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Long-duration thermo-mechanical energy storage – Present and future techno-economic competitiveness

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HIGHLIGHTS

- Unified techno-economic comparison of 6 thermo-mechanical energy storage concepts.
- ullet 100 MW ACAES and LAES exhibit lower LCOS than Li-ion batteries above \sim 4 h duration.
- New technological concepts can meet cost target below 20 USD/kWh at 200 h duration.
- Promising high-temperature thermochemical reactions for long-duration storage.
- Identified material, device and system-level advancements needed to compete with H₂.

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ABSTRACT

The extent to which long-duration energy storage (LDES) will support grid decarbonisation by enabling large penetration of renewable generation is subject to the achievement of suitable technical and economic performance. This study investigates the potential of established and novel thermo-mechanical energy storage (TMES) technologies to meet LDES targets, benchmarks TMES current and future techno-economic performance and highlights critical research developments. Results justify the priority of ensuring low storage costs over high roundtrip efficiency for LDES, thus endorsing novel concepts based on thermochemical energy storage. Besides adiabatic compressed air energy storage, novel TMES using metal oxidation/reduction and CaO hydration/dehydration reactions can potentially already meet the 20 USD/kWh cost target at 200 h duration, with current technology performance. The need for suggested and wide-ranging enhancements at material, device and system level is discussed, which may lead to TMES costs below 14 USD/kWh – competitive with long-duration solutions like hydrogen for covering the energy balancing needs of future low-carbon energy systems.

1. Introduction

Large deployment of renewable energy sources (RES) is a major way forward to decarbonise the energy system, contain anthropogenic climate change and limit global warming. RES capacity worldwide is expected to more than triple by 2030 and increase ninefold by 2050 (entailing 600 GW yearly addition of solar photovoltaics and 340 GW of wind) [1]. Propelled by the competitive costs of RES technologies per unit energy generated [2], such changes are underway. Yet, the intrinsic intermittency of wind and solar results in challenges to ensuring supply—demand matching over space and time and requires system flexibility [3].

Electrical energy storage is widely recognised as a key enabling

technology to support RES penetration [4] by increasing system reliability and decreasing unmet demand [5]. This work adopts few essential definitions to identify electrical energy storage features and operation:

- \bullet Power output, \dot{W}_{dsc} [MW]: rated electrical power generated
- ullet Capacity, W_{dsc} [MWh]: electricity delivered over a complete storage discharge
- *Duration*, τ_x [h]: time over which the power output \dot{W}_{dsc} can be sustained, starting from fully charged conditions $(\tau_x = W_{dsc}/\dot{W}_{dsc})^{\perp}$
- Idle time, φ [h]: period between the end of storage charge and the beginning of a subsequent discharge.

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 $^{^{1}}$ In this notation, storage duration value is added as subscript to the symbol τ . For instance, τ_{8h} identifies 8h storage duration.

Nomenclature		σ	Void fraction [-] Duration [h]	
Symbol	dc	τ	Idle time [h]	
		arphi	idic tinic [ii]	
Q 	Thermal power [MW]	Subscripts		
W	Power [MW]	0	Standard	
c_p	Specific heat capacity [kJ/kmolK]	h	Hot	
m.	Mass flow rate [kg/s]	C	Compressor	
h	Specific enthalpy [kJ/kg]	HX	Heat exchnager	
Λ	TES heat loss coefficient [1/h]	RT	Roundtrip	
П	Compression/expansion ratio [-]	T	Turbine	
AC	Annual cost [USD]	TES	Thermal energy storage	
CF	Conversion fraction [-]	С	Cold	
EC	Energy-specific cost [USD/kWh]	chr	Charge	
ED	Energy density [kWh/m3]	dsc	Discharge	
EG	Excess gas [-]	eq	Equilibrium	
Н	Reaction enthalpy [kJ/kmol]	f^{-}	Fluid	
MM	Molecular mass [kg/kmol]	in	Inlet	
N	Number of yearly cycles [-]	loss	Loss	
PC	Power-specific cost [USD/kW]	out	Outlet	
Q	Thermal energy [MWh]	r	Reactor	
S	Reaction entropy [kJ/kmolK]			
T	Temperature [K]	Acronym		
V	Volume [m3]	ACAES	Adiabatic compressed air energy storage	
W	Energy [MWh]	CAPEX	Capital expenditure	
d	Discount rate [%]	CES	Carbonates energy storage	
m	Mass [kg]	HES	Hydroxides energy storage	
p	Pressure [bar]	LAES	Liquid air energy storage	
q	Specific thermal energy [kJ/kg]	LCOS	Levelised cost of storage	
t	Time [h]	LDES	Long-duration energy storage	
w	Specific work [kJ/kg]	OES	Oxides energy storage	
γ	Isentropic exponent [-]	PTES	Pumped thermal energy storage	
ε	Heat exchanger effectiveness [-]	RES	Renewable energy sources	
ζ	Thermal loss coefficient [-]	STES	Sensible thermal energy storage	
η	Efficiency [%]	TCES	Thermochemical energy storage	
ν	Stoichiometric coefficient [-]	TES	Thermal energy storage	
π	Electricity purchase price [USD/kWh]	TMES	Thermo-mechanical energy storage	
ρ	Density [kg/m3]	TIVIES	Thermo incentancal energy storage	

Several works indicate a link between RES penetration and the need for storage, whose required capacity is suggested to increase from 1.5 to 6 % of the annual energy demand when moving from 95 to 100 % RES share [6]. Such capacity figures synthesise a highly variable and sitespecific set of recommendations from the literature, where even higher storage fractions are suggested for 100 % RES penetration (e.g. 8.7 % for Brazil [7]; 13.6 % for Europe [8]). Along with capacity, also duration requirements depend on RES share within the energy system (see Fig. 1) [9]. Analyses for the UK under 100 % RES penetration show that overproduction could be stored 82 % of the time [10]. Although storage is expected to shift energy across periods of 8 h or less 80-95 % of the time [11] under 80 % RES penetration, one-third of the annual stored energy comes from charging events lasting over 24 h and up to more than 60-80 h, or six consecutive days [12]. Under these circumstances, deployment of long-duration energy storage (LDES) technologies, i.e. capable of addressing energy supply variability across several days and seasons, will be crucial to achieving large RES penetrations cost-effectively, limiting overcapacity [13] or massive investments in transmission infrastructure [14]. Several cost targets have been proposed in the literature for LDES: 20-35 and 5-15 USD/kWh, respectively, for 50 h and 100 h storage duration [15], 85 USD/kWh for a 100 h storage [16] and 30-73 USD/kWh, depending on the region considered [17]. Amid such breadth of recommendations, recent studies concur in identifying 20 USD/kWh as a suitable cost target for LDES to help ensure

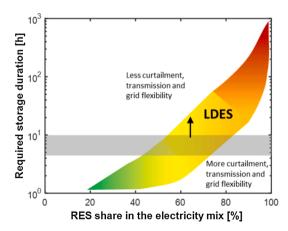


Fig. 1. Overview of the electricity storage requirements to ensure demand is met under increasing penetration of renewable energy sources (adapted from [15]).

reliable baseload electricity across different RES generation mixes [18] and with 10 % lower energy system adaptation costs [19].

Despite a consensus on the need for LDES and the broad requirements LDES should meet, less focus has been devoted to identifying

and addressing the storage technologies that can realistically deliver such long-duration services for the target costs. With over 9000 GWh installed worldwide and capital costs of 600-2000 USD/kW (5-100 USD/kWh) [4], the predominant LDES is pumped hydro, but geographical constraints limit deployment at the required scales [20]. Thermo-mechanical energy storage (TMES) technologies use commercial process engineering components for electricity conversion and storage in the form of heat and/or mechanical potential. During charge, a suitable thermodynamic process converts excess electricity into thermal and/or mechanical energy, which is stored and, during system discharge, becomes the driver of a power cycle, generating electricity back into the grid. Traditional TMES concepts are adiabatic compressed air energy storage (ACAES) and liquid air energy storage (LAES) - both at an early commercialisation stage [21] – and pumped thermal energy storage (PTES) - of which only a few prototypes exist [22]. Thermal energy is stored in these systems via sensible thermal energy storage (STES). Traditional TMES offers relatively cheap storage at ~ 1000 USD/kW, large capacities up to and above hundreds of MWh and no site constraints [21], which makes it a good proposition for LDES. Most recently, novel TMES concepts have been proposed that still rely on the same process engineering components and thermodynamic cycles of traditional concepts, but use reversible thermochemical reactions to store heat, via thermochemical energy storage (TCES) [23]. Due to reduced footprint, cheap materials and no thermal losses associated with TCES [24], these novel concepts seem particularly relevant for LDES [25]. Still, the majority of works consider TMES for applications in the range 4-8 h [26].

Given such background, this work explores and addresses the key unanswered question: to what extent TMES technologies are or will be capable in the future to meet the target techno-economic performance requirements for LDES to sustain decarbonisation? Six between traditional and novel TMES concepts were considered. Bottom-up, thermodynamic models for each of the six TMES with 100 MWe power output were developed and used to predict storage performance for LDES application at different durations, as later described in Section 2. Quantification and comparison of the underlying key performance indicators (KPIs) from Table 1 and the results of a sensitivity analysis to device technical parameters were used to: i) identify promising solutions; and ii) explore the effect and justify the need for future advancements. Both areas are essential to driving the future development of LDES for energy decarbonisation.

1.1. Originality and novelty of this work

This work studies for the first time the techno-economic competitiveness of thermo-mechanical energy storage for long-duration applications. Existing literature mostly addresses LDES performance needs in a technology-agnostic fashion. For instance, Zhang et al. [11], distinguish between four LDES technologies only based on their roundtrip efficiency; Sepulveda et al. [19] identify a design space as the

Table 1
Selected KPIs computed, used to assess and cross-compare different TMES storage concepts.

КРІ	Units	Description
Roundtrip efficiency, η_{RT}	%	Ratio between electricity output and input over a complete charge/discharge cycle
Energy density,ED	kWh/ m ³	Ratio between electricity storage capacity and total storage volume
Power-specific cost,PC	USD/ kW	Capital expenditure (CAPEX) per unit storage power output
Energy-specific cost, <i>EC</i>	USD/ kWh	Capital expenditure (CAPEX) per unit storage capacity
Levelised cost of storage, LCOS	USD/ kWh	Fixed price of electricity required to fully cover storage costs over project lifetime, for a given discount factor

combination of charge and discharge power, storage capacity cost, charge and discharge efficiency requirements, with no link to specific technologies; Cardenas et al. [9] show how the needed storage capacity across different RES penetration levels differ depending on the overall storage efficiency, but do not associate values to technologies. On the other hand, except few recent works ([21;27]) the vast majority of studies on thermo-mechanical energy storage consider only short-duration applications and daily energy balancing [26]. This creates a dichotomy where, on the one hand, the target LDES performance is identified with no information on which technologies can realistically meet it, and, on the other hand, potential long-duration solutions such as TMES are not assessed for LDES applications. The present work aims at filling such knowledge gaps through the following novel contributions to the existing body of literature:

- A techno-economic assessment of TMES for LDES, in light of the individuated design space and in relation to incumbent storage technologies
- An understanding of the potential for LDES applications of novel TMES concepts recently emerged
- 3. A cross-comparison between traditional and novel TMES concepts.

In so doing, this work represents a first-of-a-kind analysis of the present and future techno-economic competitiveness of TMES for LDES. It initiates the discussion on the value of thermochemical energy storage for LDES and explores future development pathways with the potential of extending the role TMES has to play in grid decarbonisation, beyond current daily storage, to long-duration energy storage applications.

2. Methods

The six TMES solutions investigated in this study comprise both traditional and novel concepts. Among traditional technologies, ACAES, LAES and Brayton PTES were considered. For ACAES, a 2-stage compression/expansion layout with a parallel circuit for air intercooling and re-heating was chosen [28]. The studied LAES plant comprises 3stage compression and expansion processes, hot and cold recovery, as the layout adopted by She et al. [29], while PTES uses argon as the working fluid and indirect heat transfer with the hot and cold packed bed TES [30]. Process flow diagrams are reported in Fig. 2 (and further described in Section S1.1, S1.2 and S1.3 of the supplementary material), alongside those for the novel TMES considered (described in Section S1.4, S1.5 and S1.6). These latter all rely on open Brayton cycles for power conversion, but different reactions and associated process components for TCES were chosen. The reactions are, respectively, the reversible oxidation/reduction of MnO2/Mn2O3 [23] in the oxides energy storage (OES), the largely investigated CaCO₃/CaO reaction [31] in the carbonates energy storage (CES), and the hydration reaction Ca (OH)₂/CaO currently studied, among others, by the company SaltX [32], in the hydroxides energy storage (HES). Note none of the novel TMES concepts has currently been built as a complete system; not even at prototype scale: while CaO carbonation/calcination and hydration/ dehydration reactions have so far been investigated solely as TCES, the addition of a thermo-mechanical charge and discharge process for electricity storage purposes is a novelty of this paper.

Reduced thermodynamic models for each of the TMES concepts were set up by explicitly modelling the devices involved in conversion processes between power and thermal energy and thermal energy storage, as exemplified in Fig. 2. Although relying on the assumptions listed in Table 2, such bottom-up modelling approach allows grounding performance predictions on the key technical features of each system, and is therefore particularly suitable for preliminary performance assessment and quantitative cross-comparison. It is also worth mentioning the open challenge of ensuring multicycle conversion efficiency and stability of materials for TCES and anticipating novel TMES lifetime. Several publications discuss the former topic and approaches have been tested

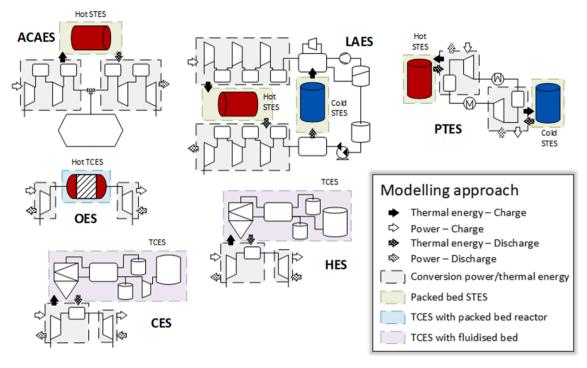


Fig. 2. Process flow diagram for the six TMES solutions considered. Dashed boxes illustrate the representation of each TMES as a combination of energy conversion and thermal energy storage processes, used for numerical model setup. Further discussion of the investigated processes can be found in Fig. S2-S7 of the supplementary material. Abbreviations: ACAES: adiabatic compressed air energy storage; LAES: liquid air energy storage; PTES: pumped thermal energy storage; OES: oxydes energy storage; CES: carbonates energy storage; HES: hydroxides energy storage; STES: sensible thermal energy storage; TCES: thermochemical energy storage.

Table 2
Major modelling assumptions in this work.

Assumption	Applied to equations
Steady-state conditions	1 to 20
Storage charging/discharging at rated conditions	1 to 20
Chemical reactions progress until thermodynamic equilibrium	12 to 20
Negligible pressure losses	1 to 6
Constant storage media properties	12 to 20
Sharp reaction front in packed bed reactors	14 to 15
Balanced hot and cold stream flows in the heat exchangers	7 and 8

which ensure over 1000 cycles without degradation [33]. However, the scope of this analysis is limited to studying the thermodynamic conditions under which TCES should operate and the subsequent technoeconomic system performance, leaving the engineering of suitable materials open to further research. A baseline 30 years lifetime for all concepts has therefore been assumed.

Models were implemented in MATLAB; the CoolProp library was used to predict fluid properties based on the implemented equation of state and an explicit formulation of the Helmholtz energy, as described in [34]; the HSC Chemistry 6.0 database was used to retrieve thermochemical properties for chemical reactions [35].

2.1. Conversion processes between power and thermal energy

Three main components enable conversion between electricity and heat in TMES: compressors, expanders and heat exchangers. For compressors and expanders, the approach proposed in [36] was followed, which conveniently models both volumetric devices and turbomachinery, based on device polytropic efficiency (η) and the definition of a thermal loss coefficient (ζ_C) for compressors and a gain (ζ_T) for turbines. The specific work of the i-th compression stage $(w_{C,i})$ and the outlet

temperature condition ($T_{out,i}$) were then computed as enthalpy (h) difference, based on the stage inlet temperature ($T_{in,i}$), the isentropic coefficient for the gas (γ) and the compression ratio (Π_C).

$$T_{out,i} = T_{in,i} \Pi_C^{\theta_C} \tag{1}$$

$$\theta_C = \frac{1}{\eta_C} \frac{\gamma - 1}{\gamma} (1 - \zeta_C) \tag{2}$$

$$w_{C,i} = \frac{h_{out,i} - h_{in,i}}{1 - \zeta_C} \tag{3}$$

Similar analysis enabled to compute the specific work produced, and the outlet temperature of the working fluid from each expansion stage:

$$T_{out,i} = T_{in,i} \Pi_T^{\theta_T} \tag{4}$$

$$\theta_T = -\eta_T \frac{\gamma - 1}{\gamma} (1 - \zeta_T) \tag{5}$$

$$w_{T,i} = \frac{h_{in,i} - h_{out,i}}{1 - \zeta_T} \tag{6}$$

Heat exchangers and intercoolers/reheaters were all modelled based on device heat transfer effectiveness (ε) and the assumption of balanced flows [37]; the thermal energy exchanged was computed as an enthalpy difference. Based on the process arrangement, the hot fluid of cold fluid temperatures (T_h and T_c , respectively) also represent the temperature of the streams exchanged with the thermal energy storage:

$$\begin{pmatrix} T_{h,out} \\ T_{c,out} \end{pmatrix} = \begin{bmatrix} 1 - \varepsilon & \varepsilon \\ \varepsilon & 1 - \varepsilon \end{bmatrix} \begin{pmatrix} T_{h,in} \\ T_{c,in} \end{pmatrix}$$
(7)

$$q_{HX} = h_{h,in} - h_{h,out} = h_{c,out} - h_{c,in}$$

$$\tag{8}$$

Ultimately, the suitable connection of compressors and heat exchangers (or turbines and heat exchangers) for each TMES concept

allows the forward and backward conversion between heat and electricity.

2.2. Thermal energy storage processes

Proper modelling of the thermal energy storage process was necessary for accurate estimation of plant performance, energy density and investment costs. In particular, the reversible chemical reactions involved in TCES are ruled by thermodynamic equilibrium so that, for any specified reactor pressure (p_r) , storage charging takes place if reactants temperature is above T_{eq} , or below for discharging [23]:

$$p_r = p_0 \exp\left(\frac{-\Delta H + T_{eq}\Delta S}{RT_{eq}}\right) \tag{9}$$

 ΔH and ΔS represent the enthalpy and entropy of reaction (per mole of gas) for the specific reactants-products group considered. Representative figures at high temperatures were computed following Hess law [38] and the tabulated values ΔH_0 and ΔS_0 at standard conditions, the stoichiometric coefficient (ν_i) for each compound (negative for reactants), and the specific heat capacity ($c_{p,i}$) of the chemical species involved, the latter being retrieved from the HSC Chemistry 6.0 database [35] as polynomial regression:

$$\Delta H(T_r) = \Delta H_0 \pm \sum_i \nu_i \int_{T_0}^T c_{p,i} dT \tag{10}$$

$$\Delta S(T_r) = \Delta S_0 \pm \sum_{i} \nu_i \int_{T_o}^{T} c_{p,i} \frac{dT}{T}$$
(11)

Three different approaches were used to model the thermal energy storage solutions considered, namely STES for ACAES, LAES and PTES, TCES with a packed bed reactor for OES and TCES with fluidised bed reactors for CES and HES.

2.2.1. Sensible thermal energy storage

The thermal energy storage volume (V_{TES}) was computed from the value of the thermal energy produced during the charging process ($\dot{Q}_{chr}t_{chr}$) and the associated temperature:

$$V_{TES} = \frac{\dot{Q}_{chr}t_{chr}}{\rho \frac{c_p}{MM}(1 - \sigma)\Delta T_{TES}}$$
(12)

 ρ and c_p represent, respectively, storage medium density and specific heat capacity, and ΔT_{TES} is the temperature difference between charged and discharged states. A 38 % void fraction (σ) was considered in the packed bed, based on previous analyses [39], and a heat loss coefficient Λ (0.08 %, from [40]) was associated to the STES, to represent storage standing losses as a percentage of the stored energy during idle time. Accordingly:

$$\frac{Q_{loss}}{\dot{Q}_{chr}t_{chr}} = 1 - exp(-\Lambda\varphi) \tag{13}$$

No standing losses were considered for TCES.

2.2.2. Thermochemical energy storage with packed bed reactor

The packed bed reactor in OES is filled with $\rm MnO_2/Mn_2O_3$ particles and has a twofold role: it enables reversible metal oxidation/reduction, but also acts as thermal energy storage by storing reaction heat; this is thoroughly explained in [23]. During OES charge, $\rm Mn_2O_3$ is reduced to $\rm MnO_2$ and heat is stored in the packed bed. This process was modelled by assuming sharp temperature and reaction fronts which move forward through the device, as the reaction progresses [41]. Therefore, air leaves the packed bed reactor at the discharged storage temperature ($T_{c.TES}$) first, and then at the equilibrium temperature conditions ($T_{eq,chr}$), until the reaction front reaches the packed bed outlet and the TCES is fully charged, reaching its maximum temperature ($T_{h.TES}$). During discharge,

 $\rm MnO_2$ is oxidised to $\rm Mn_2O_3$ and reaction heat is transferred to the air stream flowing through the packed bed. Now, the outlet air leaves the TCES first at temperature $T_{h,TES}$ and then at $T_{eq,dsc}$, until $\rm MnO_2$ has fully reacted.

Since air leaving the packed bed in OES is then expanded in the downstream turbine, the time (t_1 and t_2) over which each outlet temperature condition takes place during charge/discharge needed to be computed. An energy balance was imposed over the reactor for this purpose. As an example, Equation (14) and (15) refer to the storage charging process. The energy transported by the hot inlet air (left-hand side) provides, initially, the sensible contribution increasing packed bed temperature from $T_{c,TES}$ to $T_{eq,chr}$, as well as the required heat of reaction (Equation (14), right-hand side). Then, further heating only increases packed bed temperature up to the value $T_{h,TES}$, until the TCES is fully charged (Equation (15), right-hand side):

$$\dot{m}_{f} \frac{c_{p,f}}{MM_{f}} \left(T_{h,TES} - T_{c,TES} \right) t_{1} = V_{TES} \left(1 - \sigma \right) \left[\Delta H \frac{\rho}{MM} + \rho \frac{c_{p}}{MM} \left(T_{eq,chr} - T_{c,TES} \right) \right]$$
(14)

$$\dot{m}_{f} \frac{c_{p,f}}{MM_{f}} \left(T_{h,TES} - T_{eq,chr} \right) t_{2} = V_{TES} (1 - \sigma) \left[\rho \frac{c_{p}}{MM} \left(T_{h,TES} - T_{eq,chr} \right) \right]$$
 (15)

In the equations above, $\dot{m_f}$, MM_f and $c_{p,f}$ represent the mass flow rate, molar mass and specific heat capacity of the flow-through fluid (here, air), and MM is the molar mass of the solid reactant. Analogous energy balance equations were imposed for packed bed discharge.

Finally, storage volume was estimated, accounting for both a sensible and a chemical contribution to the storage capacity:

$$V_{TES} = \frac{\dot{Q}_{chr}t_{chr}}{\rho \frac{c_p}{MM}(1-\sigma)(T_{h,TES} - T_{c,TES}) + \Delta H \frac{\rho}{MM}(1-\sigma)}$$
(16)

2.2.3. Thermochemical energy storage with fluidised bed reactor

In the fluidised bed reactors, the below reversible reactions take place, respectively, in CES and HES:

$$CaO + CO_2 \leftrightarrow CaCO_3$$
 (17)

$$CaO + H_2O \leftrightarrow Ca(OH)_2$$
 (18)

Since reactants heat capacity is orders of magnitude lower than reaction enthalpy, an ideal recuperation process between products leaving the reactor and incoming reactants was assumed to model the fluidised bed reactors. This means the heat provided by the fluid in the open Brayton cycle during storage charge must be sufficient to sustain the chemical reaction, with no additional sensible contribution. Similarly, the full reaction heat generated during discharge, netting off heat transfer losses, is used to increase turbine inlet temperature before expansion. Additional thermal losses needed to raise reactants temperature to equilibrium conditions would have to be included, otherwise.

Outlet temperature for the air stream was computed from Equation (7) (with $T_{c,in} = T_{c,out} = T_{eq,chr}$ during charge, and $T_{h,in} = T_{h,out} = T_{eq,dsc}$ during discharge). The total thermal energy storage volume, in this case, accounted only for the chemical contribution, plus the extra volume required to store, respectively, CO_2 at 75 bar in CES, and water at ambient pressure in HES. The CO_2 and water amounts were computed from a mass balance, which includes the value of a conversion factor (*CF*) representing the reaction extent (20 % for carbonation [42] and 80 % for CaO hydration [43]) and the excess gas (*EG*):

$$V_{TES} = \frac{\dot{Q}_{chr}t_{chr}}{\Delta H \frac{\rho}{MM} (1 - \sigma)CF} + \frac{m_{CO_2/H_2O}}{\rho_{CO_2/H_2O}}$$
(19)

$$\dot{m}_{CO_2/H_2O} = (1 + EG) \frac{\dot{Q}_{chr}MM}{CF\Delta H} \left(\frac{MM_{CO_2/H_2O}}{MM} \right) \left(\frac{\nu_{CO_2/H_2O}}{\nu} \right)$$
 (20)

2.3. Estimation of plant investment costs and levelised cost of storage

Capital investment for each plant was evaluated from the cost functions for individual devices, namely compressors, heat exchangers, turbines, reactors, storage tanks, vessels and the air cavern, as well as costs of the storage media. Functions from the literature and material price from sellers reported in Table 3 were used for this purpose. So, for each considered process, the component size returned by the model for a 100 MW_e power output, together with the given storage duration were supplied as inputs to the cost functions. The total capital investment was then estimated by summing all the system components. On top of the mentioned costs, an additional 33 % on power-related costs was considered for LAES (based on [44]), covering mainly air liquefaction equipment, cryopumps and evaporator. For the other technologies, a 10 % surplus was added for auxiliaries.

Whilst acknowledging the adopted cost accounting based on preliminary design estimates may be accurate within a \pm 30 % range [45], this approach is deemed suitable for comparative analysis; a more accurate appraisal would require accounting separately for procurement and installation costs for each component, as well as engineering costs and contingencies [46]. It is also worth pointing out how cost functions in Table 3 assume a preliminary material selection that is consistent across all TMES technologies, although challenging operating conditions in terms of temperatures and pressures may require ad-hoc considerations to be evaluated in further detailed design studies. Cost figures were all adjusted to 2020 USD, using the CEPCI index and average yearly currency conversion factor.

Levelised cost of storage (LCOS) was then computed as a widespread metric which evaluates the fixed price of electricity required to fully cover project costs over a complete lifetime of Y years, when a discount factor d is considered [53]. Equation (21) expresses LCOS based on storage investment cost (CAPEX), annual costs (AC) – which includes operation and maintenance cost and electricity purchase cost $\left(\pi \frac{W_{out}}{\eta_{RT}}\right)$ – and the annual storage electricity output, W_{out} :

$$LCOS = \frac{CAPEX + \sum_{y=1}^{Y} \frac{AC}{(1+d)^{y}}}{\sum_{y=1}^{Y} \frac{W_{out}}{(1+d)^{y}}}$$
(21)

On top of investment costs, LCOS includes system lifetime, operational and maintenance costs, and most notably, the cost of charging the

TMES, which requires knowledge of plant duty cycle. This work considers full charge and discharge at rated conditions and the same charge/discharge time, separated by two idle times of equal length per cycle. Daily, weekly or monthly cycles were considered depending on the storage duration, as summarised in Table 4. For LCOS calculation, CAPEX and efficiency values output from the developed technoeconomic models were used for individual TMES. On the contrary, for the electricity purchase price, project lifetime, operation and maintenance cost a uniform distribution within a selected range was considered. A probabilistic approach inspired by McTigue et al. [27] was then followed, yielding multiple evaluations of LCOS by randomly picking each parameter value within its selected range (see Table S4). This approach allowed including the uncertainty associated with parameter estimation in the LCOS analysis. Ultimately, the annual energy output was computed as $W_{out} = W_{dsc}N$, where N is the number of yearly cycles.

3. Results and discussion

3.1. Techno-economics of thermo-mechanical energy storage for selected durations

Consensus exists for ACAES, LAES and PTES on the range and best values of operating parameters like cycle pressures and temperatures [21]. To explore the effect of operating parameters on novel TMES and illustrate the features of each concept, a preliminary optimisation of cycle charge and discharge pressure was undertaken for OES, CES and HES, maximising roundtrip efficiency. This same condition was found to also lead to the highest energy density and the lowest specific costs for the three technologies. Results for OES demonstrate performance increases monotonically with charge pressure due to overall higher

Table 4LDES duty cycle features considered for this analysis.

Cycles	Storage duration [h]	Idle time [h]	Number of cycles			
Daily	1–12	0–11	365p.a.			
Weekly	14–50	34–72	52p.a.			
Monthly	52-200	160-308	12p.a.			
Example of	Example of duty cycles					
$ au_{8h}$	Charge: 8 h; Idle: 4 h; Discharge: 8 h. N: 365					
$ au_{200h}$	Charge: 200 h; Idle 160 h; Discharge: 200 h. N: 12					

Table 3
Cost functions used to evaluate TMES investment cost. All values are expressed in 2017 kEUR.

Component	Cost function	Variable X	Notes	Ref
Compressors	$2035 \left(\frac{X}{10}\right)^{0.6}$	Power input, MW	Carbon steel	[44]
Turbines	$1002 \bullet 3 \left(\frac{X}{10}\right)^{0.67}$	Power output, MW	Carbon steel	[44]
Intercoolers and reheaters	$(1.3+1.88)65\frac{X}{1000}$	Heat transfer area, m ²	Carbon steel, $U=500 \text{ W/m}^2\text{K}$	[44]
TCES fluidised bed reactors, charge	193X ^{0.65}	Thermal input, MW	Calciner	[31]
TCES fluidised bed reactors, discharge	$3830 + 217X^{0.65}$	Thermal output, MW	Carbonator	[31]
Air cavern	$\left(\frac{7.5}{1.15}\right)X$	Capacity, MWh	Salt cavern	[47]
Liquid air tanks	$2760 \left(\frac{X}{1200}\right)^{0.6}$	Liquid mass, ton	Atmospheric pressure	[48]
Thermal storage tanks	563 + 0.22X	Tank volume, m ³	Large field tank	[44]
Pressure vessels	$\left(\frac{1.24}{1000}\right) \bullet 10^{\left[3.49 + 0.44 Log(x) + 0.11(Log(x))^2\right]}$	Vessel volume, m ³	Vertical	[49]
Gravel	$80\left(\frac{X}{1000}\right)$	Mass, ton	Material cost	[50]
MnO_2	$534\left(\frac{X}{1000}\right)$	Mass, ton	Material cost	[51]
CaO	$27.5\left(\frac{X}{1000}\right)$	Mass, ton	Material cost	[52]

temperature values in the TCES [23], while the optimum discharge pressure depends on the selected pressure value during charging. Further elaboration shows that suitably optimising discharge pressure can also mitigate OES performance sensitivity to changes in charge pressure. Indeed, roundtrip efficiency was shown to only drop by 10 % when halving charge pressure. On the contrary, for CES and HES, chemical equilibrium constraints the inlet temperature conditions before expansion. So, the optimal discharge pressure only depends on the equilibrium temperature and becomes a function of the particular material and reaction chosen for thermochemical energy storage. Values of 11.7 bar and 4 bar were found, respectively, for CES and HES, which reflect equilibrium temperatures of 900–1000 °C in the first and 500–550 °C in the second case (see Table S2 in the supplementary material).

With the optimised pressure values in Table S2, the technical performance of novel and traditional TMES concepts is compared in Fig. 3, while specific cost metrics factoring the procurement cost of individual devices and materials for each concept (see Section 2.3) are reported in Fig. 4 for τ_{8h} and τ_{200h} . A performance gap emerges between ACAES, LAES, PTES (with efficient devices) and novel TMES concepts based on thermochemical storage. None of the TMES options based on thermochemical storage reaches roundtrip efficiency higher than 30 %, even with enhanced component performance, mainly because a significant pressure difference between charge and discharge is needed for reactants to be, respectively, above and below the equilibrium temperature. Large process irreversibility is a consequence, so that nominal roundtrip efficiency values are 21 %, for OES and CES and 12 % for HES, i.e. well below indicated figures for most storage technologies [54]. However, the performance gap reduces at τ_{200h} .

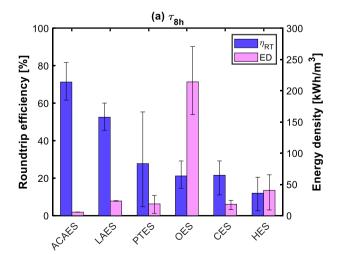
Conversely, novel TMES outperform traditional options in terms of energy density by up to 2 orders of magnitude, thanks to compact TCES. OES and HES, respectively, reach 214 and 41 kWh/m³, whereas LAES and PTES only 24 and 19 kWh/m³, and ACAES 6 kWh/m³. Despite carbonation yielding the highest reaction enthalpy among the reactions considered (\sim 10 MJ per m³ of solid reactant), CES energy density (18 kWh/m³) is comparable with TMES options relying on STES. Three conditions hinder storage compactness. First, the storage of gaseous CO₂ reactant accounts for 57 % of the overall storage volume (\sim 43500 m³) for τ_{8h} (H₂O storage in HES is only 37 %). Second, the storage of solid CaO at ambient conditions only leverages chemical energy, whereas, with a Δ T of 100 °C, 16 % more heat could be stored per unit volume. Third, the conversion efficiency for CaO in the carbonator is only 20 % [42]. Should conversion be increased to 80 % (as in HES), energy density

would reach 73 kWh/m 3 . Both the discussed sensible addition to thermal storage and the reaction conversion enhancement equally apply to HES. Hence, two key pathways for future technological improvement involve process layout configuration and material-scale studies.

Cost metrics in Fig. 4 significantly change between the investigated storage durations. Power equipment (mainly turbines and compressors) represents by far the biggest contribution (between 50 % and 83 %) to the investment cost at τ_{8h} . This is a major drawback for PTES and novel TMES relying on Brayton cycle, which require larger components as the overall net charging/discharging power is the difference between the compression and expansion work rates. However, for τ_{200h} , turbines and compressors represent only 8–17 % of the total investment cost for most TMES. Now 80-90 % is capacity equipment (i.e. vessels, STES or TCES) the only exception being CES, with prohibitive costs for pressurised CO₂ storage at large-scale – and Brayton-based concepts emerge more costcompetitive. PTES and OES become only 13 and 17 % more expensive than LAES, but, as error bars highlight, they can notably be cheaper for enhanced compressors/turbines performance. Among novel TMES solutions based on TCES, the use of CaO hydration/dehydration seems very well suited for LDES. Thermal energy storage compactness, cheap storage medium and liquid H₂O storage at ambient conditions concur in HES achieving the lowest investment with respect to OES and CES, under the assumed baseline device performance used for this study. Future device improvement captured by sensitivity analysis (i.e. enhancement of compressor and turbine polytropic efficiency and heat exchanger effectiveness) would result in up to 20 % roundtrip efficiency and further cut costs to 3235 USD/kW and 16 USD/kWh, which is less than the estimates for ACAES and, as discussed later, suits the design space identified for LDES [19].

3.2. Levelised cost of thermo-mechanical energy storage for selected durations

LCOS combines technology costs and roundtrip efficiency into a single metric [53], which is presented in Fig. 5. Results differ significantly with duration, mainly due to the variations in investment costs and roundtrip efficiency previously discussed, along with the different amount of energy shifted. Electricity purchase costs contribute by 66–77 % across most technologies to the overall LCOS breakdown, while CAPEX only represents 17–26 %, for τ_{8h} . Hence, high roundtrip efficiency is of primary importance to limit electricity expenses and should be the focus of future technology development; currently, traditional technologies (ACAES and LAES) appear more suitable than other TMES



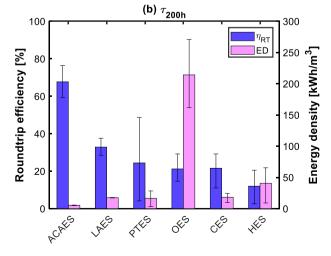


Fig. 3. Technical performance metrics for the TMES technologies investigated for τ_{8h} (a) and τ_{200h} (b). Error bars contain sensitivity analysis results where machine polytropic efficiency and heat exchanger effectiveness were varied between 0.8 and 0.9 and 0.87–0.97, respectively. Limited (28 % and 24 %) roundtrip efficiency for PTES depends on the low (0.85) machine efficiency in this study, but, under the high sensitivity to component performance shown by the error bars, values above 55 % are achieved.

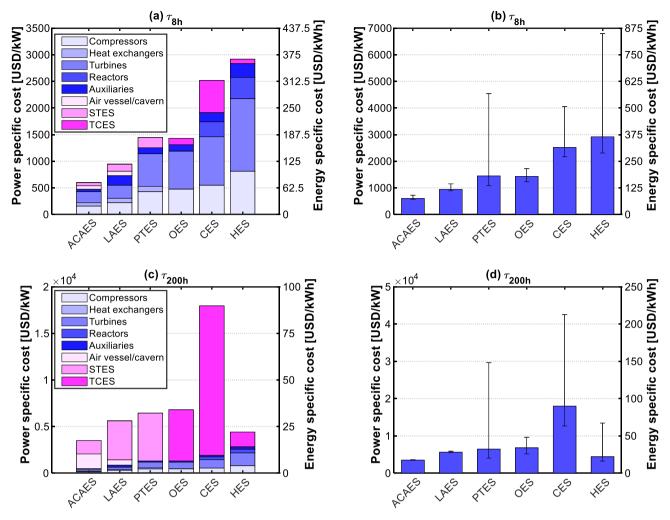


Fig. 4. Power- and energy-specific cost breakdown for the TMES technologies investigated for τ_{8h} (a) and τ_{200h} (c) and their variation with device characteristic parameters for τ_{8h} (b) and τ_{200h} (d). Error bars contain sensitivity analysis results where machine polytropic efficiency and heat exchanger effectiveness were varied between 0.8 and 0.9 and 0.87–0.97, respectively.

for short durations. For τ_{200h} , CAPEX (driven by capacity equipment costs) accounts for 46–61 % of LCOS across technologies (except for HES), which is a comparable share to that of electricity purchase costs. Under these conditions, novel TMES technologies seem to offer a favourable cost structure and are now less impacted by low efficiency. For instance, the same 10 % increase in cost and decrease in efficiency for OES results in similar LCOS variation (from 0.54 to 0.57 USD/kWh) at τ_{200h} , while for τ_{8h} 10 % efficiency reduction is much more impactful on LCOS (from 0.25 to 0.28 USD/kWh), than a 10 % CAPEX increase (LCOS 0.26 USD/kWh).

Another interesting consideration concerns OES, among novel TMES. Our results for τ_{200h} show it only requires 2 % of the storage volume of HES. Yet, OES in Fig. 5(d) presents ~ 3 times higher capacity cost than HES, due to the use of pressurised storage vessels. A different OES process layout could be thought where, rather than flowing through the packed bed reactor, the high-pressure working fluid only exchanges reaction heat with the TCES, through an intermediate indirect heat transfer step. This eliminates the need for TCES vessel pressurisation. In such a case, the estimation of TCES costs (obtained neglecting the small cost contribution from an additional heat exchanger and the performance penalty associated with indirect heat transfer) is only 32 % of the required investment for a pressurised TCES. OES total cost more than halves in this case (from 34 to 15 USD/kWh), yielding a 0.36 USD/kWh LCOS: the second-lowest value in this study and below predictions for power-to-H2-to-power pathways between 0.45 and 0.59 USD/kWh [54].

3.3. Effect of storage duration and idle time on techno-economic performance

As noted, TMES economics depends on storage duration. However, standing losses from STES occurring during idle time are the primary cause of roundtrip efficiency reduction when moving from τ_{8h} to τ_{200h} , which ultimately affects TMES technical performance. In this regard, ACAES suffers the least because multistage compression allows storing 18 % excess heat on top of the amount needed for expansion; that surplus is only 11 % for PTES. Accordingly, roundtrip efficiency starts departing from design values after ~ 2 days of idle time ($\varphi = 40$ h) for PTES and ~ 1 week ($\varphi = 140$ h) for ACAES. In LAES, the effect is immediate - as the recovery of cooling available from the process of air evaporation during discharge is always insufficient to fully liquefy air during charge [55] - and the most significant due to the associated liquid yield reduction, as noted in [39]. For τ_{200h} , roundtrip efficiency in Fig. 3 drops to 68 %, 33 % and 24 % for ACAES, LAES and PTES, from 71 %, 53 % and 28 %, respectively. Storage concepts based on TCES instead are loss-free and display no technical metrics detriment. Contrary to power-specific cost, energy-specific cost from Fig. 6 decreases with longer durations. The previously discussed favourable cost structure of novel TMES results in HES notably becoming the second cheapest solution above τ_{100h} . The high share of power-related CAPEX at τ_{200h} (45) %) means additional HES cost reduction can be achieved by further extending duration above 200 h, (see slope of the HES curve in the detail

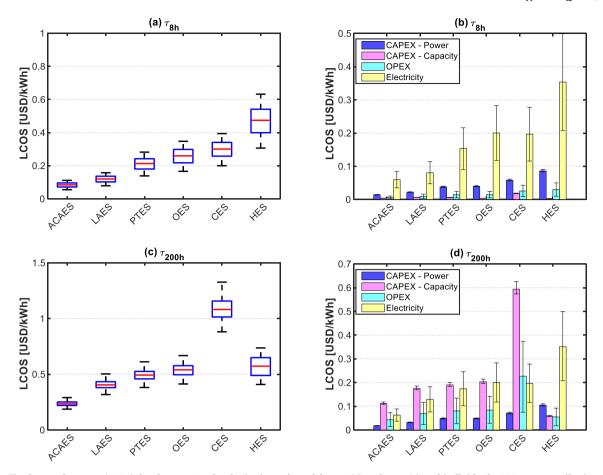


Fig. 5. Levelised cost of storage (LCOS) for the TMES technologies investigated for τ_{8h} (a) and τ_{200h} (c) and individual LCOS cost contributions for τ_{8h} (b) and τ_{200h} (d). Boxplots represent the median (red line), 25th and 75th percentiles (box) of the LCOS distribution obtained by selecting rated TMES cost and roundtrip efficiency, and an interval for the electricity cost, lifetime and O&M costs, as in [27]. In figures (b) and (d) bars report the median and error bars the extreme values from the analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of Fig. 6(a)). PTES becomes more economically viable than OES above τ_{150h} . Uncertainties on the actual future device costs and system performance may shift the exact value of storage duration where a shift in the cost ranking between TMES technologies takes place; nonetheless, the observed trends still apply. Additional sensitivity results on storage power output are reported in Table S5 of the supplementary material.

LCOS results in Fig. 6 are overlaid with those of incumbent technologies that are suited for daily and monthly cycles, namely Li-ion batteries and power-to- $\rm H_2$ -to-power [56]. For daily cycling, a crossover point exists above which LAES and CAES become more cost-effective than Li-ion batteries. In the present analysis, this happens around 2–4 h duration, for a 100 MW_e power output. Although several Li-ion battery projects with over 10 h duration are in operation world-wide [57], at sufficiently large scales and above 4 h duration, TMES should be preferred instead. 8 h duration or more is needed for PTES to be cost-competitive (others suggest 6 h [27]). Novel TMES based on TCES is less attractive on an LCOS basis, both for daily and monthly cycling, as penalised by the low roundtrip efficiency.

Fig. 6 conveys a different picture of which technology to prefer, depending on the chosen indicator being LCOS or investment costs. Such finding demonstrates how technology prioritisation depends on the assumptions about storage duty cycle and electricity price, both implicit in LCOS calculation (see Section 2.3 and S2.2), and whose representativeness of real operation can be uncertain, especially for LDES [58]. In particular, free (i.e. zero-cost) electricity can incur for charging storage with otherwise curtailed energy (sometimes even avoiding a curtailment penalty [59]); fewer cycles for LDES would favour low-CAPEX, low-efficiency solutions over capital-intensive ones. Studies describe a

monotonical increase of curtailment at high RES shares [60] and emphasise the limited importance of roundtrip efficiency at the first stages of storage deployment, i.e. when large portions of otherwise wasted energy are available [61]. Therefore, storage ranking based on LCOS and capacity-specific cost should be regarded as two limiting cases. The more the zero-cost electricity assumption is representative or the fewer the yearly cycles, the more HES becomes competitive for LDES. So, different storage technologies might become the most cost-effective solution to drive different stages of the energy transition. In addition, a lower number of stable cycles for the TCES material may be acceptable for LDES application, as fewer yearly charge/discharge cycles are involved.

On the other hand, storage roundtrip efficiency values certainly affect the competitiveness of LDES with alternative storage technologies and flexibility sources. This analysis proves that standing losses of TMES are negligible for short energy storage duration (8 h) but should be accounted in TMES assessment for LDES applications [58]. Fig. 6(b) and (c) show the range 4–150 h can be the recommended ideal scope where TMES offer the most cost-effective storage solution. Chemicals should be preferred at longer storage duration and technologies with lower power costs – like batteries – below 4 h. Indeed, a 52 h Li-ion battery with the cost and technical parameters used here would yield a 3.56 USD/kWh LCOS and the use of $\rm H_2$ for 6 h storage 0.44 USD/kWh. Clearly, selecting the best storage solution ensures lower investment requirements for the energy transition.

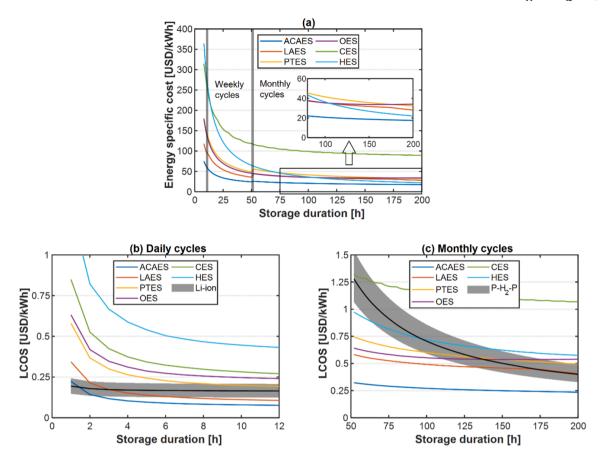


Fig. 6. Energy-specific cost (a) and levelised cost of storage (LCOS) for the TMES concepts investigated under daily (b) and monthly cycles (c). LCOS figures are compared with Li-ion battery (cost components 63 USD/kW and 232 USD/kWh, roundtrip efficiency 90% [27]) in (b) and power-to- H_2 -to-power (cost components 6380 USD/kW and 0.16 USD/kWh, roundtrip efficiency 41% [56]) in (c). The confidence range includes the LCOS distribution obtained using a range for the electricity cost, lifetime and O&M, as in [27].

3.4. The future potential of TMES for long-duration energy storage applications

To conclude, results from this study have been used to benchmark TMES solutions at 200 h duration against the target design space for LDES applications in Fig. 7. Energy-specific cost is the designated indicator in line with the scientific practice for LDES, where capacity costs are significantly more influential than power costs in the assessment [18]. Additionally, the use of energy-specific costs averts the aforementioned drawbacks of LCOS, linked with uncertainties on electricity price [62] and storage remuneration potentially coming from additional energy and reserve services [63].

With increased RES penetration, storage power requirements increase linearly while capacity exponentially [64]. Li-ion technology has proven viable for short-duration applications, but it is rarely cost-effective above 6 h, given the need for incremental units of duration [65]. Other thermochemical options, such as flow batteries, can store energy up to 80 h with minimal standing losses [66] and may achieve a well below 10 USD/kWh cost for the chemicals alone, but system cost is 1 order of magnitude higher [67]. On the contrary, cost estimations for TMES are consistent with the highly project-specific range for pumped hydro energy storage (5–100 USD/kWh) but without associated geographical restrictions to deployment [56].

In stark contrast to 88 % of the new global storage capacity addition in 2016 being Li-ion batteries [68], and given raw material price predictions of nickel-manganese-cobalt falling only to 124 USD/kWh [69], the world must turn to other technologies delivering LDES services. Still acknowledging economic results are subject to uncertain future developments through the energy transition, this work shows that ACAES

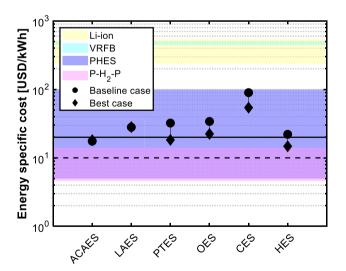


Fig. 7. Target design space for LDES: 200 h duration TMES and other incumbent storage options (cost range from [56]). Circles represent baseline case results obtained with 0.85 machine efficiency and 0.92 heat exchanger effectiveness. Diamonds denote best-case results obtained with 0.95 machine efficiency and 0.97 heat exchanger effectiveness. LDES cost targets from [19] are drawn as black lines: 20 USD/kWh (solid) to yield 10 % lower system adaptation cost and 10 USD/kWh (dashed) to start displacing firm generators.

can already meet the target cost for LDES even with the assumption of current device performance. However, when considering the effect of learning as future technological advancement [70], also PTES and HES are predicted to achieve costs below 20 USD/kWh. HES becomes the cheapest TMES solution for LDES and is competitive with power-to-H2-to-power pathways at ~ 14 USD/kWh. It may be complementary were future business cases to favour the sale of H2 as a commodity or a heat carrier [58] or should the countless H2 storage options reveal lower efficiency than expected. OES may reach the same or even better economic values than HES if the TCES can be indirectly coupled to the process via an intermediate heat transfer step, thus removing the need for a pressurised vessel.

4. Conclusion

This study thoroughly characterised all the main relevant existing and emerging TMES solutions for future applications as LDES. Results show traditional TMES (mainly ACAES and LAES) are best suited for τ_{8h} , while ACAES also meets the cost targets for LDES. They also suggest caution in projecting the technical performance obtained for short durations (e.g. τ_{8h}) to LDES assessment, especially for traditional TMES affected by standing losses.

Novel TMES technologies featuring storage compactness, limited losses and cheap storage materials represent a promising proposition for LDES, with lower efficiencies offset by a favourable investment cost structure with small capacity contributions. Particularly, the use of CaO hydration/dehydration and metal oxidation/reduction reactions seem promising pathways to develop TMES based on TCES. Future improvements for the development of these technologies are needed across material, device and process scales to: (i) achieve high thermochemical reaction conversion and cyclic stability; (ii) reduce the use of pressure vessels; and (iii) maximise the internal heat recovery within the process. Under these premises and with enhanced components efficiency, HES and OES may align with LDES cost targets and possibly become the cheapest TMES options for LDES. They could offer a cost-effective storage solution up to and potentially above 200 h duration, thus complementing H2 role in bringing relief to the power balancing needs of future low-carbon energy systems. This paper proposes the wideranging research advancements needed to make this happen.

CRediT authorship contribution statement

Andrea Vecchi: Methodology, Software, Formal analysis, Writing – review & editing, Visualization. **Adriano Sciacovelli:** Methodology, Supervision, Project administration, Writing – review & editing, Funding acquisition.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apenergy.2022.120628.

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