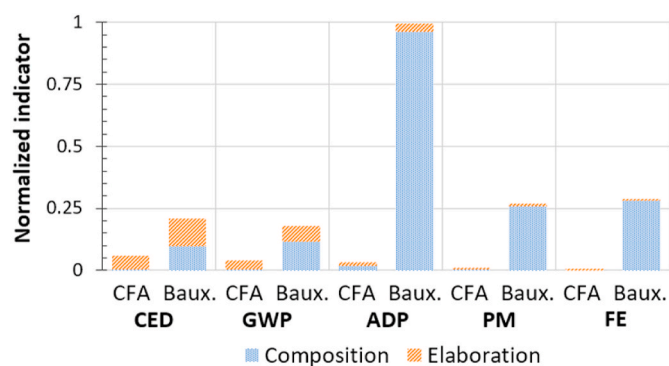


Fig. 6. Normalized impacts indicators by ton of CFA and bauxite ceramics produced.

lifecycle (respectively dangerous waste treatment and steel making), it is not accounted in the thermal storage material elaboration step (Lalau et al., 2016). In the case of the vitrified asbestos containing waste named Cofalit®, the targeted indicators would be reduced ~7 times compared to CFA ceramic. More precisely, the TES material cradle to gate impact is equal to:

- Global Warming Potential: 1.64 kgCO₂-eq kg⁻¹, 0.37 kgCO₂-eq kg⁻¹ and 0.050 kgCO₂-eq kg⁻¹ respectively for bauxite, CFA ceramic and Cofalit®;
- Cumulative Energy Demand: 28.5 MJ-eq kg⁻¹, 8.2 MJ-eq kg⁻¹ and 1.3 MJ-eq kg⁻¹ respectively for bauxite, CFA ceramic and Cofalit®.

4.3. Thermal energy storage system life cycle total impacts

The whole storage system lifecycle normalized impacts indicators are represented on Fig. 7, including error bars about 10% of the indicator value (ranking from 8 to 11%). Those error bars are the result of the sensitivity study: a variation of the considered parameters in their defined range of uncertainty lead to an error ranking from 8 to 11% depending on the indicators.

The storage system made with CFA ceramics (on the left) have lower impacts, about ~30% for GWP, ~20% for ADP, PM and FE, and ~10% for CED – that last decrease is not relevant regarding the error. The abiotic depletion potential indicator shows the higher value (77 and 97), followed by the cumulative energy demand (24 and 27), then particles emission matter and freshwater eutrophication (around 17 and 22), and finally the global warming potential (7 and 10).

The **utilization phase** is responsible of most of the cumulative energy demand (>70%), and of a more than 25% of the abiotic depletion potential, the two mains indicators concerned by this phase. The higher contributor to all utilization impacts is the fans consumption (~95% for the CED), followed by the piping replacement (~40% of resource depletion indicator). The **tanks manufacturing step** have a negligible influence on the cumulated energy demand, but a major weight on the other indicators. Those environmental impacts are mainly due to the stainless steel employed for the tank and the piping. The **ceramic manufacturing step** have noticeable impacts only when bauxite ceramics are employed, mainly in resource depletion that is consistent with the storage materials “cradle to gate” assessment. The **end of life and transport phases** have negligible impacts, lower than 2% of each indicator. As most of the storage system components are easily reusable or recyclable, the landfilling impacts are reduced (mainly due to insulation materials), and the end of life management mainly consist in transporting the recyclable materials to the recycling factory, the recycling process being out of the system boundaries.

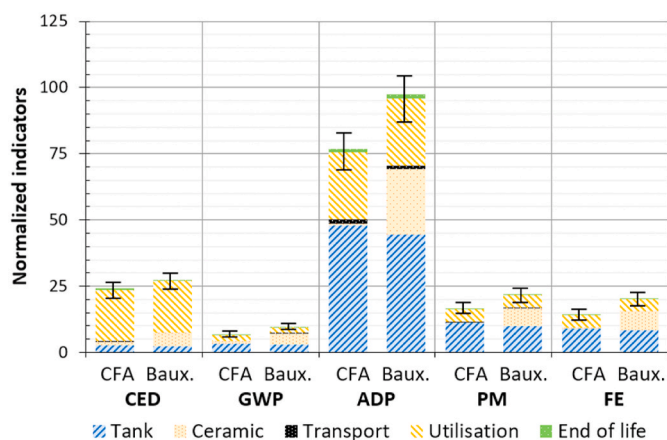


Fig. 7. Normalized impact indicators distributed through the lifecycle steps for CFA and bauxite storage systems.

Thus, both storage systems have similar impacts, with an advantage in reducing abiotic depletion potential for CFA ceramics system. They intend to provide a low-impact heat replacing natural gas consumption; their impacts should then be compared.

5. Discussion

5.1. Supplied heat impacts

Table 6 gives each indicator value per MWh of provided heat by CFA or bauxite ceramic storage system and natural gas burning. The gas environmental indicators are given by the EcoInvent process “Natural gas, burned in industrial furnace >100 kW” adapted to French mix.

5.2. PBT & EROI

5.2.1. Payback time (PBT)

The heat provided by the storage system substitutes a fraction of fossil fuel consumption at a lower environmental impact (cf. Table 6). That benefice may also be evaluated by calculating the time the system needs to payback its lifecycle ecological duty (Equation (1)). That time should be as low as possible and of course shorter than the system lifetime (25 years). Each indicator PBT is depicted on Fig. 8.

The CED and GWP payback times (PBT) are negligible before the 25 years of lifetime: both are lower than 2 months. The ‘PM PBT’ and the ‘FE PBT’ are greater, respectively about 3 and 5 year, but remain below 20% of the system lifetime. Nevertheless, the ‘ADP PBT’ is close to the middle of the lifetime: 12 years for the storage containing CFA ceramic and 17 years for the conventional one. Giving the other indicators reduction breadth that value may be accepted, but some caution should be taken with this indicator: a change in the operating conditions could quickly increase the PBT over the lifetime limit, as studied in the 5.3 section on operating scenarios.

5.2.2. EROI

The energy returned by each system is represented as function of their EROI (Equation (2)) on Fig. 8. The reference natural gas EROI is based on CED from the EcoInvent process “Natural gas, burned in industrial furnace >100 kW” adapted to French mix (consistent with reference impact indicators, see V.I). The calculated value of 21 is coherent with literature (N.B.: an EROI of 12 for electricity from CCGT plant accounts for a ~60% conversion efficiency, leading to a value of 20 for the gas combustion) (Arvesen and Hertwich, 2015). In Fig. 9. a, the EROI value of heat from gas burning (black cross), from CFA ceramic storage (grey square) and bauxite ceramic storage (yellow circle) are

Table 6
Impacts per MWh of provided heat by storage system or gas burning.

	CED MJ- eq	GWP kgCO ₂ -eq	ADP kgSb-eq	PM kgPM2.5- eq	FE kgPO ₄ ²⁻ - eq
CFA ceramic storage system	65	1.2	16 × 10 ⁻⁵	13 × 10 ⁻⁴	42 × 10 ⁻⁵
Bauxite ceramic storage system	74	1.8	20 × 10 ⁻⁵	17 × 10 ⁻⁴	60 × 10 ⁻⁵
Natural gas burned in industrial furnace	4375	254	26 × 10 ⁻⁵	12 × 10 ⁻³	22 × 10 ⁻⁴

Compared to heat from natural gas burner, the valorisation of waste heat using a TES system presents major environmental improvements.

- Excellent in decreasing CED and GWP (>98%);
- Very good in decreasing PM (almost 90%) and FE (~75%);
- Good in decreasing ADP in the case of CFA system (46%), but minor in the case of the bauxite system, just above the 10% error margin (17%).

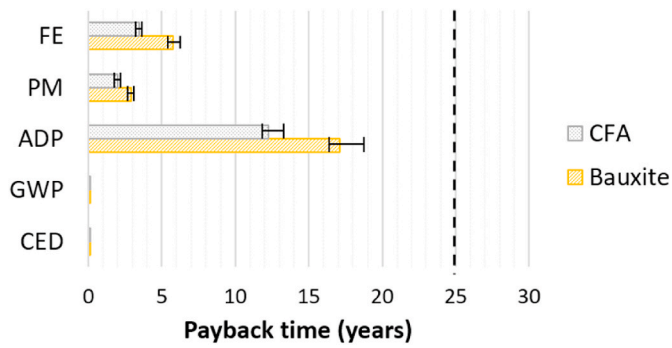
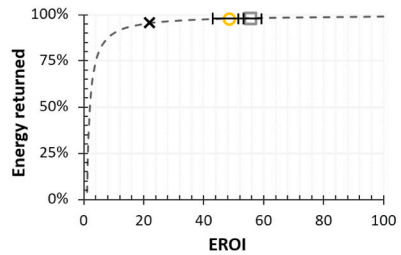
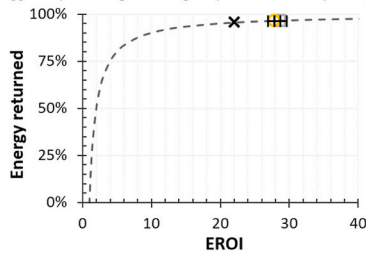


Fig. 8. Impact indicators payback time for CFA (grey dots) and bauxite (yellow lines) storage systems.



a. Energy supplied by natural gas burning or by the heat recovery storage system



b. Energy supplied by industrial furnace: conventional facility or equipped with a heat recovery storage system

Fig. 9. Energy returned as function of EROI.

presented linked to their respective energy returned values. In Fig. 9. b, there is the EROI value of heat from an industrial furnace, supplied at 100% by gas (same value than before) or at 60% by gas and 40% by valorised heat from storage. Those 40% correspond to the ratio of substitute heat in the studied case, based on fuels flows and recovered proportion of waste heat.

The heat from storage systems present EROI values more than twice higher than gas, and far away from the “energy cliff”, enlightening an excellent energetic profitability. When this system is integrated to a conventional gas furnace, the heat provided by the new facility shows an EROI increased by 30%.

These very positive results could be affected by a modification of the operating conditions: that sensitivity should be studied through different scenarios.

5.3. Operating scenarios

5.3.1. Parametric study

While keeping the unit function constant, some parameters could vary depending on site specifications:

- The annual number of charge/discharge cycles N_{cycles} : 350 to 2100 cycles per year (1–6 per day),
- The storage temperature T_{max} : 200–600 °C,
- The daily duration of storage cycles slots H_{slot} : 8–24 h per day.

The storage system dimensions (diameter and length) are adapted to fit with those specifications: e.g., a lower temperature will require a larger tank (with same L/D ratio) to reach the same storage capacity.

The parametric study objective is to evaluate how those modifications affects the storage system environmental performances. The most sensitive impact being the resource depletion, only this indicator variation is studied through its payback time to simplify the analysis: it should remain below the system lifetime (25 years). The EROI value is also considered and it should remain above the natural gas value (21). Besides, the EROI and the ADP are both sensitive sustainability indicators in the energy transition domain (Capellán-Pérez et al., 2019).

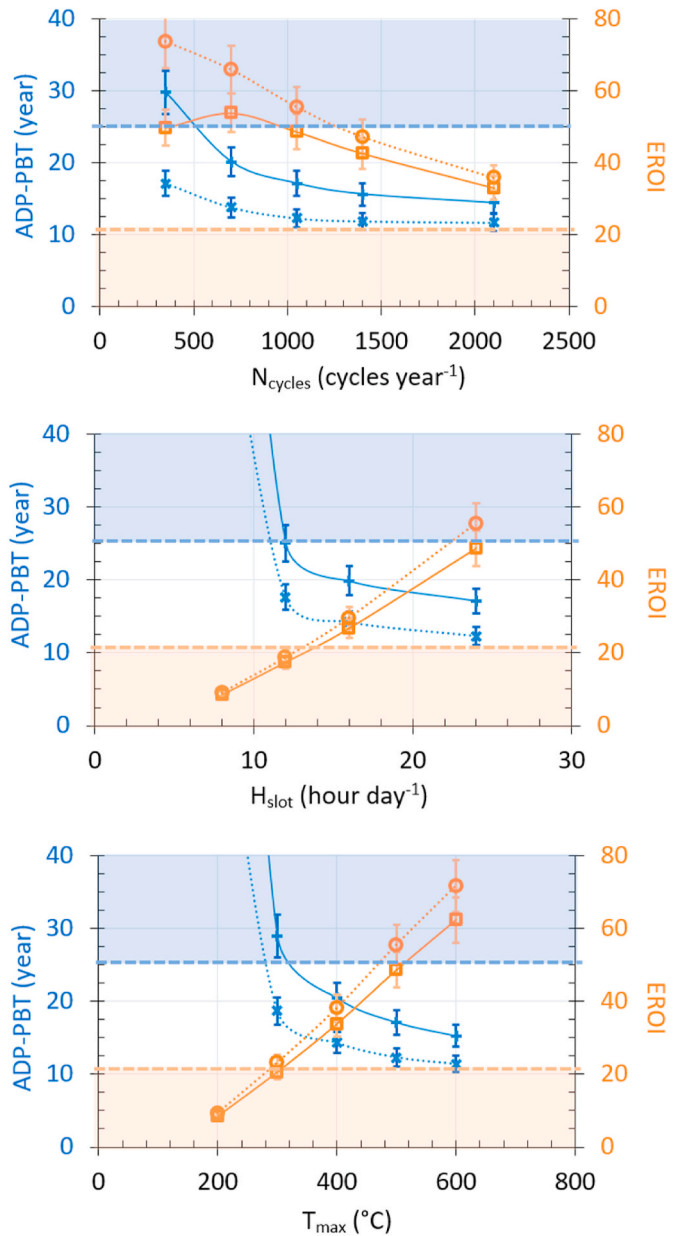


Fig. 10. Influence of (from left to right) the annual number of cycles N_{cycles} , the storage temperature T_{max} and the slot daily duration H_{slot} on the Abiotic Depletion Potential Payback time (left axis) and the EROI (right axis) of the two studied storage systems: with CFA and bauxite ceramics.

Fig. 10 depicts the results.

As it could be intuited, a higher storage temperature T_{max} and a longer daily utilization H_{slot} have a positive influence on the solution environmental profile (the PBT decreases and the EROI increases): both requires smaller storage tanks and a long H_{slot} reduces the storage system nominal power, thus the required flow and consequently the fans consumption.

An increased number of cycles N_{cycles} has antagonist effect on the ADP-PBT and the EROI. The same amount of energy is supplied by smaller tanks operating an increased number of charge/discharge cycles, thus reducing the resource consumption to build the storage tanks. In another hand, those smaller tanks have smaller diameter that involves an increase in pressure drop, thus in the fans' consumption, and thus in the energy needed to supply the heat – that reduces the EROI. The bauxite ceramic storage shows an optimal EROI: until ~ 750 cycles per year, the reduction in the tank size decreases the energy consumption of the elaboration step without dramatically increasing the pressure drop, favouring higher EROI values; but that trend is then reversed and the EROI starts to decrease due to higher fans consumption that is no longer compensated by the lower energy consumption for the elaboration.

The CFA ceramic storage appears to be more resilient against this constraint: e.g., it turns profitable again starting $T_{max} = 280$ °C while the bauxite ceramic storage needs a minimum of $T_{max} = 320$ °C or keep high performances for a wide range of number of cycles while the bauxite ceramic storage turns detrimental below 500 cycles per year. Thus, the ceramics made from wastes improves the system adaptability to various operating conditions variations.

5.3.2. Detrimental cases improvement

In the previous paragraph, some operating conditions appeared to be not suitable to the studied storage system. However, some design parameters may be altered to improve those low environmental performances cases: a smaller ratio between length and diameter (L/D) decreases the pressure drop thus increases the EROI; and a higher porosity also decreases the pressure drop but at the same time slightly increases the storage tank dimensions that increases the resources consumption. One can note that previous work showed a thermal transfer improvement between the heat transfer fluid and the heat storage media without increasing the pressure drop by using structured ceramic bed, addressing at the same time the thermal ratcheting issue (Motte et al., 2014).

The L/D ratio and the porosity management were explored as improvement drivers, and succeeded in turning profitable some of the most detrimental cases (Fig. 11): low temperature, $T_{max} = 200$ °C (left), and short cycles duration, 8 h day^{-1} (right). For both cases, the L/D ratio is set to 1 and then additionally the porosity is extended to 42%.

The first statement could be that the CED and GWP payback times are so low that they are not significantly affected by the changes in operating conditions. The CFA ceramic storage system shows quite constant payback times for PM and FE, but some significant increase may be observed with bauxite especially on FE – even if remain within acceptable ranks.

Then, it is interesting to see that the design improvements work to bring the CFA ceramic storage system up to ADP PBT lower than 25 years and EROI higher than 21, but fails with bauxite at low temperature: that confirms the added value of using materials from wastes.

However, it should be noticed that the parameters were not pushed on their optimal values to obtain cautious results: e.g., the storage efficiency is not defined at its theoretical maximum (see Table 1) or the temperature is not set at the operating maximum allowed (600 °C). Indeed, those parameters are operating constraints and not design

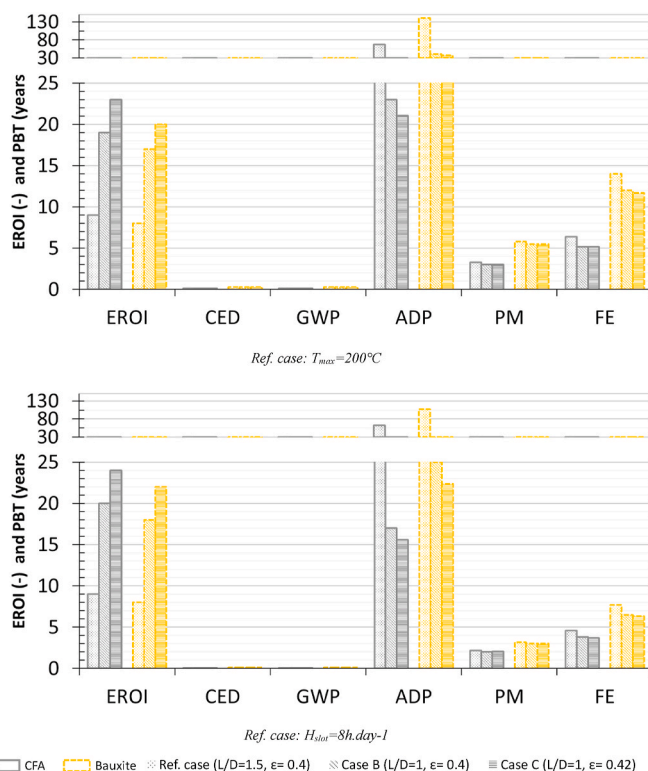


Fig. 11. EROI increase and PBT decrease by design parameters adjustment (L/D ratio and porosity) for CFA and bauxite ceramic storage systems.

options, thus in real cases (and not parametric studies) they should be considered as inputs and not variables.

6. Conclusion

This work focused on a thermal energy storage (TES) system recovering the waste heat from industrial furnace to provide a low-carbon content heat substituting natural gas. Two systems were studied, each one containing specific TES materials: bauxite and waste-based ceramic made from coal fly ashes (CFA) sintering. The recovered heat shows lower environmental impacts than gas (see Table 6): the global warming potential (GWP) and cumulative energy demand (CED) are drastically reduced (two order of magnitude); the particle emission matter (PM) and freshwater eutrophication (FE) are significantly reduced (one order of magnitude); and the abiotic depletion potential (ADP) is less impacted as the reduction leads to an indicator value of the same order of magnitude, even is close to the calculation error ($\sim 10\%$). The use of industrial wastes as mineral resources for TES materials is mainly favouring the resource depletion limitation. Finally, the heat energetic profitability is increased, with an EROI value more than twice the one calculated for natural gas (respectively 55 and 49 for TES systems with CFA and bauxite, and 21 for gas).

Thereby, the proposed energy efficiency system allows clear improvements on some heat environmental indicators (mainly CED and GWP) while is less efficient on other, namely the resource depletion. An operation out of the foreseen conditions may even lead to counter-performance on this indicator. Front to such variation, the storage system containing ceramics made from wastes kept its environmental performances over a wider range of conditions, showing a better system adaptability. However, those operating scenarios investigations were performed with an adjustment of the tanks size, but keeping the same design parameters such as the L/D ratio, the cycling monitoring or the bed porosity: the last part of this work enlighten the improvement potential, even for the worst scenarios, by an adaptation of those

parameters. Besides, most of the remaining impact indicators were payback in duration similar to reference case: the ADP-PBT improvement does not involve a deterioration of other indicators PBT.

Thus, a sizing or operating modification of an energy efficiency system may be detrimental for some impact categories: an analysis based on the real system use and integrating the relevant indicators should then be performed. That study comparing forecasted and measured benefits could be performed on an industrial system commissioned in 2020 since sufficient operating data will be available. Some environmental performances degradation may be mitigated by an optimisation of the studied design parameters: this observation could be coupled with an energetic and economic optimisation study in future work.

CRedit authorship contribution statement

Yasmine Lalau: Methodology, Investigation, Writing – original draft. **Ibrahim Al Asmi:** Software, Writing – review & editing. **Régis**

Olivès: Resources, Writing – review & editing. **Guilhem Dejean:** Resources, Writing – review & editing. **Antoine Meffre:** Funding acquisition, Writing – review & editing. **Xavier Py:** Conceptualization, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Annex A. Mid-point impacts indicators & normalization coefficient

Except for the CED indicator, all the normalization factors are recommended by the Institute for Environment and Sustainability from European Commission, corresponding to the impacts of a typical resident from the EU-27 in 2014. The CED normalization factor is defined by the French energy agency ADEME in its tool BilanProduit® as representative of an EU-15 resident impact in 2011.

Table A.1
Impact indicators robustness and normalization coefficient

Impact category	Indicator	Abb.	Unit	Robustness	Norm.
Climate change	Global warming potential	GWP	kg CO ₂ eq.	I	9220
	Ozone depletion	ODP	kg CFC-11 eq.	II	2.16E-02
Human health	Human toxicity - cancer effect		CTUh	IV	3.69E-05
	Human toxicity - noncancer effect		CTUh	IV	5.33E-04
	Particulate matter	PM	kg PM2.5 eq.	I	3.8
	Photochemical ozone formation		kg NMVOCeq.	II	31.7
	Ionising radiations		kBq U ²³⁵ eq.	II	1130
Ecosystem quality	Ecotoxicity for aquatic fresh water		CTUe	IV	8740
	Acidification potential	AP	mol H ⁺ >+ eq.	I	47.3
	Eutrophication - terrestrial		mol N eq.	II	176
	Eutrophication - freshwater	FE	kg P eq.	III	1.48
	Eutrophication - marine		kg N eq.	III	16.9
Resource depletion	Land use		kg C deficit	II	7.48 E+04
	Resource depletion - water		m ³ water eq.	IV	81.4
	Resource depletion - mineral, fossil & renewable	ADP	kg Sb eq.	II	0.101
Energy consume	Cumulative energy demand	CED	MJ eq.	–	1.53 E+05

In this work, only the most robust indicators were considered (robustness level from I to III, preferably I or II). The normalization coefficients from the above table were applied on a reference case with bauxite as TES material, and the resulting normalized impact were compared to select the most relevant.

Annex B. Fans consumption

The fans electricity consumption during one cycle is related to the power needed to overcome the pressure drop during the charge and discharge ($P_{fan, ch}$ and $P_{fan, disch}$) during the corresponding slot, respectively $H_{slot, ch}$ and $H_{slot, disch}$. Along their lifetime, the fans consume the energy E_{fans} corresponding to N_{cycles} .

$$E_{fans} = N_{cycles} \times (H_{slot, ch} \times P_{fan, ch} + H_{slot, disch} \times P_{fan, disch}) \quad (1)$$

The fans power is then related to the charge and discharge pressure drop PD_{ch} and PD_{disch} , the heat transfer fluid temperature T_{max} and charge and discharge flow rate \dot{Q}_{ch} and \dot{Q}_{disch} . It also considers the fans efficiency η_{fan} , and is calculated for:

➤ The charge:

$$P_{fan, ch} = \frac{\dot{Q}_{ch}}{3600} \times \frac{T_{max} + 273,15}{273,15} \times \frac{PD_{ch}}{\eta_{fan}} \quad (2)$$

➤ The discharge:

$$P_{fan, disch} = \frac{\dot{Q}_{disch}}{3600} \times \frac{T_{max} + 273,15}{273,15} \times \frac{PD_{disch}}{\eta_{fan}} \quad (3)$$

The total pressure drop includes the inner tank pressure drop and the inlet and outlet pressure drop due to diameter variation between the tank and the piping (PD_e and PD_s), those last ones being almost negligible. They may be calculated during:

➤ The charge:

$$PD_{ch} = \frac{PD_{ch,initial} + PD_{ch,final}}{2} + PD_e + PD_s \quad (4)$$

➤ The discharge:

$$PD_{disch} = \frac{PD_{disch,initial} + PD_{disch,final}}{2} + PD_e + PD_s \quad (5)$$

In the tank, the pressure drop differs across the cycles, as the heat transfer fluid (HTF) is cold in the charge beginning and hot in the ending (conversely in the discharge) and its properties varies with the temperature (density ρ_{HTF} , dynamic viscosity μ_{HTF}). The mean value is considered as an acceptable approximation. Besides, the heat transfer fluid is the recovered process fumes during the charge and ambient air during the discharge.

The following equations illustrate the initial and final pressure drop calculation, based on the HTF speed v in the granular media of a given porosity ϵ .

$$PD_{ch,initial} = L \times \left(\frac{150 \times \mu_{HTF} \times (1 - \epsilon)^2}{d_{TES\ material}^2 \times \epsilon^3} \times v_{initial} + \frac{1,75 \times \rho_{HTF} \times (1 - \epsilon)}{d_{TES\ material} \times \epsilon^3} \times v_{initial}^2 \right) \quad (6)$$

with:

$$v_{ch,initial} = \frac{\dot{Q}_{ch}}{3600 \times \pi \times \left(\frac{D}{2}\right)^2} \quad (7)$$

$$PD_{ch,final} = L \times \left(\frac{150 \times \mu_{HTF,T} \times (1 - \epsilon)^2}{d_{TES\ material}^2 \times \epsilon^3} \times v_{final} + \frac{1,75 \times \rho_{HTF,T} \times (1 - \epsilon)}{d_{TES\ material} \times \epsilon^3} \times v_{final}^2 \right) \quad (8)$$

with:

$$v_{ch,final} = \frac{\dot{Q}_{ch}}{3600 \times \pi \times \left(\frac{D}{2}\right)^2} \times \frac{T_{max} + 273,15}{273,15} \quad (9)$$

The discharge fan is approximatively twice smaller than the one use for the charge, respectively around 600 W and 1300 W.

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