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Comparative sustainability study of energy storage technologies using data envelopment analysis



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ABSTRACT

The transition to energy systems with a high share of renewable energy depends on the availability of technologies that can connect the physical distances or bridge the time differences between the energy supply and demand points. This study focuses on energy storage technologies due to their expected role in liberating the energy sector from fossil fuels and facilitating the penetration of intermittent renewable sources. The performance of 27 energy storage alternatives is compared considering sustainability aspects by means of data envelopment analysis. To this end, storage alternatives are first classified into two clusters: fast-response and long-term. The levelized cost of energy, energy and water consumption, global warming potential, and employment are common indicators considered for both clusters, while energy density is used only for fast-response technologies, where it plays a key role in technology selection. Flywheel reveals the highest efficiency between all the fast-response technologies, while green ammonia powered with solar energy ranks first for long-term energy storage. An uncertainty analysis is incorporated to discuss the reliability of the results. Overall, results obtained, and guidelines provided can be helpful for both decision-making and research and development purposes. For the former, we identify the most appealing energy storage options to be promoted, while for the latter, we report quantitative improvement targets that would make inefficient technologies competitive if attained. This contribution paves the way for more comprehensive studies in the context of energy storage by presenting a powerful framework for comparing options according to multiple sustainability indicators.

1. Introduction

Electricity is among the cornerstones for most economic activities and human living conditions [1]. Electricity systems link generators with consumers through transmission and distribution grids. Traditionally, secure electricity systems were designed based on key technical parameters such as stability, flexibility, resilience, adequacy, and robustness [2]. This, among other factors, led to the widespread deployment of dispatchable technologies such as those based on fossil fuels or nuclear energy [3]. More recently, environmental concerns have pushed the adoption of various clean and renewable electricity production technologies [4]. However, while conventional generation technologies can easily adapt to the inherent fluctuating demand, renewable sources like solar and wind are intermittent, unpredictable, and uncertain, therefore compromising the system capacity to match supply with demand [5].

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One of the most promising solutions to rapidly meet the electricity demand when the supply comes from non-dispatchable sources is energy storage [6,7]. Electricity storage technologies convert the electricity to storable forms, store it, and reconvert it to be released in the network when needed [8]. Electricity storage can improve the electricity grid's reliability, efficiency, safety, security, and stability [9,10] while bringing down the cost of electricity supply [11] through the reduction of the number and duration of costly electrical interruptions [12]. In a context where global power generation with renewable technologies is expected to rise from 25% in 2020 to 86% in 2050 [11], it is not surprising that energy storage is deemed key to achieving the energy sector's decarbonization [13].

Energy storage technologies can be classified according to their functions, the storage duration, and the form of stored energy [14], with no single technology performing well in all situations [9]. For instance, large-scale mechanical energy storage options can shift a large volume of electricity from one time to another, while batteries are rapidly responding to a signal [11]. Thus, a myriad of energy storage technologies with different characteristics are and will be needed to provide service in different applications [14].

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Nomenclature

a .	
δ^*	Supper efficiency of DMU _o
ε	Non-Archimedean infinitely small value
λ	Intensity vector that reports the weight of each DMU in the virtual DMU
Λ	Intensity vector that reports the weight of each DMU in the virtual DMU
T^*	Efficiency of the DMU _o
CG	Coal Gasification
CCS	Carbon Capture and Storage
DEA	Data Envelopment Analysis
DEDLF	Delivered Energy During Lifetime
DOD	Depth Of Discharge
ED	Energy Density
FEDLF	Feed Energy During Lifetime
FTEJ	Full Time Equivalent Jobs
GWP	Global Warming Potential
LCOE	Levelized Cost Of Energy
LHV	Lower Heating Value
m	Number of inputs
NC	Number of Cycles during lifetime of technology
RE	Renewable Energy
REC	Rated Energy Capacity
RTE	Round-Trip Efficiency
WE	Water Electrolysis
WS	Water Splitting
s ₁	Number of desired outputs
s ₂	Number of undesired outputs
S _i -	Slack of inputs in SBM model
S _r ^g	Slack of desired outputs in SBM model
Sr ^b	Slack of undesired outputs in SBM model
t	Charnes-Cooper linear transformation coefficient
Xio	Input i related to the DMU ₀
X	Matrix of inputs
y _{ro} ^g	Desired output r related to the DMU
y _{ro} ^b	Undesired output r related to the DMU_0
Yg	Matrix of desired outputs
Y ^b	Matrix of undesired outputs
	•

Abbreviation and full name of fast-response energy storage technologies

Short-term

FlywheelsFlywheelsSMESSuperconducting Magnetic Energy Storage

Medium-term

FB-VR	Flow battery - Vanadium Redox
FB-ZB	Flow battery - Zinc Bromine
LA	Lead-Acid
Li-ion	Lithium-ion
Li-Fe-Ph	Lithium-Ferro-Phosphate
Li-Ni-Mn-Co	Lithium-Nickel-Manganese-Cobalt
Na-Ni-Cl	Sodium-Nickel-Chloride
Na-S	Sodium-Sulphur
Ni-Cd	Nickel-Cadmium

Abbreviation and full name of long-term energy storage alternatives

H ₂ , CG	Power to H_2 , Coal Gasification
H ₂ , CG-CCS	Power to H ₂ , Coal Gasification with carbon capture and storage (CCS)
H ₂ , SMR	Power to H ₂ , Steam Methane Reformation
H ₂ , SMR-CCS	Power to H ₂ , Steam Methane Reformation with CCS
H ₂ , WE-Grid mix	Power to H ₂ , Water Electrolysis by Grid mix
H ₂ , WE-Hydropower	Power to H ₂ , Water Electrolysis by Hydropower energy
H ₂ , WE-Solar	Power to H ₂ , Water Electrolysis by Solar energy
H ₂ , WE-Wind	Power to H ₂ , Water Electrolysis by Wind energy
H ₂ , WSCL	Power to H ₂ , Water Splitting by Chemical Looping
NH ₃ , SMR	Power to NH ₃ , Steam Methane Reformation
NH ₃ , SMR-CCS	Power to NH ₃ , Steam Methane Reformation with CCS
NH ₃ , WE-Grid mix	Power to NH ₃ , Water Electrolysis by Grid mix
NH ₃ , WE-Hydropower	Power to NH ₃ , Water Electrolysis by Hydropower energy
NH ₃ , WE-Solar	Power to NH ₃ , Water Electrolysis by Solar energy
NH ₃ , WE-Wind	Power to NH ₃ , Water Electrolysis by Wind energy
NH ₃ , WSCL	Power to NH ₃ , Water Splitting by Chemical Looping
1113, WOOL	i ower to ming, water opinting by chemical hooping

To facilitate the development and deployment of energy storage technologies, these must satisfy economic, environmental, and technical targets [5]. For instance, the European Association for Storage of Energy, in their 2017 roadmap, suggested that storage technologies should aim to achieve a levelized cost of stored energy lower than the levelized cost of energy appointed to other flexibility options such as grid upgrades or flexible generators [15]. Meanwhile, the 2030-plan for stationary energy storage systems aims at the cost of $\approx 0.05 \text{ €/(kWh, cycle)}$, in addition to achieving 10000 cycles durability and 20 years lifetime [16]. Further cost reductions besides technical and/or environmental improvements could make these technologies even more attractive. Therefore, gathering quantified and updated information about energy storage technologies is one of the first tasks needed to pave the way for their development and widespread application.

Several works have compared energy storage technologies based only on economic, technical [17], or environmental aspects [18]. For instance, Koohi-Fayegh et al. compared the performance of fast-response storage technologies in frequency regulation applications based on their energy and power density, cycle efficiency, lifetime, and capital costs [19]. A review concerning the life cycle cost of energy storage technologies was presented by Zakeri and Syri [20], including elements such as capital, operational, maintenance, and replacement costs. They concluded that the power conversion system and the energy storage unit section strongly affect an energy storage system's technical and economic performance [20].

On the other hand, some works shifted the focus from economic and technical considerations to the environmental implications of storage technologies. In this context, a cradle-to-gate life cycle assessment (LCA) study of vanadium redox flow batteries and zinc-cerium batteries illustrated that the electrolyte production process contributes significantly to most impact categories [21]. A similar conclusion was reported for lead-acid, lithium-manganese, and lithium-iron-phosphate batteries [22].

Very few studies considered environmental aspects and other sustainability dimensions simultaneously, and in case they did, it was limited to only one or a few storage technologies. Stougie et al. employed a multi-dimensional life cycle assessment for analyzing the sustainability of five different energy storage units considering economic and environmental criteria, in addition to each technology's total cumulative exergy loss [23]. Another multi-objective optimization was reported by Li et al. to select energy storage systems. They used economic and environmental objectives and technical constraints, finding flow batteries and hydrogen energy storage as the optimal solutions for emerging distributed energy systems [24]. Kapila et al. compared energy, life cycle greenhouse gas emissions, and costs for large-scale mechanical energy storage systems. Their results revealed that the unit's energy consumption (i.e., amount and energy source) is more relevant than the construction material considering greenhouse gas emissions from a life-cycle perspective [25].

Going beyond previous research, this contribution aims to provide a comprehensive assessment of a wide range of energy storage technologies (11 fast-response and 16 long-term storage options) considering a total of six key performance indicators that allow covering the three sustainability dimensions concurrently (*i.e.*, economic, environmental, and social perspectives). To ensure a fair comparison, technologies are first classified into two clusters according to their response time. In addition, we analyze for the first time the sustainability performance of ammonia as an energy storage material. We include the operation phase of storage technologies within the scope of our work, thus advancing previous research in both depth and breadth.

Comparing options in terms of multiple sustainability indicators is a challenging task calling for multicriteria decision-making methods. Among available tools, we resort to Data Envelopment Analysis (DEA) [26] because it can combine multiple indicators into a single score, avoiding the definition of subjective weights between the indicators. The uncertainty associated with the data used for the analysis is considered to draw robust and reliable conclusions. In the past, DEA has been widely used to investigate and compare engineering systems for the energy sector, such as nuclear energy [27], thermal powerplants [28], power industries [29], energy-consuming equipment [30], renewable energy production [31–37], and the electricity mix in European countries [26]. Nevertheless, to the best of the authors' knowledge, there is only one previous work on applying DEA to benchmark energy storage technologies. It compares pumped hydro storage, compressed air energy storage, lead-acid battery, and lithiumion battery using sustainability indicators and employing DEA, reporting lithium-ion battery as the most efficient case [38].

The analysis carried out first grades technologies within each cluster as either efficient or inefficient, providing evidence to support policies promoting the most appealing technologies to underpin the sustainability transition. For inefficient technologies, quantitative guidelines regarding directions and targets for improvement are provided. This valuable information will be key for technology developers to push their designs to the frontier of the best-performing options available in the market.

As will be further detailed later in the manuscript, the results obtained suggest that flywheel, nickel-cadmium, lithium-ion, and sodiumsulfur batteries are the preferred options among fast-response technologies, while green ammonia from water electrolysis powered by solar energy is ranked first between long-term storage options. Other efficient options for the later cluster are green hydrogen from solar and wind.

The remaining of this contribution is structured as follows. The methodology applied for the multicriteria assessment of energy storage technologies is described in the second section, while the results are presented and discussed in the third section. Finally, the conclusions drawn on how these results can be used for effective policymaking are presented in section four.

2. Methodological approach

In this contribution, we use DEA to compare the performance of energy storage technologies through the lens of sustainability (i.e., economic, environmental, and social perspectives). DEA is a linear programming method that allows measuring the relative efficiency of a set of homogenous entities (called Decision-Making Units, DMUs) in transforming one or several input(s) into one or several output(s) [39]. Producing more outputs using fewer inputs is the general criterion for higher efficiency (see Eq. (1) [40]), yet if some outputs are undesirable (e.g., carbon emissions), producing lower amounts of them is desired [41].

$$efficiency = \frac{weighted sum of outputs}{weighted sum of inputs}$$
(1)

In the context of this contribution, we model energy storage technologies as DMUs consuming m inputs and producing s_1 desired outputs and s_2 undesired outputs. The value of inputs and outputs for each DMU are obtained from different sustainability indicators, which are classified in these three categories depending on whether they are resources "consumed" by the technology (i.e., inputs, e.g., water), beneficial outputs from the process (i.e., desirable outputs, e.g., the creation of new job opportunities) or outputs that are not desired (i.e., undesirable outputs, e.g., polluting emissions to air). The DMUs considered are presented in Section 2.1, while details on inputs and outputs are provided in Section 2.2.

With input and output values at hand, an efficiency score will be obtained for every DMU by solving a DEA model for each of them. The model will return an efficiency score of 1 if the DMU is efficient, that is, if and only if there is no other DMU with lower or equal levels of inputs and undesirable outputs while simultaneously achieving greater or equal output levels.

Note that, in DEA, the efficiency score is given by a weighted (i.e., linear) combination of indicators, which is a common approach also used in other multicriteria decision-making methods and algorithms and



Fig. 1. Classification of the energy storage technologies (or DMUs under the concept of DEA) considered. SMES: Super Magnetic Energy Storage, FB: Flow Battery, VR: Vanadium Redox, ZB: Zinc-Bromine, LA: Lead-Acid, Li-Fe-Ph: Lithium-Ferro-Phosphate, Li-Ni-Mn-Co: Lithium-Nickel-Manganese-Cobalt, Na-Ni-Cl: Sodium-Nickel-Chloride, Na-S: Sodium-Sulfur, Ni-Cd: Nickel-Cadmium, CCS: Carbon Capture and Storage, SMR: Steam Methane Reformation, WE: Water Electrolysis, WS: Water Splitting.

in environmental impact metrics such as the EcoIndicator99 [42]. This approach is not always underpinned by a physical interpretation but is still useful for estimating technological options' relative ranking. In addition, the use of DEA avoids the need to define these weights in advance, thus preventing a subjective bias in the assessment of the different technological options. This is equivalent to implicitly assuming that all the inputs, desired outputs, and undesired outputs are equally important. The case where a higher weight is assigned to environmental indicators is later analysed in Appendix A.1. Specifically, we use a unit-invariant model that makes it possible to combine different indicators without their units altering the final efficiency score obtained [43]. Further, the monetized analysis, presented in Appendix A.2, translates all the indicators to their monetary values before combining them.

2.1. DMU modelling

Storage technologies can be classified based on the duration and frequency of power supply, and each class has its particular characteristics and applications [44]. To prevent unfair comparisons across classes, technologies are grouped into fast-response (including shortand medium-term storage technologies) and long-term (Fig. 1), considering their response time. H₂ produced using coal gasification is considered in our study, while NH₃ produced from such H₂ is discarded due to the lack of reliable data. Except for it, all the other considered alternatives are the same for H₂ and NH₃.

Fast-response technologies show response times ranging between a few seconds to a few hours, reason why these technologies are typically used for voltage stabilization, frequency control, peak shaving, load leveling, and daily storage. They should be able to offer several cycles during a day and, therefore, the number of cycles during their lifetime and their power and energy density are very relevant aspects [2]. This cluster includes two short- and nine medium-term technologies. Flywheel and super magnetic energy storage (SMES) are short-term storage technologies, while vanadium redox flow battery (FB-VR), zinc-bromine flow battery (FB-ZB), leadacid (LA), lithium-ion, lithium-Ferro-phosphate (Li-Fe-Ph), lithiumnickel-manganese-cobalt (Li-Ni-Mn-Co), sodium-nickel-chloride (Na-Ni-Cl), sodium-sulfide (Na-S), and nickel-cadmium (Ni-Cd) batteries are the options considered as medium-term energy storage technologies. There are various types of lithium-ion, redox flow, and sodium batteries with different battery characteristics regarding the materials used for the electrolytes and electrodes. For instance, a graphite-based anode, a cathode made using a lithium metal oxide (e.g. Li-Fe-Ph, Li-Ni-Mn-Co, etc.), and an electrolyte are the main components of a lithium-ion battery [9]. Significant research efforts are taking place to identify the material combination leading to the best battery characteristics in each case. All these DMUs are entirely based on data found in the literature (see supplementary information, section B). Pumped hydro storage and compressed air energy storage are two medium-term storage technologies not considered in this contribution since they are large-scale storage options and including them in the analysis could lead to unfair comparison.

On the other hand, long-term storage alternatives are usually used for dark-calm control during the no sun-no wind weeks in cold regions, prioritizing a low self-discharge rate. Since they have a very high energy



Fig. 2. Flow diagrams for H_2 and NH_3 production. Grey H_2/NH_3 : H_2 or NH_3 produced using fossil fuels, Blue H_2/NH_3 : H_2 or NH_3 produced using fossil fuels and the emitted carbon captured by CCS, Green H_2/NH_3 : H_2 or NH_3 produced using renewable energy sources.

density and acceptable reliability for long-term applications, they have strategic importance. We consider a total of 16 long-term energy storage alternatives (i.e., DMUs under the concept of DEA) for long-term storage, which result from combining two energy carriers (namely H_2 and NH_3) with the different production processes (e.g., hydrogen obtained from steam methane reforming with carbon capture or without it). Data for these DMUs is not readily available in the literature, so we modeled them as follows.

Hydrogen can be obtained using a variety of fossil fuels and renewable energy sources, which will result, in turn, in multiple DMUs (e.g., H₂ from coal gasification, H₂ from water electrolysis using solar energy, etc.). Renewable (or green) hydrogen is produced from water electrolysis powered by renewable energy sources. Blue hydrogen refers to the hydrogen produced using fossil fuels and a carbon capture and storage (CCS) unit used to capture carbon emissions [45]. The carbon captured during this process can be used in applications like enhanced oil recovery [46] or valuable chemical production [47]. Hydrogen produced using fossil fuels without capturing the associated carbon emissions is called grey hydrogen (Fig. 2) [45]. The hydrogen produced can be used as a standalone energy storage alternative or as raw material for ammonia production. For the former, three conversion steps are required. First, hydrogen is compressed using a reciprocating compressor with ~90% efficiency. Then, the compressed gas is stored with 98% efficiency. Finally, hydrogen can be reconverted back to electricity using different alternatives: an open cycle gas turbine with 35% efficiency, a combined cycle process with 60% electricity generation efficiency [48], or a fuel cell with about 20-50% efficiency [49]. Intending to capture all these nuances, we assume the widest range (i.e., 20-60% conversion efficiency) and use its average as the nominal case; this results in 40% efficiency for this final step. Therefore, the overall conversion efficiency of 35% is obtained for hydrogen (i.e., 90% 98% 40%).

In case hydrogen is used as a raw material to produce ammonia through the Haber-Bosch process [50], several DMUs with similar color codes as for hydrogen (i.e., blue, green, and grey ammonia) will result, depending on the energy source and hydrogen production process. Converting hydrogen to ammonia requires an additional air separation unit to obtain N_2 from the air, in addition to the other process units included

in a typical Haber-Bosch process, such as separation equipment [51]. Based on experimental studies, 22 kg/s NH₃ are produced using 4 kg/s H₂ [50]. According to the reaction's stoichiometry, 22.67 kg ammonia can be achieved using 4 kg H₂. Therefore, we consider a conversion of 97% (22/22.67) for this step. This value is consistent with other studies reporting conversions above >90% [52] or even 98% [53]. Ammonia is then separated from the remaining reactants with a recovery of 97% (>95% [52]). Ammonia storage efficiency is the same as hydrogen, while different options arise for converting ammonia into electricity. For instance, one can use fuel cells at efficiencies varying between 37% [54] and 54% [55], while a solid oxide fuel cell-gas turbine combined cycle of ammonia can achieve efficiencies of 68% [56]. For the sake of simplicity, we consider the average of the widest range based on the reported data (i.e., 37%-68%), which is 53% for the efficiency of the ammonia reconversion step. Therefore, the overall efficiency of storing and transforming ammonia to electricity is about 52% (i.e., 98% 53%).

Independent DEAs will be carried for each cluster, where different inputs and outputs will also be assessed (i.e., gravimetric energy density will be assessed for fast-response technologies, but not for long-term energy storage alternatives). The specific list of inputs and outputs (desired and undesired) included in each analysis is described in detail in the next section. Note that some of these technologies can be used for different purposes (e.g., flywheel for power quality control and distributed generation support), potentially achieving different performance (i.e., indicator values) in each case [20]. For simplicity, we assume only a certain value based on their more common application (e.g., power quality control in the case of flywheels). The explicit consideration of the uncertainty associated with the data is expected to cover all the potential inaccuracies stemming from the different modelling choices and assumptions, leading to robust conclusions (see Section 2.4).

2.2. Inputs and outputs

A total of six performance indicators are selected to cover the three sustainability dimensions. These indicators are classified as inputs and desired or undesired outputs of the DMUs in each cluster, as reported in Fig. 3. The only difference between the indicators considered in the



Fig. 3. Diagram of DMUs for each of the clusters: fast-response and long-term storage systems. LCOE: Levelized cost of energy. GWP: Global warming potential. In practice, the inverse of energy density (expressed in kg/GJ) is used instead of the normal indicator to align with the input concept of "the less, the better".

two clusters is that the gravimetric energy density (GJ/kg) is used for fast-response technologies and not for the long-term. The reason is that size is a crucial factor for fast-response technologies, which are many times used for portable applications [7], while it is not so determinant for long-term options.

Data for fast-response technologies are mainly gathered from literature, using the average value among available data where more than one reference is available for a certain indicator. Data gaps are filled based on average values of similar technologies (e.g., the LCOE reported for Liion is used for Li-Fe-Ph). All the indicators in this cluster are downscaled to unitary values through their division by the amount of energy delivered during the lifetime of technology, e.g., they are expressed per GJ of energy delivered (see supplementary information, section A1 for details on the calculation of the delivered energy for fast-response technologies). Note that these values indirectly consider factors such as dedicated lifespans, number of cycles, energy capacities, round-trip efficiency, and depth of discharge.

For long-term storage alternatives, hydrogen data are collected from the relevant literature, with missing data replaced by the average values of similar options (e.g., water consumption for hydrogen production using coal gasification is considered the same as steam methane reformation (SMR), since both are based on fossil fuels process). Likewise, ammonia data are collected from the relevant literature. Missing data for ammonia-based DMUs is sometimes estimated from hydrogen (e.g., cost of ammonia = cost of hydrogen + their typical cost difference). Data can be estimated per kg of NH₃ considering the reaction stoichiometry and the efficiency of the different conversion steps. In addition, data of a typical carbon-capturing unit is used to estimate the energy consumption of hydrogen and ammonia production with CCS. In this cluster, indicators are divided by the amount of energy delivered by 1 kg gas, as estimated using the gravimetric energy density of the gas and the efficiency of the gas-to-power processes (i.e., compression, separation, storage, and reconversion). Details are provided in the supplementary information, section A2. These values are implicitly affected by indirect factors such as the lifespan of the facilities involved in these processes (e.g.,10000 working hours equivalent to five years lifespan for the equipment used in hydrogen production based on water electrolysis using wind energy [57]).

All the data collected, their references, and the assumptions considered to estimate the missing data are reported in detail in section B of supplementary information. The following subsections briefly explain the indicators considered (further details provided in the supplementary information, section B), while the final values used in our investigations are reported in Tables 1 and 2.

2.2.1. Economic indicators

The Levelized cost of energy (LCOE) is used here to capture the economic dimension, similar to other works [58]. Typically, the LCOE considers all relevant initial, variable, and end-of-life costs [59]. We source the LCOE directly from the literature for fast-response technologies, although values used neglect end-of-life costs due to the unavailability of data [20]. In the case of long-term storage options, the LCOE is obtained from the cost of producing 1 kg of gas using each of the production routes considered.

2.2.2. Environmental indicators

Four environmental indicators (three in the case of the long-term cluster) are considered in the analysis. Acknowledging all impact categories are important, we focus on energy consumption, energy density, water consumption, and global warming potential (GWP) since data on other indicators such as ecotoxicity or human health were not found for all the technologies considered. Regarding their use in DEA, environmental indicators are classified as follows: energy consumption, the

Table 1

Input and output indicators for fast-response technologies. LCOE: Levelized cost of energy, GWP: Global warming potential. FTEJ: Full Time Equivalent Jobs.

Storage	Inputs			Undesired output	Desired output	
options	LCOE (€)	Energy (GJ)	1/Energy density (kg/GJ)	Water (m ³)	GWP (CO ₂ -eq emissions)	Employ (FTEJ)
Flywheels	163.89	1.39	5291.0	1.8	44.2	$2.68 \cdot 10^{-4}$
SMES	102.50	1.74	101010.1	1.2	115.5	$3.35 \cdot 10^{-4}$
FB-VR	98.61	1.40	12345.7	$2.9 \cdot 10^{-3}$	8.9	$1.72 \cdot 10^{-4}$
FB-ZB	59.44	1.58	4830.9	$2.9 \cdot 10^{-3}$	12.1	$2.11 \cdot 10^{-4}$
LA	90.28	1.64	6944.4	$2.0 \cdot 10^{-3}$	20.0	9.14·10 ⁻⁴
Li-ion	172.92	1.40	1111.1	$0.5 \cdot 10^{-3}$	3.0	$13.19 \cdot 10^{-4}$
Li-Fe-Ph	172.92	1.54	1111.1	$0.5 \cdot 10^{-3}$	14.3	$14.06 \cdot 10^{-4}$
Li-Ni-Mn-Co	172.92	1.40	1111.1	$0.5 \cdot 10^{-3}$	11.8	$13.15 \cdot 10^{-4}$
Na-Ni-Cl	97.22	1.45	2314.8	4.8·10 ⁻³	18.2	9.80·10 ⁻⁴
Na-S	68.06	1.41	1388.9	$1.8 \cdot 10^{-3}$	0.2	$3.06 \cdot 10^{-4}$
Ni-Cd	117.36	1.51	1763.7	$0.2 \cdot 10^{-3}$	2.8	$22.86 \cdot 10^{-4}$
Average	119.65	1.50	12656.62	$276.3 \cdot 10^{-3}$	22.8	$8.65 \cdot 10^{-4}$
Std. deviation	43.43	0.12	29507.67	$626.8 \cdot 10^{-3}$	33.0	$6.8 \cdot 10^{-4}$

Table 2

Input and output indicators for long-term energy storage alternatives. LCOE: Levelized cost of energy, GWP: Global warming potential. FTEJ: Full Time Equivalent Jobs.

Storage	Inputs			Undesired outputs	Desired output
options	LCOE (€)	Energy (GJ)	Water (m ³)	GWP (CO ₂ -eq emissions)	Employ (FTEJ)
H ₂ , CG	124.82	5.52	2.74	2392.7	$1.12 \cdot 10^{-3}$
H ₂ , CG-CCS	261.81	9.72	2.74	103.2	$2.34 \cdot 10^{-3}$
H ₂ , SMR	139.79	5.52	2.74	1611.2	$1.25 \cdot 10^{-3}$
H ₂ , SMR-CCS	234.25	8.27	2.74	110.2	$2.10 \cdot 10^{-3}$
H ₂ , WE-Grid mix	650.67	5.52	2.74	4170.0	$5.82 \cdot 10^{-3}$
H ₂ , WE-Hydropower	918.86	5.52	1.31	175.0	$8.22 \cdot 10^{-3}$
H ₂ , WE-Solar	1949.45	5.52	1.31	699.9	17.44·10 ⁻³
H ₂ , WE-Wind	1080.48	5.52	1.31	87.5	9.67·10 ⁻³
H ₂ , WSCL	134.72	5.52	4.61	210.0	$1.20 \cdot 10^{-3}$
NH ₃ , SMR	279.64	5.42	0.46	554.15	$2.50 \cdot 10^{-3}$
NH ₃ , SMR-CCS	301.57	5.80	0.46	346.35	$2.70 \cdot 10^{-3}$
NH ₃ , WE-Grid mix	635.02	5.42	0.46	512.61	$5.67 \cdot 10^{-3}$
NH ₃ , WE-Hydropower	821.84	5.42	0.29	69.45	7.36·10 ⁻³
NH ₃ , WE-Solar	1539.72	5.42	0.30	233.40	$13.77 \cdot 10^{-3}$
NH ₃ , WE-Wind	934.56	5.42	0.30	90.65	8.36·10 ⁻³
NH ₃ , WSCL	275.98	5.42	0.30	68.27	$2.50 \cdot 10^{-3}$
Average	642.70	5.93	1.55	714.65	$5.75 \cdot 10^{-3}$
Std. deviation	539.83	1.23	1.33	1122.36	4.83·10 ⁻³

inverse value of energy density, and water use are considered inputs, while GWP is deemed an undesired output.

Note that energy consumption is not an impact *per se*, yet it is included here to capture other potential direct and indirect impacts associated with generating any energy carrier. Energy is required to manufacture storage units, operate them (in some cases), and is also fed for storage; therefore, the total energy consumption aggregates these three contributions.

For fast-response storage technologies, the energy consumption for manufacturing is retrieved mainly from techno-economic studies of the different technologies, while their operational energy is considered zero, and the energy fed for storage is calculated based on typical usage as reported in the supplementary information, section B. For long-term storage alternatives, the energy required to produce 1 kg gas is used as their total energy consumption.

The energy density is also used as an environmental indicator as a higher gravimetric density will result in lower material use and subsequent environmental impacts [60].

Since all the indicators are expressed per GJ, its reciprocal value (1/energy density) is used.

Water use stems mainly from the manufacturing stage for fastresponse technologies and is collected from techno-environmental studies. The operational water consumed to produce 1kg of gas is used for long-term energy storage options. The global warming potential (i.e., CO_2 -eq emissions) is quantified using Life Cycle Assessment (LCA), a standardized tool for evaluating environmental impacts incurred during all life stages of a product [22,61]. The CO2-eq emissions for fast-response technologies are usually provided per capacity of the storage unit. Likewise, for long-term storage, the CO₂-eq emissions are related to the production of 1 kg gas. Additional calculations necessary to obtain the GWP per GJ of delivered energy are reported in section B of the supplementary information.

2.2.3. Social indicators

Employment, energy security, health, public acceptability, and safety are the most widely used indicators of the energy sector [58]. Despite this, only direct employment is used in this contribution as data for the remaining indicators were missing for all or most technologies [62].

In the context of DEA, labor could arguably be considered either as a desired output with positive social effects or as an input for the construction or operation tasks. Without loss of generality, we here adopt the former approach because the energy sector is a promising source of new job opportunities worldwide.

2.2.4. Summary

The final indicator values used for each DMU in the corresponding DEAs are presented in Tables 1 and 2.

2.3. Data uncertainty

As already discussed, the value of specific indicators may vary within a certain range due to simplifications and the diversity of alternatives available for some storage options, sometimes affected by regional differences. We characterize the uncertainty associated with each input and output using uncertainty distributions to consider this variability and obtain more robust conclusions. This implicitly assumes that the propagation of uncertainties associated with the individual factors needed to compute a particular indicator falls within the uncertainty considered for the final value of the indicator itself.

Specifically, three types of distributions are used as follows. For parameters where we could find only one value in the literature (i.e., no range of values is available), uncertainty is characterized based on a uniform distribution with support of $\pm 10\%$ of the nominal parameter. This is the case of water use and feed energy during the lifetime of technologies (see Tables B3, B6, and B7 of supplementary information).

For parameters with a range of values available, we select triangular distributions whose limits correspond to the lowest and highest data reported in the literature for the indicator and its mode to the nominal value (i.e., the average among all values in the literature). The indicators belonging to this group are LCOE, energy consumption, energy density, and employment. Note that for each of these indicators, there are a few DMUs for which we have only a single value rather than a range for them. For these cases, $\pm 10\%$ of the nominal parameter is used to estimate a range following a triangular distribution.

Uncertainties on GWP are characterized following the guidelines from the Ecoinvent database [63]. According to this, each life cycle inventory entry (e.g., methane emissions, carbon dioxide emissions, etc.) used to compute the final life cycle impact (e.g., global warming potential) is assumed to follow an individual lognormal distribution whose uncertainty is characterized based on the so-called Pedigree Matrix [63]. This matrix assigns scores to different data quality categories for each dataset, which are later translated into the corresponding standard deviation (σ). These categories are reliability, completeness, temporal correlation, geographical correlation, and future technical correlation [64].

For simplicity, we assume that the standard deviation of the life cycle impact can be characterized based only on the most significant stressor among life cycle entries (i.e., CO_2 emission in the case of the GWP). This simplification, previously used elsewhere [65], avoids characterizing uncertainty distributions for hundreds of life cycle entries that would contribute very little to the variability in the final impact. Further details on this approach are provided in the supplementary information, section C.

After characterizing all the uncertain distributions for the different parameters, these distributions are discretized using Monte Carlo sampling to generate 100 independent scenarios for each indicator. Finally, a DEA model is solved for each scenario and DMU, following the approach in Ewertowska et al. [40]. It results in 100 different efficiency scores for each DMU (one for each scenario), which will be reported in results as error bars over efficiency scores. The next section provides the details on the DEA model used.

2.4. DEA model

With the values of the different indicators, we can finally use DEA to estimate the efficiency score of each DMU in each scenario. Without loss of generality, the non-oriented undesired output slackbased model (Un-Outputs SBM) is used in this study (M1, where variables are represented in italics, while parameters appear in normal Energy Storage Materials 48 (2022) 412-438

font) [66].

$$\tau^{*} = \min t - \frac{1}{m} \sum_{i=1}^{m} \frac{S_{i}^{-}}{x_{io}}$$

s.t. $1 = t + \frac{1}{s_{1} + s_{2}} \left(\sum_{r=1}^{s_{1}} \frac{S_{r}^{g}}{y_{ro}^{g}} + \sum_{r=1}^{s_{2}} \frac{S_{r}^{b}}{y_{ro}^{b}} \right)$
 $x_{o}t = X\Lambda + S^{-}$
 $y_{o}^{g}t = Y^{g}\Lambda - S^{g}$
 $y_{o}^{b}t = Y^{b}\Lambda + S^{b}$
 $S^{-} \ge 0, S^{g} \ge 0, S^{b} \ge 0, \Lambda \ge 0, t > 0.$ (M1)

Here, T* is the efficiency score of DMU o under evaluation, with the * superscript denoting the optimal value of the variable, that is, the value the variable takes in the optimal solution to the corresponding problem. Variable t is the Charnes-Cooper linear transformation coefficient, necessary to transform the original nonlinear undesired output SBM model into a linear model like M1 (see Li et al. for more details [66]). Also, m, s1, and s2 stand for the number of the inputs, the number of desired outputs, and the number of undesired outputs, respectively. In all cases, subscript *i* refers to inputs, while subscript *r* refers to outputs. Variables Si^{-} , Sr^{g} , and Sr^{b} correspond to the slacks in inputs, desirable (good), and undesirable (bad) outputs. In this model, slack variables quantify the distance from each DMU to the efficient frontier. This will be further discussed in the next paragraph. Similarly, x_{io} , y_{ro}^{g} , and y_{ro}^{b} refer to the input, the desired output, and the undesired output of DMU_o, respectively. X is the matrix of inputs (X= $[x_1, x_2, ..., x_n]$), while Y_g and Y_b are the analogous matrices for desired $(Y^g = [y_1^g, y_2^g, ..., y_n^g])$ and undesired outputs $(Y^b = [y_1^b, y_2^b, ..., y_n^b])$. Finally, Λ reports the weights with which efficient DMUs are combined to constitute the virtual DMU of the DMU assessed. Therefore, elements of Λ with non-zero values correspond to efficient DMUs belonging to the reference set of the DMU assessed. The concept of virtual DMU is further explained in the next paragraph.

According to model M1, a DMU is efficient ($T^*=1$) when all the slack variables are zero for all the inputs and outputs ($S_i^- = 0, S_r^g = 0, S_r^b = 0$, $\forall i, r$). Efficient DMUs are linearly combined to form the so-called efficient frontier. Conversely, if $T^* < 1$, the DMU is inefficient, and at least one of the slacks $(S_i^{-}, S_r^{g}, S_r^{b})$ has a strictly positive value. In this case, the exact efficiency score (T^*) depends on the distance to a reference point on the frontier. The reference point is constructed by combining the efficient DMUs that are closer to the inefficient DMU under evaluation. Therefore, the reference point presents an efficient-equivalent version of the inefficient DMU assessed; this point is the so-called virtual DMU. Slack variables provide the distance from the inefficient DMU to its virtual DMU. SBM models are non-radial in the sense that they handle input(s) and output(s) without imposing proportional changes for them [67] (i.e., slacks can freely take any value independently from each other). Note that slack values can guide improvement efforts (i.e., the required decrease in inputs and undesired outputs and the required increase in the desired outputs). For more clarification, see Fig. 4 of Zurano-Cervelló et al. [26].

Model M1 assumes that the ratio of outputs to inputs does not depend on the inputs' level. This assumption is known as constant returns-toscale (CRS) in DEA literature [26]. The use of CRS is consistent with the use of normalized data (i.e., per GJ), as explained in Section 2.2.

One well-known limitation of DEA models is that efficient DMUs cannot be further discriminated, thus preventing the obtention of a ranking that might help decision-making purposes [68]. To overcome this, a super-efficiency model that allows the efficiency score to be higher than one is used for efficient DMUs [41]. In a super-efficiency model, the DMU to be evaluated is removed from the set of candidate DMUs that can form the efficient frontier, thus, measuring the distance from this DMU to the frontier created with the remaining ones [68,69]. The undesired output slack-based super-efficiency DEA model (M2) is selected in this study. The mathematical formulation for the fractional



Fig. 4. Distribution of efficiency scores for fast-response energy storage technologies under uncertainty. Short-term technologies: Flywheel, SMES; Medium-Term: rest of the technologies. Grey region: inefficient, Light green region: efficient. Median efficiency: dashed line inside the plots. 1st (lower) and 3rd (upper) quartiles: dot lines inside the plots. Changes in the plot's width: distribution of the data standing for the corresponding efficiency. Percentages next to violins: number of efficiency scores that are equal to or more than 1 (i.e., probability of being efficient).

form of this model is as follows, although we use the linear form in our investigations. Additional details on the model and how to transform it to a linear-equivalent version can be found elsewhere [66,70,71].

$$\delta^{*} = \min \frac{1 + \frac{1}{m} \sum_{i=1}^{m} \frac{S_{i}}{x_{io}}}{1 - \frac{1}{s_{1} + s_{2}} \left(\sum_{r=1}^{s_{1}} \frac{S_{r}^{g}}{y_{ro}^{g}} + \sum_{r=1}^{s_{2}} \frac{S_{r}^{b}}{y_{ro}^{b}} \right)}$$

s.t. $x_{o} \geq \sum_{j=1, \neq o}^{n} \lambda_{j} x_{j} - S$
 $y_{o}^{g} \leq \sum_{j=1, \neq o}^{n} \lambda_{j} y_{j}^{g} + S^{g}$
 $y_{o}^{b} \geq \sum_{j=1, \neq o}^{n} \lambda_{j} y_{j}^{b} - S^{b}$
 $1 - \frac{1}{s_{1} + s_{2}} \left(\sum_{r=1}^{s_{1}} \frac{S_{r}^{g}}{y_{ro}^{g}} + \sum_{r=1}^{s_{2}} \frac{S_{r}^{b}}{y_{ro}^{b}} \right) \geq \epsilon$
 $\lambda, S, S^{g}, S^{b} \geq 0$ (M2)

Here, δ^* is the super efficiency score of DMU o under evaluation, with the * superscript denoting the value the variable takes in the optimal solution to the corresponding problem (i.e., the optimal value of the variable). Note that, since this model is used to rank the efficient DMUs, the super efficiency score is always greater than or equal to one ($\delta^* \ge 1$). The highest value of δ^* corresponds to the first ranked DMU among those under evaluation. Also, λ is the intensity vector that reports the weights of efficient DMUs in constructing the corresponding virtual DMU, ε is the non-Archimedean infinitely small value that forces the denominator to be positive even after the growth of undesirable output(s). All other symbols are the same as in M1 [70]. Note that, in this model, the slack values can be understood as a safe margin of changes for input(s) and output(s) in the sense that the DMU assessed can afford worsening its performance by these quantities while remaining efficient. Therefore, slacks are proportional to the stability level of the efficient condition of the DMU (i.e., the higher the super-efficiency slacks, the more likely it is the DMU will remain efficient). It could be said that safe margins hint at sustainability indicators that the efficient technology relies on to be deemed efficient. In addition, these margins provide a safe room for technology development, quantifying affordable downgrades in key performance indicators that may arise due to the enhancement of other features of the technology.

Models M1 and M2 need to be solved for each DMU and scenario, providing 100 (super)-efficiency scores for each DMU.

3. Results and discussion

The results obtained for fast-response technologies are reported in Section 3.1, while the results for long-term options are presented in Section 3.2. Safe margins of change are also reported for efficient DMUs, providing insight into why a particular technology is efficient. In addition, improvement targets are presented for inefficient DMUs, offering guidelines for technology developers to enhance their performance. Further, Section 3.3 discusses DEA results in the context of the technology readiness level (TRL) of the different technologies, providing insight into the future of the energy storage systems market. Finally, Section 3.4 incorporates the stakeholders' concerns for technology section.

3.1. Fast-response technologies

Fig. 4 depicts the efficiency scores distribution obtained by solving models M1 and M2 for each DMU in the fast-response cluster for 100 scenarios. Each of these scenarios corresponds to a particular realization of the uncertain parameters, obtained by applying Monte Carlo sampling to the associated uncertain distributions. As a result, instead of having a single efficiency score for each DMU, 100 values are obtained, i.e., one for each scenario. The lowest efficiency score obtained for each DMU corresponds to its worst scenario, while its highest efficiency score represents its best scenario. The efficiency scores obtained for the remaining scenarios are distributed between these extreme values. This distribution is depicted using a violin plot, where wider violin sections correspond to efficiency scores with a higher probability of occurrence (i.e., the DMU achieves this efficiency score in more scenarios). Technologies whose violins are wholly located in the efficient area have obtained an efficiency score greater than or equal to one in all the scenarios and, therefore, have a 100% probability of being efficient according to our data (e.g., Ni-Cd and Na-S). Note that the violin plots in this figure have been truncated to report the minimum and maximum efficiency

obtained for each technology; meanwhile, the median efficiency is presented using a black dashed line inside the plots, and the lower (first) and upper (third) quartiles are shown with dot lines inside the plots. The percentage next to each violin provides the probability of each technology being efficient. For instance, a 70% probability means that, among the 100 scenarios discretized using Monte Carlo sampling, the technology is efficient in 70 scenarios and inefficient in the remaining 30.

Flywheel is the most efficient option among fast-response technologies, with an efficiency score of 2 in 99% of the scenarios (inefficient only in one scenario). Flywheel is a short-term technology with high power density [44] and cheap power cost [20], despite having a more challenging energy density and energy cost (the fourth highest LCOE in this group, see Table B1 of supplementary information). Therefore, according to these results, the flywheel should be the technology of choice, especially for applications such as power quality control, where power is more important than energy. Indeed, the flywheel is suitable for frequent and rapid charge-discharge cycles that are required in a short time horizon, as otherwise, its long cyclability and high self-discharge might hamper its performance [5,10].

Ni-Cd, Li-ion, and Na-S batteries stand at the second to fourth positions with median efficiency scores of 1.61, 1.36, and 1.32, respectively. As shown in Fig. 4, all types of lithium-ion batteries are found more efficient than a lead-acid battery, consistent with previous findings [38]. In the case of Ni-Cd batteries, it is worth noting that cadmium is the byproduct of zinc production processes (3 kg Cd/ton Zn), which suggests that the amount of cadmium production might be inflexible. Cadmium is a limited and valuable resource that is also dangerous for human health. Hence, its safe application needs extra caution [72]. Other factors attracting attention towards this battery include its high strength, good performance at low temperatures, and fast-charging capacity [10,73]. Conversely, sodium batteries take advantage of this low-cost and nontoxic material with high recyclability potential, making these batteries a promising candidate for high power applications [44,74]. Nonetheless, technical features such as low conductivity, volume expansion of cathode, and corrosion of their anode by the electrolyte need some improvements [75].

Other Li-ion battery types such as Li-Fe-Ph and Li-Ni-Mn-Co have a certain chance of being efficient (70% and 50%, respectively). Still, there are scenarios where their performance can drop down to 0.34 and 0.31, respectively. Overall, lithium batteries, widely used in electric vehicles [76], reveal an acceptable performance regarding the sustainability indicators considered in this study, with median efficiency scores between 0.99 and 1.36. Using them in electric vehicles means exposing them to hit in the event of an accident. Therefore, the explosion danger of these batteries, mainly caused by the elevated temperatures, needs to be solved [77,78]. One promising solution for this issue is to endow separators with thermal shutdown functions in their structure [77]. Nonetheless, their widespread use should be cautioned until more environmental studies investigate their impact on human toxicity and other environmental categories.

FB-ZB, SMES, and FB-VR with median efficiency scores below 0.07, 0.05, and 0.03 are by far the most inefficient technologies.

Note that the variability of the efficiency scores of the SMES, FB-VR, FB-ZB, LA, Na-S, and Ni-Cd is small, which suggests that their obtained efficiencies are reliable. Conversely, technologies such as Li-ion, Li-Fe-Ph, Li-Ni-Mn-Co, and Na-Ni-Cl show significant changes depending on the scenario, to the point of affecting their classification as efficient (green-shaded background in Fig. 4) or inefficient (i.e., dark-shadowed region). For instance, while flywheel is inefficient only in one scenario, Ni-Cd and Na-S are always efficient. FB-VR, SMES, FB-ZB, and LA have no chance to be efficient (efficiency scores always lower than 1), while Li-ion, Li-Fe-Ph, Li-Ni-Mn-Co, and Na-Ni-Cl may be efficient or inefficient. Within this last group, Li-ion, with a 93% probability of being efficient, is always more efficient than the other three and, therefore, should be the preferred choice. Indeed, comparing the probability of Li-ion and Na-S batteries of being efficient, we find that Na-S is deemed efficient in

all the scenarios, while there is a small probability (7%) that Li-ion batteries show an inefficient performance, reaching efficiency scores as low as 0.33 in the most pessimistic estimates. Despite this, Li-ion achieves a higher median efficiency score (1.36 vs. 1.32) and a higher maximum efficiency score (1.53 vs. 1.43), suggesting that Li-ion can perform better than Na-S batteries in the most optimistic scenarios. A risk-taker policy/investor may be inclined towards Li-ion batteries, capable of achieving better performance. In contrast, a risk-averse policy/investor will bet on Na-S, as there is "no risk" of it being a non-competitive technology. Likewise, Li-Fe-Ph (70% chance to be efficient) shows a better performance than Li-Ni-Mn-Co and Na-Ni-Cl in each of the scenarios, which suggests that it can be the selected alternative in case of storage technology with high energy density or low water consumption is needed (see Tables B5 and B6 of supplementary information). Similarly, Li-Ni-Mn-Co is more efficient than Na-Ni-Cl, with the former showing a 50% probability of being efficient, while this is only 18% for the latter. All this is, of course, besides other considerations not explicitly assessed in our DEA (e.g., Li-ion technology is already well developed [79], and is more accessible).

Fig. 5 shows the safe margin of change for the fast-response technologies deemed efficient in at least one scenario, with the values depicted corresponding to the average safe margins among the 100 scenarios. These margins represent the maximum percentage of change that can be introduced *ceteris paribus* to the current value of the indicators (added for inputs and deducted for outputs) before the DMU stops being efficient.

The simultaneous analysis of Fig. 4 and Fig. 5 indicates that the more efficient a technology is (i.e., a higher super-efficiency score), the higher its safe margin. Flywheel, the first-ranked technology among fast-response options, has a 198% safe margin on its employment. Similarly, Ni-Cd and Na-S batteries also reveal safe margins on employment of about 122% and 64.5%. This suggests that the top three technologies in this cluster are mainly selected because of their relatively higher job creation ability. The deployment of these technologies may be challenging in regions with a high wage rate, affecting their LCOE. In this case, automation may help overcome the lack of skilled laborers or their high wages.

Li-ion also relies on its appealing performance on employment (52.5% safe margin), yet it also counts with the contribution of energy density (20.7% safe margin) to ensure its efficient status. Note that the inverse of energy density is used here as an input, which means that the safe margin of 20.7% in the indicator translates into an equivalent 82.8% that can be afforded in its real energy density (from 0.9 MJ/kg to 0.75 MJ/kg).

Following the same pattern, Li-Fe-Ph and Li-Ni-Mn-Co show safe margins on employment and 1/energy density. Yet, Li-Ni-Mn-Co could afford a marginal increase of 1.56% in its energy consumption, while the LCOE of Na-Ni-Cl could slightly worsen (no more than 1.79%), making it the only technology of this cluster with such a cost advantage.

We next turn our attention to inefficient technologies, for which improvement targets provide the minimum changes (i.e., reductions for input and undesired output and increments for desired outputs) required to make them efficient Fig. 6. presents the average improvement targets for inefficient fast-response technologies across the 100 scenarios, including technologies that are only inefficient in some scenarios. In this latter case, the average is calculated over the 100 scenarios. For DMUs with improvement targets on more than one indicator, changing only one of the indicators will not be sufficient to make the DMU efficient. Note that, although improvement targets are reported for all the indicators used, some of them might not be controllable directly due to design features, production factors, or other constraints [80]. Hence, technology developers could use DEA models with different orientations if improvements in some particular directions are preferred.

We detect an urgent need to improve the performance of SMES in most of the categories: it requires more than 80% reduction in its LCOE and energy consumption, while even more demanding changes (>90%)



Fig. 5. The safe margin of change for efficient fast-response energy storage technologies under uncertainty (%). Employment (desire output): allowed reduction, Rest of the indicators (inputs): allowed increase. LCOE: Levelized cost of energy.

Tachnologias		Inp	Undesired output	Average		
recimologies	LCOE	Energy consumption	1/Energy density	Water use	GWP	Average
Flywheel	-0.87	-0.85	-0.88	-1.00	-0.99	-0.92
SMES	-83.61	-87.07	-99.45	-100.00	-100.00	-94.03
FB-VR	-91.42	-92.05	-96.31	-99.46	-97.83	-95.41
FB-ZB	-80.00	-90.16	-86.24	-98.98	-97.70	-90.62
LA	-40.69	-57.65	-54.44	-96.74	-93.50	-68.60
Li-ion	-4.27	-2.31	-1.79	-5.41	-3.22	-3.40
Li-Fe-Ph	-5.54	-5.22	-1.90	-7.23	-22.79	-8.54
Li-Ni-Mn-Co	-9.82	-7.23	-4.28	-11.54	-34.70	-13.51
Na-Ni-Cl	-24.03	-39.42	-16.88	-79.30	-72.20	-46.37
Average	-37.81	-42.44	-40.24	-45.42	-47.54	
-100% -90%	-80%	-70% -60%	% -50%	-40% -3	0% -20%	-10% 0%

Fig. 6. Improvement targets for inefficient technologies of fast-response energy storage cluster under uncertainty (%). All the indicators: demanded reduction. LCOE: Levelized cost of energy. GWP: Global warming potential.

are required in its energy density, water use, and GWP. Recalling that the sustainability indicators used in this contribution correspond to unitary values expressed per GJ of delivered energy, the results obtained can be explained by a very low net delivered energy of SMES compared with its nominal value. Acknowledging that most storage technologies have certain losses, these are dramatic for SMES, which, undoubtedly, makes it necessary to control the amount of energy wasted before attempting to achieve the performance levels requested by DEA. In a SMES, the superconducting coil is cryogenically cooled to avoid loss of stored energy [7]. Making coils out of superconducting materials like mercury and vanadium can decrease resistance and the need for cooling. Submerging the coil in liquid helium can also contribute to keeping it cold, avoiding losses, and gaining the superconducting advantage [14]. Note that these modifications might also affect the economic performance of SMES, which is already critical with an operational cost ten times higher than for batteries [11].

The two flow batteries evaluated, FB-VR and FB-ZB, need about a 90% reduction on their input indicators and their GWP to become efficient. Although they need considerable improvements in all the categories, these batteries are still at the early stages of their development (technology readiness level of 6 out of 9 [79]), therefore, such changes may not be out of reach for them. It seems that a remarkable rise in the efficiency score of these batteries can be achieved by a better understanding of their construction that could help to improve their currently low energy density [81,82]. Today, the unclear reaction mechanisms and electrolyte performance are barriers to their improvements [9]. In this context, the application of finite element simulations could guide the construction of their electrode structure, providing information about the dimension of channels in electrode fibres and the path of electrolyte flow while helping to understand the reactions mechanisms better [83]. The application of 3D nanostructures as electrocatalysts is also useful to increase the catalytic activity (that affects the polarization resistance and energy loss [84]) and the cycling stability of flow batteries, which are necessary factors to make these batteries viable at the commercial level [85]. Still, poor cycling [9], corrosivity of electrolytes, and the environmental pollution associated with flow batteries also need significant improvements [86]. Currently, the cycling of redox flow batteries is limited by the sluggish anode redox chemistry [87]. Regarding FB-VR, the number of operational cycles during their lifetime is expected to rise from about 12000 cycles [14,20] to 50000 cycles by 2030 [88]. This ceteris paribus change would increase their delivered energy during its lifetime by 428.4% and decrease, in turn, the value of their energy consumption by 3.8% (i.e., 76.7% reduction in manufacturing energy consumption, while the feed-in energy to charge the technology will not change), and its GWP by 85.2%. This battery's targeted improvement for energy consumption and GWP is 92.0% and 97.8%, respectively (see Fig. 6 for improvement targets). Hence, rising the number of cycles is very helpful but still not enough to reach the improvements targeted. Acknowledging that a significant part of the battery value depends on the raw materials used in its structure [89], essential developments in material science are required to guarantee the achievement of such targets. For instance, ammonium-based electrolytes [90], and lowcost non-corrosive advanced ion-conducting membranes are the most promising solutions to solve the cyclability issue and the stability and environmental concerns associated with flow batteries [91]. In addition, employing nonaqueous electrolytes solves the corrosivity problems and is more environmentally friendly [92]. Also, application of organic redox-active species can significantly reduce the cost of flow batteries in the future [93]. It seems that, the development of low-cost membranes, increasing the power density, and rising the life cycle would help these technologies to improve their performance significantly [81,94].

Two other technologies need significant improvements to become efficient. The first is LA, with improvement targets above 40% for LCOE, energy consumption, and energy density and above 90% for water use and GWP. The second one is Na-Ni-Cl, with targets around 20% on LCOE and energy density, 40% on energy consumption, and more than 70% on water use and GWP. These two technologies ranked second and third expensive between the batteries based on the cost of their storage section, while their operational cost is lower than most of the batteries. Therefore, decreasing the cost of the storage section will lead to a significant improvement in their LCOE [20]. In the absence of practical methods to decrease the cost of the storage section, raising their lifetime and life cycle, which currently is not interesting for LA, will lead to delivering more energy during their lifetime and consequently a lower LCOE [10]. On the other hand, their energy consumption can be decreased by improving the power stack materials manufacturing, the plant balance, and the power control system, which are the steps with the highest energy consumption [95]. Also, the economy of scale can be to the benefit of their environmental impacts. For instance, the GWP per GJ of an 8 MW lead-acid battery is much lower than a 2 MW one [96]. Certainly, recycling battery materials will decrease their associated environmental impacts and increase material availability [59]. This observation is underpinned by studies reporting that the impacts of recycling are lower than the impacts of producing fresh materials [95]. Currently, lead-acid is the only battery with a very good recycling operation procedure, and a high percentage of recycled lead is usable in the production of new batteries [72]. Also, its good performance in different temperature ranges makes it a potential candidate for operations with fluctuating temperatures [10].

Two batteries have a remarkable chance to be efficient, Li-Fe-Ph and Li-Ni-Mn-Co, with average improvement targets of 8.54% and 13.51%, respectively. To this end, they should decrease their GWP (23 and 35%, respectively). Like the lead-acid battery, this group of batteries can gain from the economy of scale to decrease their GWP. Recycling operations exist for lithium-ion batteries (not economical) [97], but these are not well established yet and need further attention. For instance, in the case of their cathode, the material used, its morphology, and microstructure affect the recycling process and production of hazardous pollutions [98]. High nickel-metal carbons and spinels Li-rich Mn are recommended for the cathode to improve the battery capacity and operating voltage. At the same time, graphite containing silicon oxides attract attention for the application on the anode side [89]. On the other hand, the LCOE of these batteries requires improvements beyond 5% for Li-Fe-Ph and beyond 9% for Li-Ni-Mn-Co. Regarding the fast and broad research concerning these batteries' characteristics and costs, the cost reduction deemed is not far to achieve [15]. Layered lithium-rich low-cost cathode materials are the most attractive option for this purpose [99]. In addition, controlling other indicators, such as water use, will be necessary. Battery manufacturing processes should be highlighted to control water consumption since these processes contribute more in their material consumption than operation and end-of-life processes [100]. Furthermore, Li-ion needs to obtain safety and environmental certificates to solve the legal issues that ban its extensive application [88]. The latest development suggests that its safety can be improved by using a physical separator between electrodes, which provides a reliable bridge for ion transport [101]. Their frequent charging needs and long charging time are other issues that need to be solved [9,10].

Finally, flywheel and Li-ion are almost always efficient, translating into very small improvement targets (always below 5.5%). These could be achieved by the targeted choice of material [100], of course, so as not to worsen other aspects (e.g., self-discharge, lifetime, charging time, etc.).

The estimated improvement target for employment is always zero for all the technologies, reason why it is not depicted in Fig. 5.

3.2. Long-term energy storage options

Fig. 7 presents the efficiency scores for long-term energy storage alternatives. Like Fig. 4, violin plots have been truncated to report each alternative's minimum and maximum efficiency. Median efficiencies are presented using a black dashed line inside the plots, while the lower (first) and upper (third) quartiles are shown with dot lines inside the



Fig. 7. Distribution of efficiency scores for long-term energy storage options under uncertainty. The colors for hydrogen and nitrogen are indicated underneath the x-axis labels. Grey region: inefficient, Light green region: efficient. Median efficiency: dashed line inside the plots. 1st (lower) and 3rd (upper) quartiles: dot lines inside the plots. Changes in the plot's width: distribution of the data standing for the corresponding efficiency. Percentages next to violins: number of efficiency scores that are equal to or more than 1 (i.e., probability of being efficient).

plots. The percentage next to each violin provides the probability of each technology being efficient. For clarification of the used colors for hydrogen and nitrogen underneath the x-axis labels, see Fig. 2.

As shown in Fig. 7, green ammonia from solar energy is the only long-term storage option that is always efficient, achieving a median efficiency of 1.265. This efficiency score is mainly due to this technology's high employment and low water consumption (Table B7 and B11). Green hydrogen produced with solar energy and then the one produced with wind energy are the next two alternatives, with median efficiency scores of 1.09 and 1.08, respectively. The former takes advantage of its low energy consumption, while the latter offers important employment opportunities. Green ammonia from hydropower and wind energy are ranked fourth and fifth, with median efficiencies of 1.07 and 1.02, respectively. These results reveal that the top five long-term storage alternatives relate to water electrolysis using renewable energies (i.e., green hydrogen and ammonia). Although green hydrogen and ammonia are still very expensive compared with grey and even blue ones, it is expected that future cost reduction of renewable energy sources will help them to become the cheapest options [45]. As will be further discussed later, other options like green hydrogen using hydropower, ammonia produced by water electrolysis using grid mix energy, and ammonia produced by chemical looping processes have a certain chance of being efficient despite displaying median efficiencies below one: 0.53, 0.44, and 0.39, respectively. Moving to any of the remaining eight long-term storage options results in a dramatic decrease in the median efficiency score, below 0.33 in all the cases. It means that these technologies are, in principle, inferior to the other eight when assessed based on the economic, environmental, and social indicators we considered. Between these alternatives, grey hydrogen does not have any chance of being efficient, regardless of the technology employed in its manufacturing (max efficiency of 0.33 for coal gasification and 0.32 for SMR). In addition to their high GWP, their high energy and water consumption also exacerbate their situation. Therefore, they need remarkable improvements to be comparative with other options in the market.

Among the five alternatives that are efficient in all or most of the scenarios, two are related to hydrogen and three to ammonia, and all of them use renewable energy. It means that only 22% of hydrogen alternatives can be deemed efficient, while 43% of ammonia-based options are found efficient in at least some scenarios. Compared to hydrogen, ammonia is safer and generally needs lower space for storage due to its higher volumetric density [102]. However, developments like applying complex hydride materials can help store hydrogen safely and efficiently [103].

Green hydrogen or ammonia have better performance than their grey alternatives, even between inefficient options. Despite this, grey alternatives still are widely used owing to the higher reliability of fossil fuels and the possibility of gas production with higher capacity. Maybe in the form of policies, extra efforts seem necessary to move to cleaner sources [104].

Employing a CCS unit is essential for processes that use fossil fuels to control CO_2 emissions, although we did not find this to cause a fundamental change in their efficiency scores. Indeed, the maximum difference in efficiency scores of production units (hydrogen or ammonia) with and without CCS is approximately 0.05 (based on the median). This happens because when a CCS unit is used, the reduction of GWP comes at the cost of increasing LCOE and energy consumption. Hopefully, reducing the cost of electricity generation using cleaner sources besides other technical improvements will increase the contribution of more sustainable gas production methods such as green hydrogen and ammonia. For instance, projections indicate that, in 2030, the cost of ammonia production using renewable sources will be lower than its production using methane reforming [105].

In terms of the variability of the efficiency scores, we find that green ammonia from solar energy is always efficient. The next green alternatives based on water electrolysis are ammonia from hydropower, hydrogen from wind, and hydrogen from solar, showing 92%, 91%, and 89% probability of performing better than the remaining alternatives in each scenario. Among the other options, green ammonia from wind (NH₃, WE-Wind) and hydrogen relying on the grid for water electrolysis (H2, WE-Grid mix) have a 76% and 38% probability of being efficient. Other options like ammonia produced using water splitting by chemical looping (NH₃, WSCL), ammonia produced by steam methane reformation and using carbon capture and storage (NH₃, SMR-CCS), hydrogen produced by water electrolysis powered by hydropower energy (H2, WE-Hydropower), ammonia produced by steam methane reformation (NH₃, SMR), ammonia produced by water electrolysis powered by grid mix (NH₃, WE-Grid mix), hydrogen by steam methane reformation using CCS (H₂, SMR-CCS), hydrogen from coal gasification using CCS (H₂, CG-CCS), and hydrogen produced using water splitting by chemical looping (H2, WSCL) have smaller probabilities of being efficient (33%-4%), which suggests these technologies are among the best performance alternatives only in some particular contexts. For instance, green ammonia based on wind energy might be particularly suitable for regions with strong and constant winds and without reliable solar energy potentials. Meanwhile, grey hydrogen production options such as coal gasification (H2, CG) and steam methane reforming (H2, SMR) are inefficient in all the scenarios. Even blue hydrogen alternatives such as hydrogen production based on the coal gasification with CCS (H2, CG-CCS) and hydrogen production based on the SMR and with CCS (H2, SMR-CCS) only have a small chance to be efficient (less than 5%). This suggests they might still be viable alternatives for some niche applications, although less likely than the alternatives previously discussed. Overall, green processes are preferred over blue and grey due to their higher efficiency according to the indicators employed in this contribution.

Comparing hydrogen use as a standalone energy storage option with ammonia obtained from the same hydrogen, we can see that ammonia has better performance almost always. The only exception is when wind energy is used for water electrolysis. In this case, the median efficiency of hydrogen is 0.055 higher than for ammonia. Therefore, converting hydrogen to ammonia is recommended if the ultimate goal is to have a long-term energy storage alternative.

We next investigate why some technologies are efficient. To this end, we plot the safe margins of change for alternatives that are efficient at least in one scenario (see Fig. 8). Overall, efficient alternatives have safe margins only on one or two of their indicators at most, while the rest should remain at equal or better values to retain their efficient condition. The case that attracts the most attention is the 48.3% safe margin on employment that is related to ammonia produced using water electrolysis by solar energy (NH₃, WE-Solar). Supposing that technological innovations and automation decrease labor needs for this alternative as it moves from 5 to higher technology readiness levels [106,107], the next storage options in terms of their efficiency scores could be explored as alternatives. Note that, in this case, the safe margin on employment is also high for the third and the fourth most efficient options, hydrogen produced by water electrolysis powered by wind (H₂, WE-Wind) and ammonia produced by water electrolysis powered by hydropower (NH₃, WE-Hydropower), with values of 15% and 12.9%, respectively. However, the second standing alternative, (H2, WE-Solar), can also afford to increase its low energy consumption by 24.7%, while (H₂, WE-Grid mix) could raise its LCOE by 9.7%, if deemed necessary for introducing additional innovations affecting other features. In addition, about 4.53%, 3.04%, and 1.79% rise in LCOE is allowed for (NH₃, WSCL), (NH₃, SMR-CCS), and (NH₃, SMR), respectively, which highlights the economic competitiveness of the chemical looping method compared to other options for energy production. For the rest of the alternatives, the safe margin is only obtained on LCOE and is lower than 1%. These are the alternatives that have a slight chance to be efficient.

Improvement targets are next computed for alternatives that are inefficient in at least one scenario (see Fig. 9). As in Fig. 6, the

values depicted correspond to perceptual changes concerning the current value of the indicators, calculated as the average of all 100 scenarios.

Fig. 9 reveals that the minimum reduction required in LCOE is 0.6% and is related to (H₂, WE-Wind), which could be naturally achieved by falling costs of renewable energies, including wind. Meanwhile, the maximum reduction in LCOE is 11.76% for (H₂, WE-Grid mix). The rest of the options are demanded LCOE reductions within this range, but generally lower for ammonia than for hydrogen. Note that the cost of producing 1 kg hydrogen is higher than producing 1 kg ammonia [108]. Also, since converting the gas to electricity using a fuel cell is more expensive than with gas turbines [20], replacing fuel cells with gas turbines can help control the LCOE for hydrogen. In addition, the cost of storing the gas generated changes from $130 \notin$ /kWh for above-ground storage to $3.7 \notin$ /kWh for underground storage [20]. Therefore, the DMUs needing to reduce their LCOE, can move to underground gas storage to help reach their target, provided this option is socially and politically accepted in the region.

The highest reduction required in energy consumption is about 90% related to grey and blue hydrogen, followed by WSCL. The current pilot efforts for CCS systems consume significant amounts of energy, but new technologies are being investigated that could bring the energy demand down [109]. For green hydrogen and ammonia, the reduction required in energy consumption is meager, and (H_2 , WE-Wind) with 0.45% demanded reduction in energy consumption is the last and the best one in this indicator. It is worth noting that there is considerable heat waste in the gas production reactors [50], and heat integration can improve their performance.

Regarding water use, the situation presents a similar pattern to energy consumption but with different values, ranging from 1.21% for (NH₃, WE-Hydropower) to 97.09% for (H₂, SMR).

Almost all the investigated long-term storage options need to decrease their GWP. Between the inefficient DMUs, changes requested start at 0.31% for (H₂, WE-Wind) and climb until 96.99% related to (H₂, SMR). Using CCS, CO₂ emissions can be controlled, but as already discussed, the contribution of CCS does not seem very effective in improving the efficiency score of DMUs (only 0.05), as CCS worsens the performance in energy consumption and LCOE. Despite this, a CCS unit is necessary for ammonia production since the existence of CO₂ in the reactor can affect the hydrogen and nitrogen reaction catalyst [110].

The improvement target obtained for employment of all the long-term storage options is zero and, therefore, not represented in Fig. 9.

3.3. Future market and development trajectory of energy storage options

A technology may look much more appealing than the other technologies in a comparative analysis such as DEA regarding the considered indicators. Despite this, not all energy storage technologies will successfully access the energy market [106]. The technology readiness level (TRL) of each technology allows us to assess the maturity and its level of expected changes in the future [88]. When TRL is low, and the technology is still in its research or concept stage, it is highly uncertain whether the technology will be able to be implemented or not [79]. If it succeeds, its development potential will be high at first. Still, as it gets closer to the level of full development (i.e., being marketable), this potential will decrease, following a learning curve. Technologies with a higher TRL value are developed very well, and further development may not be easy. In such a case, if the technologies are efficient, they may finally penetrate the market but probably not in the near future, and if they are not efficient, they will have a long and challenging way to go.

The future market access potential of fast-response energy storage technologies and long-term energy storage alternatives are discussed in Sections 3.3.1 and 3.3.2, respectively.



Fig. 8. The safe margin of change for efficient long-term energy storage alternatives under uncertainty (%). Employment (desire output): allowed reduction, Rest of the indicators (inputs): allowed increase. The colors for hydrogen and nitrogen are indicated underneath the x-axis labels. LCOE: Levelized cost of energy.

3.3.1. Future market of efficient and inefficient fast-response technologies

To explore the chance of each efficient or inefficient technology to access the market, we plot their median efficiency versus their TRL, as shown in Fig. 10. The median efficiency score of fast-response technologies used in Fig. 10 corresponds to the one obtained with DEA and previously reported in Fig. 4. The technology's current TRL is retrieved from different literature sources, as reported in Table (B.12) of supplementary information. Each technology should meet specific criteria to qualify for a certain level [79], according to nine levels ranging from 1 (non-mature) to 9 (mature). Usually, a TRL lower than four is related to technology at the research or concept test stage. If TRL is between 4 and 6, the technology is in the demonstration or technical development stage. When TRL is above 7, the technology is at a specific test certification stage or is marketable (i.e., TRL close to 9) [107].

Fig. 10 is divided into different zones using dashed lines placed at an efficiency score of one (i.e., splitting the region between efficient and inefficient technologies), and TRLs of 4 and 7 (i.e., splitting the region according to how close the technology is to be marketable in the future). The combination of the different criteria met gives rise to the classification of the technologies assessed into different groups as follows:

– Efficient ($\delta^* \ge 1$) and marketable (7 < *T RL* \le 9): flywheel, Ni-Cd, Li-ion, Li-Fe-Ph and, with a bit of optimism, Li-Ni-Mn-Co belong to this group. According to our results, all these technologies are winning choices since they show a competitive performance and a reliable level of development. The best technology selection from this group for a particular application can be underpinned by Fig. 5, which provides the indicators that each of these technologies performs best. For instance, flywheel, Ni-Cd, and Na-S are promising options when creating new job opportunities is essential for developing a region. Meanwhile, Li-ion, Li-Fe-Ph, and Li-Ni-Mn-Co are also

important employment creators, on top of showing a good performance in energy density, which makes them suitable for electric vehicles [76]. It is forecasted that, by 2030, Li-ion batteries will remain the dominant technology in mobility applications, while for stationary applications, a broader choice of technologies will be used [89]. Continued research on Li-ion batteries makes solid-state Li-ion batteries marketable by 2025. From 2025 to 2030, solid-state lithium metal, advanced solid-state Li-ion, and metal-air batteries probably will access the market [89]. Despite this, their TRL is still lower than 9, which indicates that some aspects still need to be improved before they can effectively penetrate the market of electricity storage technologies. For instance, the gravimetric energy density and number of cycles during the lifetime of Li-ion batteries are about 250 Wh/kg and 3000 cycles, respectively [20]. These parameters are expected to reach 350 Wh/kg [106] and 15000 cycles [89] according to the 2030 set plan target offered by the European association for energy storage, which presents the roadmap of energy storage technologies development.

- Efficient ($\delta^* \ge 1$) but not marketable ($4 < TRL \le 7$): only Na-S, the promising alternative to lithium-ion batteries [111], belongs to this group. The relatively low cost of this battery, the natural abundance of sodium resources [111], its high theoretical capacity, and the high energy density of sulphur [112], make the Na-S technology a promising candidate to attract investment, with real possibilities to infiltrate the market. Our DEA results confirm this, showing that Na-S is always efficient with a median efficiency score of 1.32. Although not a completely mature technology, its TRL is also relatively high, suggesting that Na-S is already highly developed, and that further development may not be easy or rapid. At this point, the main technical challenge of this battery is lowering its operational temperature [107]. The bottleneck to solving this problem is developing and designing suitable anode materials [111]. Hollow nanostructured market.

Long-term		Inputs	Undesired output	Average	
storage	LCOE	Energy consumption	Water use	GWP	Weruge
H ₂ , CG	-9.70	-89.29	-96.37	-96.06	-72.86
H ₂ , CG-CCS	-5.89	-83.12	-91.58	-66.46	-61.76
H ₂ , SMR	-7.57	-88.95	-97.09	-96.99	-72.65
H ₂ , SMR-CCS	-5.58	-81.48	-91.19	-68.81	-61.77
H ₂ , WE-Grid mix	-11.76	-38.45	-57.33	-60.12	-41.92
H ₂ , WE-Hydropower	-3.56	-25.45 -64.53		-25.51	-29.76
H ₂ , WE-Solar	H ₂ , WE-Solar -0.56		-0.57 -7.01		-3.75
H ₂ , WE-Wind	-0.60	-0.45 -6.66		-0.31	-2.01
H ₂ , WSCL	H ₂ , WSCL -8.18		-85.61 -94.36		-67.91
NH ₃ , SMR	-5.03	-60.30	-64.36	-72.67	-50.33
NH ₃ , SMR-CCS	-6.05	-56.29	-60.52	-63.03	-46.47
NH ₃ , WE-Grid mix	-3.09	-45.49	-55.01	-72.12	-43.93
NH ₃ , WE-Hydropower	-0.55	-1.40 -1.21		-0.36	-0.88
NH3, WE-Wind -1.28		-2.12 -2.31		-0.90	-1.65
NH ₃ , WE-WSCL -6.21		-52.85	-50.17	-31.86	-35.27
Average	-4.78	-44.49	-52.47	-46.60	
-100% -90% -8	-70%	-60% -5	60% -40%	-30% -20%	-10% 0%

Fig. 9. Improvement targets for inefficient technologies of long-term energy storage cluster under uncertainty (%). All the indicators: demanded reduction. GWP: Global warming potential.

terials capable of storing sodium ions in their structures through the combined conversion and alloying reactions are a class of promising materials for this purpose [113,114]. For example, indium tin oxide nanoparticles decorated onto activated carbon cloth enable operations in sub-zero by immobilizing the higher-order Na-polysulfides and promoting their conversion into insoluble end-discharge products [74].

- Inefficient ($\delta^* < 1$) but marketable ($7 < TRL \le 9$): LA and SMES are in this zone. Our analysis (Fig. 6) revealed that significant improvement targets are necessary to increase the efficiency of these technologies. SMES requires more than 80% fall in all the indicators except for employment. On the other hand, LA asks for about 50% decrease in LCOE, energy consumption, and 1/energy density, while its required decline for water use and GWP is even higher and above 90%. The fact that these technologies are marketable suggests that considerable time has already been spent in their development. At this point, they no longer have remarkable development potential. In the absence of a technological breakthrough, it is hard to imagine how these technologies could reach the required performance. For all these reasons, they are not particularly promising alternatives at this point.
- Inefficient ($\delta^* < 1$) and not marketable ($4 < TRL \le 7$): technologies in this zone are Na-Ni-Cl, FB-ZB, and FB-VR. These technologies need further improvement before they can become efficient. Still, since they are not yet at a marketable stage, it is not far from the expectation that they can move to the "efficient and marketable" zone. However, their improvement targets (Fig. 6) do not reveal a smooth path ahead. For instance, Na-Ni-Cl needs to decrease its water use and GWP in the range of 70-80%, along with reductions in its LCOE, energy consumption, and 1/energy density (16-40%). Meanwhile,

flow batteries (FB-ZB and FB-VR) need about 90% decrease in all the indicators except employment. For this latter group of batteries, extending the number of operational cycles and increasing the power density are requested (e.g., from 12000 cycles [14,20] to 50000 cycles [88] for FB-VR) [94].

- Research stage ($TRL \leq 4$): none of the technologies assessed qualified in this zone, whether efficient or inefficient. In this contribution, only the technologies whose data were available were considered. Usually, the data of technologies at the research step are not open to the public. It is expected that more players will be on the scene in the future, including organic batteries, metal batteries, onion shuttle-based batteries, high power primary regenerative batteries based on reactive metals [89], and hybrid energy storage technologies that include the advantages of different technologies [115]. Rapid growth is expected for technologies that manage to pass this step successfully.

Overall, the efficiency and TRL of all the energy storage technologies are influenced by some common factors. For instance, in addition to the environmental impacts and technical factors like energy capacity (kWh), energy density (Wh/kg), efficiency, response and discharge time, and the number of cycles during the lifetime of technology, the capital cost (\$/kW), and LCOE (\$/kWh) influence the technologies TRL value [116]. Almost all these factors are used in our DEA analysis explicitly (i.e., as an indicator) or implicitly (i.e., in calculations related to estimating the final value of the indicators). Therefore, the efficiency and TRL of these technologies will change simultaneously. So, in the next paragraph, we present some general improvement guides, the challenges ahead, and the solutions to deal with them.



Fig. 10. Market penetration chance of efficient and inefficient fast-response energy storage technologies. Short-term: Flywheel, SMES. Medium-term: Rest of the technologies. Grey region: Inefficient. Light green region: Efficient. Development potential decreases moving from TRL=0 to TRL=9.

The main challenge is developing advanced materials that enable high power and energy density to improve technical and environmental factors. To this end, the focus should be on adapting cathode and anode materials and stabilizing the formulation of electrolytes [89]. However, the unbalanced distribution of raw materials in different countries and the marginal extraction rate are obstacles to accelerating the development [89]. Reaching the production target of 300 GWh/a battery in the European Union alone will require 270000 tons battery-grade graphite, 30000 tons of silicon for the anode, 225000 tons of class 1 high purity nickel, 29000 tons of cobalt, 84000 tons of manganese, and 59000 tons of lithium for the cathode [89]. Mapping the availability of raw materials in different countries helps raise awareness regarding the available resources [106]. Beyond the application of sensing technologies, big data from sensors embedded in battery cells, the discovery of self-healing materials [117], artificial intelligence-based tools, physicsaware models, and autonomous synthesis robotics will enable scientists to learn the interplay between battery materials and interface. Therefore, several battery chemists will propose using novel developed materials and taking advantage of the knowledge obtained about the interplay between different materials [89].

Furthermore, according to the European 2030 technology development roadmap, liquid discharge from battery-grade material processing should be zero, and CO_2 emissions in material extraction and processing should be reduced by 50% [89]. To this end, currently used batch processes should be replaced by continuous processes, new smelting, and slag engineering technologies, and new recoverable reagents for battery metal leaching and extraction should be addressed [89]. Also, beyond 2025, some investments in the facilities that recycle materials from energy storage technologies will become necessary to reduce the pressure exerted on the environment [117], while recovery of metals from industrial or urban wastes can also contribute to this end [89]. Further, to decline the technology cost, its volume (i.e., large-scale manufacturing) and standardization (i.e., proper connection of system components, installation, maintenance, and performance) are factors that should be noted [106].

3.3.2. Future market of efficient and inefficient long-term energy storage alternatives

Fig. 11 presents the median efficiency versus TRL for long-term energy storage alternatives. Their median efficiency is obtained from DEA analysis, as previously reported in Fig. 7. Unfortunately, the TRL for many long-term storage options like hydrogen produced with solar, wind, or fossil fuels is not reported reliably and explicitly in the literature. However, generally, the TRL for grey processes is about 8-9 and is higher than for blue processes, which varies from 6 to 8. Finally, with a TRL of 1-3, green processes still have a long way ahead [118]. We select the median of the ranges reported as the TRL of each production method [106,107]. In the case of NH₃ production, the TRL is not reported for any of the routes. Therefore, the TRL of each power-to-NH₃ production





Fig. 11. Market penetration chance of efficient and inefficient long-term energy storage alternatives. Grey region: Inefficient. Light-green region: Efficient. Development potential decreases moving from TRL=0 to TRL=9. Grey H_2/NH_3 : H_2 or NH_3 produced using fossil fuels, Blue H_2/NH_3 : H_2 or NH_3 produced using fossil fuels, and the emitted carbon captured by CCS, Green H_2/NH_3 : H_2 or NH_3 produced using renewable energy sources.

method is considered the same as its power-to- H_2 equivalent (see Table (B.13) of supplementary information).

Fig. 11 is equivalent to Fig. 10, with the plotted area divided into six regions as given by the combination of the different categories achieved by the alternatives regarding their efficiency scores (efficient vs. inefficient) and their TRL level (research stage, not marketable, marketable). In this case, no alternative is both efficient and marketable. Only grey options, like power-to-gas based on water electrolysis using the grid mix, are close to a marketable situation. Despite this, grey alternatives are still far from efficient, with improvement targets on water use, GWP, and energy consumption asking for reductions beyond 80%. Considering that these alternatives are already well developed and almost marketable, these improvements seem out of reach.

The following alternatives in terms of TRL are blue options, such as power-to-gas using steam methane reforming or coal gasification, both coupled with carbon capture and storage. Compared to grey ones, blue alternatives present a better situation regarding their GWP, which, combined with their lower TRL, may result in a better performance once they are completely developed. However, even if they manage to infiltrate the market, such domination will only be a transition period before growing environmental concerns put green alternatives in action.

Only the green power-to-gas-to-power routes, such as H_2 and NH_3 based on the water electrolysis powered with wind or solar, and NH_3 based on water electrolysis using hydropower, are found efficient. These

alternatives are still at the research step and have good development potential, which could help them perform even better and achieve higher efficiency scores.

Overall, the bottleneck for developing the power-to-gas processes is water electrolysis. Due to different reasons, different electrolytic cells and processes have managed to achieve distinct TRL levels. For instance, alkaline electrolysis is well developed, with a TRL of almost 9, yet the technology is considered not marketable because of its high costs. Other options like polymer electrolyte membranes with a TRL of 8, solid oxide electrolytic cells, and the co-electrolysis of water and CO₂, both with a TRL of 5.5, still need to increase their energy efficiency and lifespan while reducing maintenance costs and environmental impacts [107]. In this context, large-scale production might help decline costs [88]. At the same time, the use of nanomaterials to store the gas shows an improved environmental performance compared to other choices like caverns [119].

Finally, it is also essential to consider the market readiness level when predicting the future of energy storage alternatives. Market readiness level records a value of 7-8 (out of 12) for power-to-hydrogen, which means the market has identified the need for this storage alternative and its application [88]. In addition, it is forecasted that raising concerns about CO_2 emissions will increase motivations for further investments in power-to-gas energy storage alternatives. Hence, while mobility is the most significant anticipated market for hydrogen in the short

term, balancing the energy system and grid reinforcement is expected to be the most demanding source for hydrogen in the longer term [88]. Further development of fuel cells, facilitation of gas storage, and cost reduction are the key factors that will pave the development trajectory of power-to-gas energy storage alternatives [79]. Preparing the necessary infrastructures and certificates for gas production processes and their application operations will require political and financial support [88].

3.4. Incorporating concerns from stakeholders in the technology section

In this contribution, we resorted to DEA to compare the energy storage options, where the absence of predefined weights is equivalent to implicitly assuming that all indicators are equally important. However, in real applications, the stakeholders' preferences or local contexts could challenge this assumption and even make some technologies rejected initially. Also, engineers, customers, or policymakers might show some preferences over selected indicators, which may vary with time, location, and the application of the technology. Therefore, we address the technologies comparison by presenting two additional investigations. In the first analysis, we include predefined weights as parameters in the model formulation. Indeed, we solved an additional problem where we assigned 80% of the total weight to environmental indicators and 20% to the rest. However, different weights could be used if stakeholders were more inclined towards other aspects of the problem (i.e., economic, or social). This exercise, discussed in detail in Appendix A.1, reflects the view of a stakeholder inclined towards using cleaner technologies.

This evaluation reveals that, between fast-response technologies, flywheel, Ni-Cd, Li-ion, Na-S, and Li-Fe-Ph face a significant decrease in their efficiency scores; nevertheless, they all remain efficient. This means that, although the main strengths of these technologies rest on their economic and social performance, they are also good choices for environmentally-friendly applications. The efficiencies of Li-Ni-Mn-Co, Na-Ni-Cl, LA, FB-ZB, SMES, and FB-VR rise remarkably. However, only Li-Ni-Mn-Co moves from inefficient to efficient, raising its median efficiency marginally from 0.98 to 1. In general, the prioritization of environmental indicators causes the efficiency scores of efficient technologies to decrease and that of inefficient technologies to rise. However, these changes are not enough to cause modifications in the relative ranking of the different technologies, which remains unaltered.

Regarding alternatives in the long-term cluster, hydrogen production based on water electrolysis powered with solar (H_2 , WE-Solar) and wind energy (H_2 , WE-Wind), and ammonia production based on water electrolysis powered with solar (NH_3 , WE-Solar), wind (NH_3 , WE-Wind) and hydropower (NH_3 , WE-Hydropower) present a decline in their efficiencies. This happens because increasing the weight of environmental indicators reduces the weight assigned to employment, which is the most favourable indicator for most of these alternatives. Nonetheless, all these technologies remain efficient. In contrast, the rest of the alternatives face an increase in their efficiency scores, although insufficient to make them efficient.

Overall, the list of technologies recommended based on the results from our original DEA is still valid when environmental aspects are placed at the core of the assessment since the relative ranking of the technologies remains unaltered. Further discussion about this alternative study where environmental indicators are prioritized is presented in Appendix A.1.

Alternatively, other stakeholders may argue that the cost of reverting the damage caused by increased environmental impacts is higher than the cost of prevention [120]. To illustrate such an approach, in our second investigation, we monetized the different indicators, translating indicators into a consistent monetary term based on the corresponding externalities. Using monetized indicators in a unit-invariant DEA model, like the one used in this contribution, would result in the same efficiency scores as obtained with the original indicators since using monetization coefficients to transform the indicators is equivalent to changing their units. To overcome this, we avoid DEA and, instead, use the monetized indicators to estimate the efficiency of the DMUs using the basic definition of efficiency, that is, the summation of (monetized) desired outputs minus the summation of (monetized) undesired outputs, divided by the summation of (monetized) inputs. This provides a measure of the absolute efficiency of the technology, which is conceptually different from the relative efficiency score provided by DEA. Combining monetized indicators into a single score is helpful to rank alternatives, nevertheless, opposite to DEA, it does not provide information on how to improve the worst-performing technologies. In addition, it is improbable that two technologies will tie with precisely the same score and, hence, it might be more challenging to decide a threshold below which the remaining technologies should be discarded for further analysis. This issue is solved with DEA, which mathematically classifies all the alternatives between efficient and inefficient. Therefore, we avoid any comparison of the efficiency scores obtained with the two approaches and we only compare the relative ranking achieved by the energy storage options with each of the two methods.

According to the results obtained for fast-response technologies, only flywheel, Na-S, and SMES report a decline in their ranking. Although this ranking degradation is significant for the flywheel, it keeps its position as the best short-term option. The rest of the technologies improve their ranking. For instance, Ni-Cd, which is the second-best option in the original DEA, ranks first in this new analysis. Regarding the alternatives of the long-term cluster, some changes are observed in their relative ranking, but green alternatives still occupy the first positions, and usually green hydrogen alternatives rank better than the green ammonia alternatives. More details are reported in Appendix A.2.

4. Conclusions

Storage technologies improve grid flexibility, which is particularly appealing when there is a high share of renewables. However, insufficient attention to energy storage technologies has slowed down their development, hindering, in turn, the penetration of renewables. To unlock this situation, energy storage technologies need to be assessed considering all the sustainability dimensions concurrently to ensure that unsustainable practices in energy storage do not offset the benefits from the increased use of renewables in the grid.

In this contribution, we benchmark energy storage options using DEA, considering uncertainty in the data through the generation of 100 scenarios. Storage options are classified into two clusters according to their response time, the first for fast-response technologies and the second for long-term storage options. Independent DEAs are carried out for each cluster, finding the technologies with the best performance in each situation and providing evidence for policymakers to develop better-informed regulations.

Flywheel, with a median efficiency score of 2 and efficient in 99% of the scenarios, is the most efficient option among fast-response technologies. Since it is a short-term storage technology, it is the recommended choice for power applications such as power quality control. Ni-Cd, Liion, and Na-S batteries are standing at the next positions in terms of efficiency. Indeed, these medium-term technologies were found efficient in all or most scenarios and, therefore, should be promoted for applications like electric vehicles, where modular storage technologies are needed. In this case, it is suggested to employ a separator in the battery structure to improve its safety. Fortunately, most of them are well developed and easy to access. Only, Na-S needs further developments to be marketable. Conversely, FB-VR, SMES, and FB-ZB, with efficiencies lower than 0.1, are graded as the most inefficient technologies. For these technologies, quantitative improvement targets are reported to help technology developers improve and make them competitive. For instance, in the case of SMES, controlling energy loss would be key to increasing its performance. Also, in the case of flow batteries, increasing the number of operational cycles is several times recommended. The application of nanomaterials and recently developed membranes are beneficial for this purpose. According to their TRL, these technologies are not entirely

developed and still can improve their performance and infiltrate the market. However, even technology like LA that is already developed is deemed inefficient in our evaluation. In such a case, the technology does not have an easy way ahead to improve its performance. Since the indicators influence each other, the developers should avoid worsening other indicators when improving one of them. For this case, the reported safe margin of changes gives insight into the worsening indicators can afford without turning a technology inefficient. We note that, in this contribution, we could not explore the effect of batteries on toxicity and human health owing to the lack of data for some of the options considered. This critical topic could be an object of future research since it might play an important role in shaping technological and political decisions.

Among the 16 long-term storage options evaluated, only ammonia produced from hydrogen obtained by water electrolysis powered by solar energy is efficient in all the scenarios. The most efficient options are green ammonia (median efficiency scores of 1.265 when based on solar and 1.07 if based on hydropower) and green hydrogen (1.09 using solar and 1.07 using wind). Grey hydrogen from coal gasification and from SMR processes are not efficient alternatives in any of the 100 scenarios considered. Apart from these two options, the other options in the longterm cluster emerge as efficient, at least in some scenarios. Again, we report improvement targets to guide developers in enhancing the different technologies. For instance, most options need reductions between 0.55-11.76% in their LCOE, which can be achieved by employing cheaper options like gas turbines instead of expensive fuel cells for gas reconversion, reducing the cost of renewable energies in the case of green alternatives and enhancing the availability and suitability of salt caverns for underground gas storage in the case of blue options. More significant actions are needed for other indicators like energy consumption of non-green hydrogen and ammonia, where technologies should target improvements between 38.45% and 89.29% to become efficient. These can be pursued by avoiding heat loss in reactors. Note, however, that it is expected that improving one of the indicators might affect others: for instance, insulation of reactors will result in a higher LCOE. In addition, the safe margin of change obtained for efficient alternatives reveals that most of them rest on employment generation for being deemed efficient. Despite this, their establishment may be challenging in regions with a high wage rate or lacking skilled laborers. Even in this case, automation may be helpful, and considering that these green alternatives are still at the research step, they have a promising development potential. Blue and green alternatives are close to being marketable according to their TRLs. However, they are not efficient, and they may dominate the market only during a transition period before a clean energy sector is completely developed. Meanwhile, we find that ammonia reveals better or at least the same performance compared with hydrogen. Therefore, when storing energy for the long-term is the target, using hydrogen to produce ammonia is better than its application as an energy storage alternative.

Although the results presented are helpful to facilitate technology selection and improvement, each storage technology is especially suited for some specific applications due to its design and characteristics. Therefore, the technologies reported as efficient are not necessarily the best choices for all the applications, and inefficient ones are not completely useless. For a practical application, several additional aspects will affect the project's social, economic, environmental, and technical implications and should be considered before making the final decision. We dealt with some of these concerns by doing further analyses like weighted DEA and monetization. Our results revealed that although the efficiency score and relative ranking of technologies report some changes, still the results obtained by the original DEA are reliable. Other aspects including the location of the storage unit, the availability of raw materials, cost and availability of skilled labors, market energy prices, geographical limitations, deployment and construction time, the technology response time, the complicity of manufacturing, minimum and maximum charging rate, maximum operating temperature, packing issues, ease of installation-operation-and control, needed accessories, ability to join with other technologies, application purpose, commercial availability of technology, its strength and durability, maturity, safety, grid connection impacts, recyclability and after use impacts also should be considered. Therefore, while this contribution provides a powerful framework for comparing storage technologies considering multiple sustainability dimensions through DEA, more comprehensive evaluations are still necessary.

Acknowledging that improving some technologies to make them efficient might take some time or even be impossible in some cases, it is suggested to design energy systems in a way that they do not depend strongly on storage technologies found inefficient in this contribution. In this context, policies promoting the use of the most efficient technologies through incentives might be as effective as regulations based on the taxation of the poorest performing options while alleviating the direct economic burden placed on companies and citizens. Further, the evaluation of hybrid energy storage technologies is an unexplored necessity that should be considered in future studies.

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.

CRediT authorship contribution statement

Fatemeh Rostami: Conceptualization, Methodology, Software, Writing – original draft, Visualization. Zoltán Kis: Conceptualization, Validation, Writing – review & editing. Rembrandt Koppelaar: Validation. Laureano Jiménez: Resources, Writing – review & editing, Supervision, Project administration. Carlos Pozo: Conceptualization, Methodology, Validation, Writing – review & editing, Supervision, Project administration.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ensm.2022.03.026.

Appendix A: Further analyses

In the evaluation presented in the main manuscript (models M1 and M2), the absence of predefined weights between indicators implicitly considers them equally important. However, in practice, stakeholders might be more concerned about certain facets of the sustainability problem. Bearing this in mind, we next present two additional analyses addressing the same problem of comparing energy storage technologies, but this time using different perspectives reflecting potential concerns from diverse stakeholders. In the first analysis, reported in Appendix A.1, we still use DEA but enforce environmental indicators to be more important than economic and social ones, thus prioritizing cleaner technologies for a sustainability transition. In the second one, provided in

Table A.1

Predefined weights for indicators of each cluster. LCOE: levelized cost of energy. GWP: global warming potential.

Indicators	Weight of indicators for the fast-response cluster (%)	Weight of indicators for the long-term cluster (%)
LCOE	10.00	10.00
Energy consumption	20.00	26.67
1/Energy density ¹	20.00	-
Water used	20.00	26.67
GWP	20.00	26.67
Employment	10.00	10.00
Total	100.00	100.00

¹ This indicator is not considered in the evaluation of long-term energy storage alternatives.

Appendix A.2, we explicitly account for the positive and negative externalities that would result from the deployment of each energy storage technology considered in this contribution.

Appendix A.1: Weighted-SBM

Without loss of generality, we explore a hypothetical situation where an environmentally conscious stakeholder assigns 80% of the total weight to environmental aspects and 20% to the rest (economic and environmental). We assume that these weights are divided equally between indicators within each sustainability dimension. For fast-response technologies, this translates to 20% weight for each of the four environmental indicators (i.e., 80/4=20). For long-term alternatives, each of the three environmental indicators is assigned 26.67% weight (i.e., 80/3=26.67). The dedicated weight to LCOE and employment in both clusters is 10%, as these are the only indicators in the economic and social categories, respectively. The weights considered are summarised in Table A.1.

Then, the undesired output slack-base model (i.e., model M1) is modified to consider these weights as parameters. The revised model (model M3 hereafter) is a weighted-SBM model presented in [43] and formulated next.

$$\begin{aligned} \tau^* &= \min t - \frac{1}{m} \sum_{i=1}^{m} \frac{W_i S_i}{x_{io}} \end{aligned} \tag{M3}$$

$$s.t. \ 1 &= t + \frac{1}{s_1 + s_2} \left(\sum_{r=1}^{s_1} \frac{W_r^g S_r^g}{y_{ro}^g} + \sum_{r=1}^{s_2} \frac{W_r^b S_r^b}{y_{ro}^b} \right)$$

$$x_o t &= X\Lambda + S^-$$

$$y_o^g t &= Y^g \Lambda - S^g$$

$$y_o^b t &= Y^b \Lambda + S^b$$

$$w_i^- &= 0.2i = energy \ consumption, \ water \ used, \ (1/energy \ density)$$

$$w_{r=GWP}^b = 0.2$$

$$w_{i=LCOE}^- &= 0.1$$

 $w_{r=Employment}^{g} = 0.1$

$$S^{-} \ge 0, \ S^{g} \ge 0, \ S^{b} \ge 0, \ \Lambda \ge 0, \ t > 0.$$

In this model, w_i^- , w_r^g , and w_r^b are the weights for input i, desired (good) output r, and undesired (bad) output r, respectively, as provided in Table A.1. The rest of the parameters and variables are the same as

model M1. Analogous changes can be introduced in the super-efficiency model M2. The weighted undesired output SBM and the weighted superefficiency model are solved for each DMU in the fast-response and longterm cluster for 100 scenarios. The scenarios were obtained by applying Monte Carlo sampling to the uncertain distributions associated with the sustainability indicators. The resulting median efficiency for each technology is compared with its median efficiency from the original DEA (i.e., equally important indicators) in Figs. A.1 and A.2, with the former comparing fast-response technologies and the latter benchmarking long-term energy storage alternatives.

Fig. A.1 shows the median efficiency obtained for each technology in the fast-response cluster from both analyses. The Y-axis presents the median efficiency when 80% weight is dedicated to the environmental indicators, while the X-axis presents the median efficiency when the indicators are weighted equally (i.e., the median efficiency from the original DEA). This figure is divided into four quadrants; (i) light green region: the technologies are efficient regardless of the weight considered for the indicators; (ii) dark green region: the fate of the technology is affected by the weight of the indicators, and it is efficient only when the indicators are weighted unequally; (iii) grey region: technologies are always inefficient regardless of the relative importance assigned to the different indicators; and (iv) dark green region: the fate of the technology is affected by the weight of the indicators and is efficient only when all the indicators are considered equally important. Also, using a diagonal line, technologies are classified into two groups: those that report a higher efficiency when no predefined weights are assigned to indicators (i.e., located on the right side of the diagonal), and those that benefit from larger weights to the environmental indicators (i.e., located on the left side of the diagonal).

As Fig. A.1 shows, using higher weights for environmental indicators results in important changes in the efficiency scores obtained. Technologies on the right side of the diagonal, namely flywheel, Ni-Cd, Li-ion, Na-S, and Li-Fe-Ph, see their efficiencies decrease from 2, 1.61, 1.36, 1.32, and 1.03, respectively, to 1.05, 1.04, 1.03, 1.03, and 1. This happens because increasing the weight of environmental indicators is equivalent to decreasing the weight of their most favourable indicator, which, in the case of the flywheel, Ni-Cd, Li-ion, and Na-S, is employment (see Fig. 5 for more information). Li-ion and Li-Fe-Ph also rely on their energy density to be efficient. It is an environmental indicator and increasing its weight benefits the efficiency score. However, this benefit is counterbalanced by the other environmental indicators and results in a lower median efficiency score. In any case, all these technologies remain efficient in this analysis, too, which demonstrates they are still robust options with a solid environmental performance.

On the other hand, the technologies on the left side of the diagonal, namely Li-Ni-Mn-Co, Na-Ni-Cl, LA, FB-ZB, SMES, and FB-VR, benefit from increasing the weight of environmental indicators. They improve their median efficiency scores from 0.98, 0.4, 0.24, 0.07, 0.05, and 0.03, respectively, to 1, 0.84, 0.81, 0.7, 0.66, and 0.57. These improvements are enough for Li-Ni-Mn-Co to become efficient. Despite improving their median efficiency scores, the other technologies stay inefficient, although they now have a better chance for competition against efficient technologies. Indeed, using predefined weights in the analysis produced a narrower distribution of efficiency scores than in the original case: from 1.05 (flywheel) to 0.57 (FB-VR), compared to 2 (flywheel) and 0.03 (FB-VR). However, none of the technologies deemed efficient in the original DEA stops being so in this new analysis. From a mathematical point of view, model M1 is a relaxation of model M3 because (implicit) weights can take any value to favour the technology under assessment. Considering that the efficiency scores provided by DEA are relative (and not absolute) efficiency measures, in practice, this means that model M1 can produce more distinct results across the technologies. This is done, for instance, by assigning a high weight to employment for some technologies since this allows them to achieve high-efficiency values. Remarkably, the relative ranking of the technologies is not affected by increasing the weights of environmental indicators, i.e., sorting the



X: Median efficiency in the original DEA

Fig. A.1. Comparing the median efficiency of fast-response energy storage technologies when the indicators are weighted equally and unequally. Grey region: always inefficient, Light green region: always efficient. Dark green region: inefficient when the indicators are equally weighted (i.e., X-axis) and efficient when the environmental indicators are weighted more (i.e., Y-axis), or vice versa.

technologies from highest to lowest median efficiency scores yields the same result as in the analysis presented in the main manuscript.

We next turn our attention to the long-term energy storage alternatives (Fig. A.2). Again, the X-axis presents the median efficiency of the original DEA, while the Y-axis reports the median efficiency when 80% weight is dedicated to the environmental indicators. This figure is equivalent to Fig. A.1, with the plotted area divided into four quadrants achieved by the alternatives regarding their efficiency scores (efficient vs inefficient). Similarly, the diagonal divides the alternatives into two groups, with alternatives on the left side of the diagonal benefitting from unequal weights and alternatives on the right side of the diagonal being harmed.

As shown in Fig. A.2, the alternatives on the right side of the diagonal, including green alternatives like hydrogen based on water electrolysis powered with solar (H_2 , WE-Solar) and wind energy (H_2 , WE-Wind), and ammonia based on water electrolysis powered with solar (NH_3 , WE- Solar), wind (NH₃, WE-Wind) and hydropower (NH₃, WE-Hydropower) see their efficiencies decline when higher weights are dedicated to the environmental indicators. This change is particularly remarkable for (NH₃, WE-Solar), with its efficiency score changing from 1.26 to 1.02. Despite this, all of them remain efficient, and this is a piece of evidence that they are reliable choices for environmentally friendly applications. Except for (H₂, WE-Solar), the rest of these alternatives are efficient mainly thanks to their employment generation capabilities. Therefore, the reduction in their efficiency is caused by a fall in the weight dedicated to employment. The fall in the employment weight results from increasing the weight of environmental indicators. Hydrogen production based on water electrolysis using solar energy (H2, WE-Solar) is mainly efficient because of its low water use (see Fig. 8 for more information). Therefore, the fact that its median efficiency decreases despite the higher weight assigned to its most-favourable indicator (i.e., water used) restates the necessity of improving its other environmental indica-



Fig. A.2. Comparing the median efficiency of long-term energy storage alternatives when the indicators are weighted equally and unequally. Grey region: always inefficient, Light green region: always efficient. Dark green region: inefficient when the indicators are equally weighted (i.e., X-axis) and efficient when the environmental indicators are weighted more (i.e., Y-axis), or vice versa. Grey H_2/NH_3 : H_2 or NH_3 produced using fossil fuels, Blue H_2/NH_3 : H_2 or NH_3 produced using fossil fuels, Blue H_2/NH_3 : H_2 or NH_3 produced using fossil fuels and the emitted carbon captured by CCS, Green H_2/NH_3 : H_2 or NH_3 produced using renewable energy sources.

tors (i.e., energy consumption, energy density, and GWP). The rest of the alternatives of the long-term cluster, which are located at the left side of the diagonal, enjoy a climb in their median efficiency compared to the results from the original DEA. This includes all the blue and grey options and power to hydrogen based on water electrolysis using hydropower energy (H₂, WE-Hydropower). Despite this, their improvements are not impressive, and none succeed in becoming efficient. Like fast-response technologies, the difference between the highest and the lowest median efficiency when unequal weights are used is lower (i.e., (1.02-0.73) vs. (1.26-0.23)). Also, in both types of analysis, (NH₃, WE-Solar) has the highest median efficiency, while (H₂, CG) presents the lowest median efficiency. The alternatives selected by the original DEA are attractive when social, economic, and environmental aspects are all essential. At the same time, the efficiency scores of this second "weighted" DEA look more reliable when environmental aspects are the most important.

Appendix A.2: Monetization of the indicators and monetized efficiency

Monetization uses predefined coefficients to translate environmental indicators into monetary terms. These coefficients are lower than one for some indicators and, therefore, decrease their value (e.g., water use, GWP), while the coefficients are greater than one for others, thus increasing the indicator values (e.g., energy consumption, Employment, and 1/energy density). In this way, monetization of the indicators helps to consider their relative importance from an economic point of view [121]. In this contribution, we monetize not only the environmental indicators but also employment. Employment provides the number of full-time equivalent jobs, as expressed in [number of jobs]. For its monetization, an average wage of 123816 \pm /year is considered [122], together with a currency exchange rate of US\$=0.85 \pm [123]. Then, the received wage for 30 years is estimated (i.e., 123816 \pm /year-0.85-30



Fig. A.3. Comparing the median efficiency obtained from original DEA using per GJ values of indicators (i.e., X) with the efficiency obtained using monetized indicators (i.e., Y) for fast-response energy storage technologies. Left side: technologies' ranking in X. Right side: technologies' ranking in Y. Short-term: flywheel, SMES. Medium-term: rest of the technologies. The number of positions that each technology climbs (+) or drops (-) in the monetized analysis compared with the original DEA is reported by the numbers in the parenthesis. The (=) presents the case that there is no change in the ranking of the technology. Green, blue, and red lines, respectively, present the climbed, maintained, and dropped ranking.

years=3157308). The coefficients used to transform the value of the indicators (i.e., the values reported in Tables 1 and 2) to their monetary values are presented in Table (B.14) of the supplementary information. As stated in the main body of the manuscript, using monetized indicators in a unit-invariant DEA model as the one employed in this contribution would not affect DEA results since monetizing the indicators is equivalent to changing their units. Hence, in this case, we avoid the use of DEA and calculate efficiency as the summation of the monetized desired output (i.e., employment) minus the summation of the monetized undesired output (i.e., GWP), divided by the summation of the monetized inputs (i.e., LCOE, energy consumption, energy density, and water used). This provides a measure of the absolute efficiency of the technologies, which is conceptually different from the relative efficiency provided by DEA to the point that it is no longer possible to classify units as efficient or inefficient based on a score of 1 or any other cutoff value. Finally, Figs. A.3 and A.4 show the results for fast-response and long-term clusters obtained using monetized indicators.

Fig. A.3 depicts the efficiency obtained using monetized indicators (i.e., Y) versus the median efficiency obtained from DEA (i.e., X) for each technology in the fast-response cluster. This figure is divided into two regions using the dashed line. On the left side, technologies are sorted according to their ranking in the original DEA, while on the right side, they are sorted based on their ranking in the monetized analysis. The signs and numbers inside the parenthesis beside the name of technologies introduce the changes in the ranking of technology in Y compared with X. The green line and a (+) sign reports an improvement in the ranking, while a red line and a (-) sign presents falling in the ranking. When the technology reports the same ranking, a blue line and an (=) sign are used.

Flywheel drops dramatically, mainly owing to its low energy density, being surpassed by seven batteries in the ranking. As reported in Table (B.14), the monetization coefficient of energy density is lower than one and decreases the indicator value. Note that, in this contribution, we use its inverse term (i.e., 1/energy density). Therefore, an initially low energy density that is dropped by monetization, results in a higher monetized 1/energy density, which finally leads to a lower monetized efficiency. Despite this, flywheel is still the best short-term alternative since SMES performs very poorly again (from second-to-last, to last). Among batteries, Na-S drops two positions while most of the rest improve their relative ranking, including Ni-Cd as the best performing. While Ni-Cd relies on its high employment and energy density to rank first, Na-S seeks for an increase on these indicators to improve its monetized ranking. Li-Fe-Ph, the next technology with a high value on its employment indicator, becomes the second-best option in the monetized analysis. The rest of the technologies, Li-Ni-Mn-Co, Na-Ni-Cl, LA, and FB-VR, improve their ranking by climbing from 6th, 7th, 8th, and 11th, respectively, to 4th, 5th, 7th, and 10th, while Li-ion and FB-ZB are the technologies that maintain their relative ranking.

Although in some cases, the technologies' ranking changes quite a lot in the monetized analysis compared with the original DEA, choices from the original DEA (i.e., flywheel for short-term and Ni-Cd for mediumterm) still stand as the preferred options. Despite this, this new analysis unveils the potential of other technologies such as Li-Fe-Ph, which might also have certain opportunities to infiltrate the market.

Fig. (A.4) is equivalent to Fig. A.3. On the left side of the dashed line, the alternatives are sorted based on their ranking in the original DEA (i.e., X), while on the right side, they are sorted regarding their ranking in the monetized analysis, which is marked by symbol Y. Similarly, an improvement in the ranking is presented by a green line and a (+) sign, the degradation with a red line and a (-) sign, and a maintained position with a blue line and a (=) sign. The number beside the sign accounts for the ranking changed.

As Fig. (A.4) shows, several "pairs" swap their ranking. Overall, green alternatives are still the best, but, with externalities, they are sorted according to the power source, with green H₂ always preferred over green NH₃ because of its higher employment generation. For instance, the rankings of (NH₃, WE-Solar) and (NH₃, WE-Hydropower) fall from 1st and 4th, respectively, to 2nd and 6th, while the rankings



Fig. A.4. Comparing the median efficiency obtained from original DEA using per GJ values of indicators (i.e., X) with the efficiency obtained using monetized indicators (i.e., Y) for long-term energy storage alternatives. Left side: alternatives' ranking in X. Right side: alternatives' ranking in Y. The number of positions that each alternative climbs (+) or drops (-) in the monetized analysis compared with the original DEA is reported by the numbers in the parenthesis. The (=) presents the case that there is no change in the ranking of the alternative. Green, blue, and red lines, respectively, present the climbed, maintained, and dropped ranking. Grey H₂/NH₃: H₂ or NH₃ produced using fossil fuels, Blue H₂/NH₃: H₂ or NH₃ produced using renewable energy sources. WE: Water Electrolysis.

of (H_2 , WE-Solar) and (H_2 , WE-Hydropower) climb from 2nd and 6th, respectively, to 1st and 5th. The opposite happens with blue and grey sources, where ammonia processes always rank above their H_2 -based counterpart. Also, all processes based on water electrolysis are placed on the top of the list. Overall, these results show significant agreement with those from the original DEA and, therefore, demonstrate the robustness of the conclusions drawn.

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