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The TWh challenge: Next generation batteries for energy storage and electric vehicles

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ABSTRACT

Energy storage is important for electrification of transportation and for high renewable energy utilization, but there is still considerable debate about how much storage capacity should be developed and on the roles and impact of a large amount of battery storage and a large number of electric vehicles. This paper aims to answer some critical questions for energy storage and electric vehicles, including how much capacity and what kind of technologies should be developed, what are the roles of short-term storage and long-duration storage, what is the relationship between energy storage and electrification of transportation, and what impact will energy storage have on materials manufacturing and supply chain. Accelerating the deployment of electric vehicles and battery production has the potential to provide terawatt-hour scale storage capability for renewable energy to meet the majority of the electricity need in the United States. However, it is critical to greatly increase the cycle life and reduce the cost of the materials and technologies. Long-lasting lithium-ion batteries, next generation high-energy and low-cost lithium batteries are discussed. Many other battery chemistries are also briefly compared, but 100 % renewable utilization requires breakthroughs in both grid operation and technologies for long-duration storage. New concepts like dual use technologies should be developed.

1. Introduction

The importance of batteries for energy storage and electric vehicles (EVs) has been widely recognized and discussed in the literature. Many different technologies have been investigated [1–3]. The EV market has grown significantly in the last 10 years. In comparison, currently only a very small fraction of the potential energy storage market has been captured [3,4]. There is still a large debate regarding the roles of different technologies and how they can be deployed, and what will be the research priorities for the community. For example, the estimated amount of energy storage need varies widely. Some analysis suggests that a few terawatt-hours (TWh) of storage capacity is needed [5], but seasonal variation requires long-duration storage of up to more than a month. The long-duration needs will significantly increase both the storage capacity needed and the cost of storage. The United States (US) Department of Energy (DOE) Energy Storage Grand Challenge sets a goal of $0.05/kWh for long energy storage [6], which is 3–10 times lower than what most of the state-of-the-art technologies available today can offer. There have been intense discussions of alternate technologies for long-duration storage, including new battery chemistries and hydrogen storage, but all these technologies have significant challenges, including difficulties in production, transportation and storage [7]. Lithium-ion (Li-ion) batteries are considered the prime candidate for both EVs and energy storage technologies [8], but the limitations in term of cost, performance and the constrained lithium supply have also attracted wide attention [9,10].

This paper provides a high-level discussion to answer some key questions to accelerate the development and deployment of energy storage technologies and EVs. The key points are as follows (Fig. 1): (1) Energy storage capacity needed is large, from TWh level to more than 100 TWh depending on the assumptions. (2) About 12 h of storage, or 5.5 TWH storage capacity, has the potential to enable renewable energy to meet the majority of the electricity demand in the US. (3) Accelerated deployment of standalone battery storage devices and EVs can play a critical role in meeting the TWh storage challenge. (4) New mechanisms need to be developed to manage the whole infrastructure, and the cost effectiveness and cycle life should be significantly improved. (5) The uneven distribution of materials and manufacturing is a

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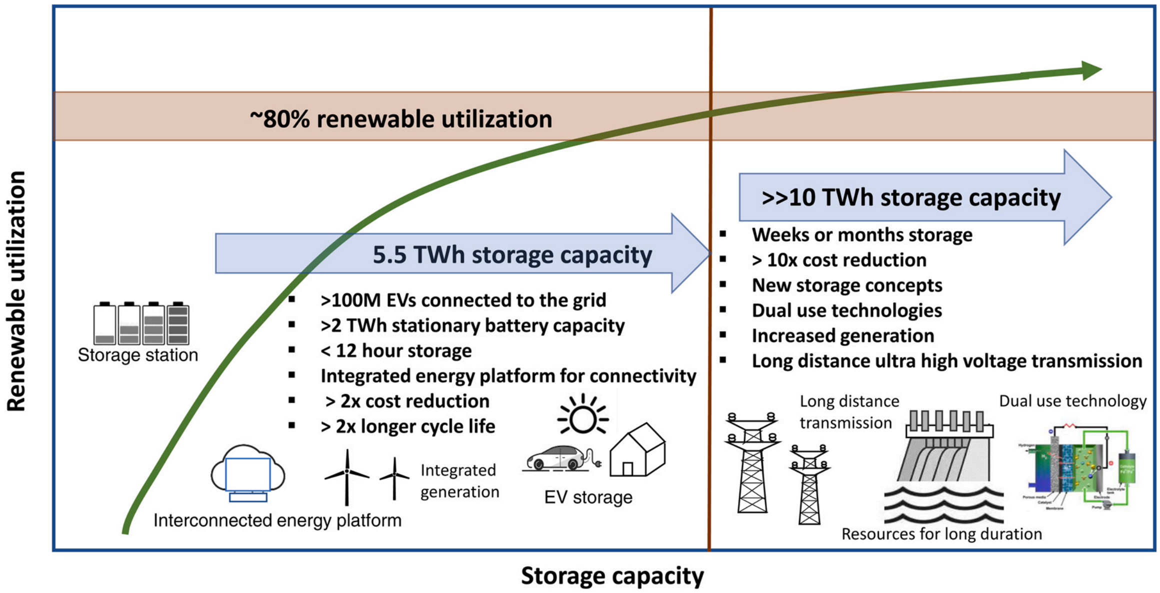

Fig. 1. Road map for renewable energy in the US. Accelerating the deployment of electric vehicles and battery production has the potential to provide TWh scale storage capability for renewable energy to meet the majority of the electricity needs. It is critical to further increase the cycle life and reduce the cost of the materials and technologies. 100 % renewable utilization requires breakthroughs in both grid operation and technologies for long-duration storage.

challenge for rapid deployment. (6) 100 % renewable utilization requires significant long-duration storage and breakthroughs in grid infrastructure and storage technologies.

It should be noted that energy storage is geographically dependent. This article is based on data and analysis available in the US, but the scientific principles and technical approaches discussed will be helpful for defining the future research directions and challenges in other regions.

2. Energy storage needs and cost

Renewable energy is fundamentally different from traditional sources and requires a paradigm shift from a centralized, top-down infrastructure to a distributed, variable infrastructure (Fig. 2) [11]. Renewable energy is highly variable and unpredictable. The generation is usually small and distributed over large areas. The generation is also location-constrained and weather-dependent. In addition, renewable generation is non-synchronous in nature. With an increasing level of renewable penetration, electric grid operation will need to address many issues, including ensuring power quality, providing adequate transmission and distribution capability, maintaining the stability of the system, and maintaining real-time balance of power supply and demand.

Energy storage is considered the most effective approach to addressing the variable nature of renewable energy. [1,7] Energy storage can provide a wide range of services, including improving stability and reliability, improving flexibility to manage renewables technology integration, improving grid resilience, economic efficiency, and deferring infrastructure upgrades.

The key question is how much storage capacity is needed and at what cost, and how to achieve the capacity. The amount of energy storage needed has been extensively investigated and the estimate covers a wide range. Earlier studies suggested that 10–20 % storage capacity will be needed for additional new generation capacity brought into the grid [12]. A recent study reported that several TWh of storage capacity will be needed for 43–81 % renewable penetration by adding together all the short-duration storage (< 12 h), but this value will be much higher if more than 80 % renewable penetration is reached with the need for long-duration storage (Fig. 3) [4]. The estimate for long-duration storage is more scattered. Traditionally, the stability of the power grid and the quality of the electricity (voltage, phase and frequency) are maintained by interconnecting large generators and

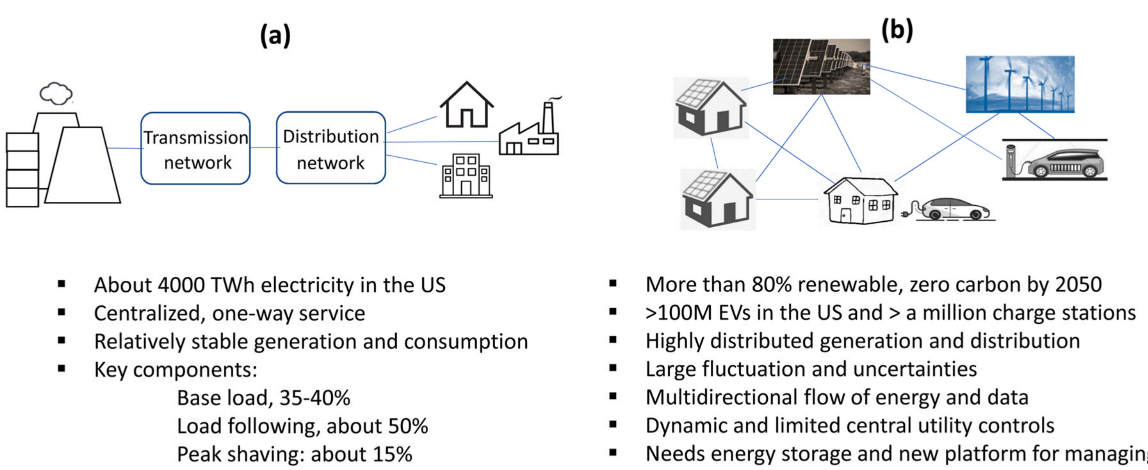

Fig. 2. Attributes of a traditional energy system and a distributed energy system. (a) Centralized, top-down energy system. (b) Distributed and variable energy system. More than 100 TWh energy storage capacity could be needed if it is the only approach to stabilize the renewable grid in the US.

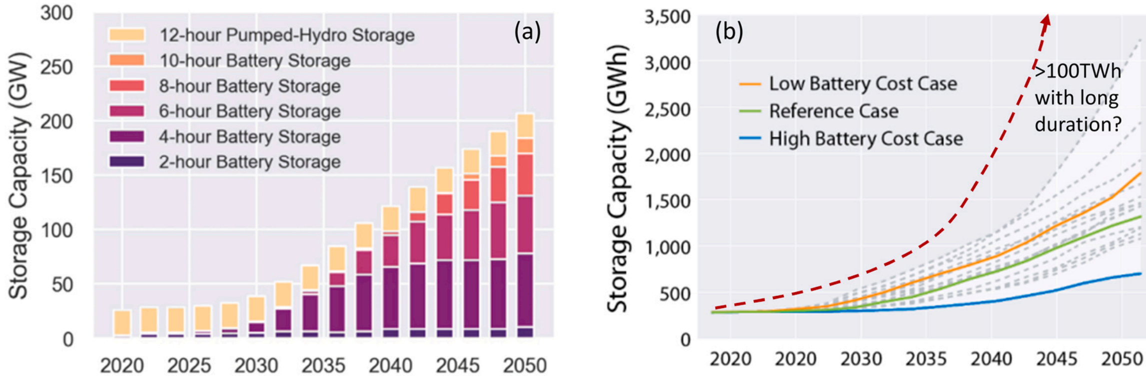

Fig. 3. US storage capacity prediction. (a) The estimated storage capacity (GW) for different short durations. (b) Short-term storage only requires a few TWh, but the total storage capacity (GWh) could be more than 100 TWh if energy storage is the only solution (reproduced from Ref. [5] with permission) [5].

carefully matching demand and production [13]. There are three different kinds of generations: (1) base load, the inviable portion of power generation; (2) load following, the generation that adjusts with the fluctuation of demand throughout the day; and (3) peak shaving, the short-term, fast-responding generation to meet demand at the peak hour. Base load is usually 35–40 % of the maximum load and is operated 24 h a day, but load following and peak shaving generation are only operated about 50 % and 15 % of the time, respectively. If more than 80 % generation is replaced by renewable energy, the same principles may not work anymore. Large storage capacity could be needed to stabilize the grid. Roughly 4000 TWh of electricity is consumed in the US per year. If only 10–20 % of storage capacity is considered, more than 100 TWh will be needed. Indeed, recent studies estimate that long-duration storage will require 85–140 TWh of energy capacity by 2040 that can store up to 10 % of all electricity consumed [14]. Providing more than 100 TWh storage capacity is a daunting challenge – not to mention the cost and performance requirements (discharge durations and number of cycles, etc.) – and cannot be easily done by storage alone. A large hydro dam like the Grand Coulee produces 20 TWh of electricity per year.

Cost is a critical factor in how different applications can be anticipated. Fig. 4 summarizes the projection of the capital and life cycle cost (LCC) or levelized cost for various storage technologies [4]. Overall, pumped hydro is the least expensive for large-scale applications at \$100-\$200 kWh^{-1}, but the service life is normally over 50 years. This makes the LCC extremely low, around \$0.05 (0.025–0.10) kWh^{-1} [15]. The problem is the geographic constraints. Currently, the most flexible storage technology is electrochemical storage using Li-ion batteries [16]. The cost of Li-ion batteries has been dramatically reduced (by an order of magnitude) over the last 10 years. The cell-level cost of Li-ion batteries is already less than \$150 kWh^{-1}, to about \$100 kWh^{-1}, a huge reduction from even a few years ago. The trend is still continuing today [17]. For energy storage, the capital cost should also include battery management systems, inverters and installation. The net capital cost of Li-ion batteries is still higher than \$400 kWh^{-1} storage. The real cost of energy storage is the LCC, which is the amount of electricity stored and dispatched divided by the total capital and operation cost [18]. Li-ion batteries have a typical deep cycle life of about 3000 times, which translates into an LCC of more than \$0.20 kWh^{-1}, much higher than the renewable electricity cost (Fig. 4a). The DOE target for energy storage is less than \$0.05 kWh^{-1}, 3–5 times lower than today's state-of-the-art technology. A combination of 2x cost reduction and 2x extension of cycle life could meet the DOE goal. Other important considerations include the service year and how frequently the storage is used, which are related to the storage duration. From Fig. 4b, if the storage is used two times a day (< 12-h storage), the LCC is quite acceptable after 10–20 years of service (< < \$0.10 kWh). If the storage is used for a long duration, i.e., over a period of more than 1 day, the LCC is exceptionally high even after 20 years (> \$1.00 kWh^{-1}). The cost needs to be reduced by more than 20 times depending on the duration, as shown here and pointed out in the literature [19]. Therefore, long-duration storage is extremely difficult not only because of the magnitude, but also because of the cost.

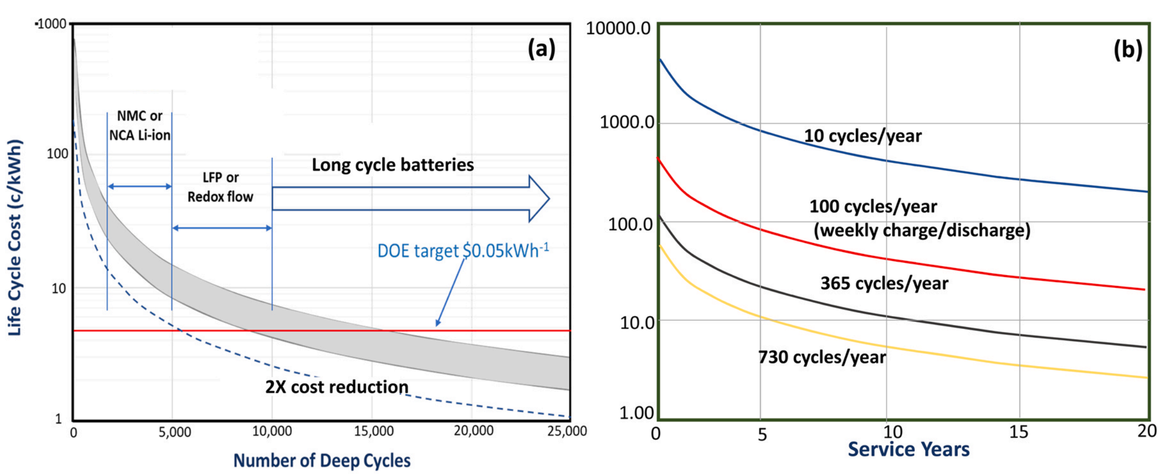

Fig. 4. Energy storage life cycle costs as a function of the number of cycles and service year. (a) Life cycle cost of batteries as a function of cycle life [4]. (b) Life cycle cost as a function of service years for different storage durations (the number of times a battery is charged and discharged in a year). Storage for more than 1 day would be very expensive, even after 20 years of service.

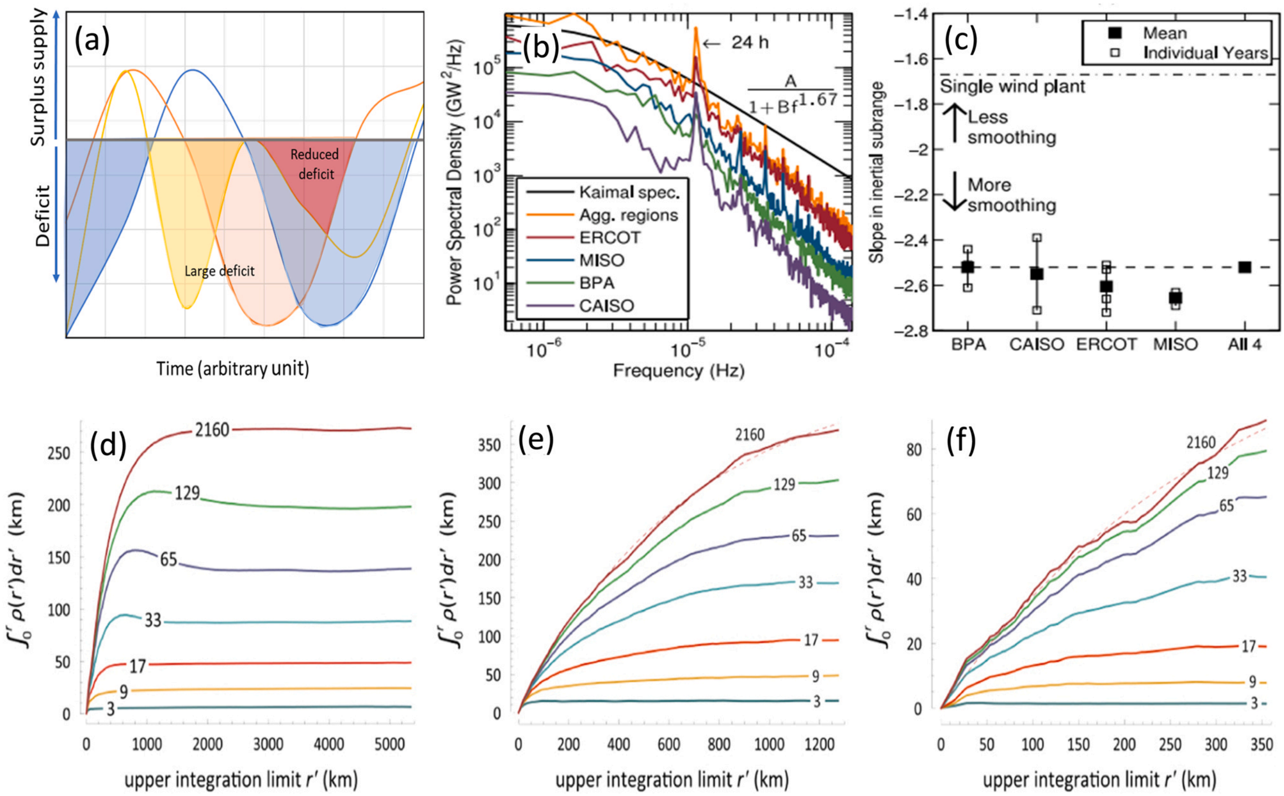

Fig. 5. Smoothing effect by combining different sources across different regions. (a) Additive effect of different hypothetical non-overlapping sources reduces the deficit [21]. (b) Power spectrum densities (PSDs) for 2009 wind power output of different regions in the US [23]. Five regions combined produce smooth power. (c) Slopes in the inertial subrange for each region and the interconnected regions for all years of available data and the means over time (reproduced from Ref. [23] with permission) [23]. The PSD slope in the inertial subrange reflects the relative proportion of fast- and slow-ramping units required to balance wind power output. (d) Integrated correlation of wind power in Canada with filter pass from 3 to 2160 h. (e) Integrated correlation of wind power in Australia with filter pass from 3 to 2160 h. (f) Integrated correlation of wind power in Bonneville Power Administration (BPA) with filter pass from 3 to 2160 h. The upper correlation length is 273 km for Canada, 368 km for Southeastern Australia, and 89 km for tower wind-speeds in the BPA region The correlation length for BPA region is underestimated because the saturation is not reached (reproduced from Ref. [25] with permission) [25].

3. Integrated approaches to reduce energy storage

The above discussion suggests that developing and deploying an exceptional amount of storage capacity to meet both short-term and long-term requirements is difficult and costly today. However, there has been plenty of research and analysis to separate the different needs for storage. In the real world, system fluctuations can be substantially reduced by integarting different resources (Fig. 5) [20]. Fig. 5a shows how three hypothetical sources added together can significantly reduce the deficit (yellow region) [21]. Typically, solar power reached its peak at mid-day while wind power did the opposite [22]. Similarly, the fluctuations can be significantly reduced if the generation from different regions can be aggregated (Fig. 5b and 5c) [23]. The power spectrum densities are an indicator of the fluctuation. Fig. 5c shows that integrating the wind generation of five regions in the US produces the least fluctuation. Fig. 5d shows that generation from all the generators in a particular region is always smoother than generation from an individual plant, judging by the ratio for fast to slow ramping. Other studies from other regions also suggest that mixed solar and wind can significantly reduce the fluctuation, and thus the needed storage [24]. The viability of solar and wind generations usually decrease exponentially with distance, with a characteristic correlation length of 200–500 km in North America (Figs. 5d–5f) [25].

These discussions suggest that the fluctuation is reduced by integarting different sources across different regions. While this is not a new idea, a close analysis can provide a more realistic estimate of the magnitude of the problem. Fig. 6 shows an analysis performed by Shaner et al. [26] This study assumed different ratios of solar and wind generation, with 1x and 1.5x of the total electricity generation capacity needed across a broad area, with and without 12-h storage capacity. It is noted that mixed generation of solar and wind in general improves the reliability across a large area because the fluctuations of solar and wind cancel the effect of each other to some extent compared to pure wind and solar (Fig. 6a). When 12-h storage is introduced, the reliability improves significantly, with the best result in the middle (60 % solar and 40 % wind). The overall reliability is better than 85 % over a large area (Fig. 6b). However, when the generation capacity is increased (1.5x), the reliability furthers improves significantly (Fig. 6c). With the addition of 12-h storage capacity, 90–99 % reliability can be achieved over large areas (> 10,000–1,000,000 km^2; 10,000 km^2 is roughly 100 km across) with the proper mixture of solar and wind (Fig. 6d). Therefore, this analysis provides a good lesson on how to achieve reliable electricity generation without resorting to a large amount of long-duration storage: aggregating excess solar and wind generation over larger areas and adding a reasonable amount of short-duration storage.

Shaner et al. further clarified how much electricity demand can be met with different generation and storage capacities. Fig. 7 shows that it is difficult to meet more than 60 % electricity demand without storage for pure solar generation, but with 12-h storage, the percentage met is increased to more than 90 % with 1x generation. Similar results are observed for 100–50 % solar (0–50 % wind). For 50–0 % solar (50–100 % wind), the storage also brings improvement, but the benefit is less pronounced. For much longer duration storage, the improvement is limited. This study also shows that it is difficult to meet 100 % electricity demand without resorting to large excess generation

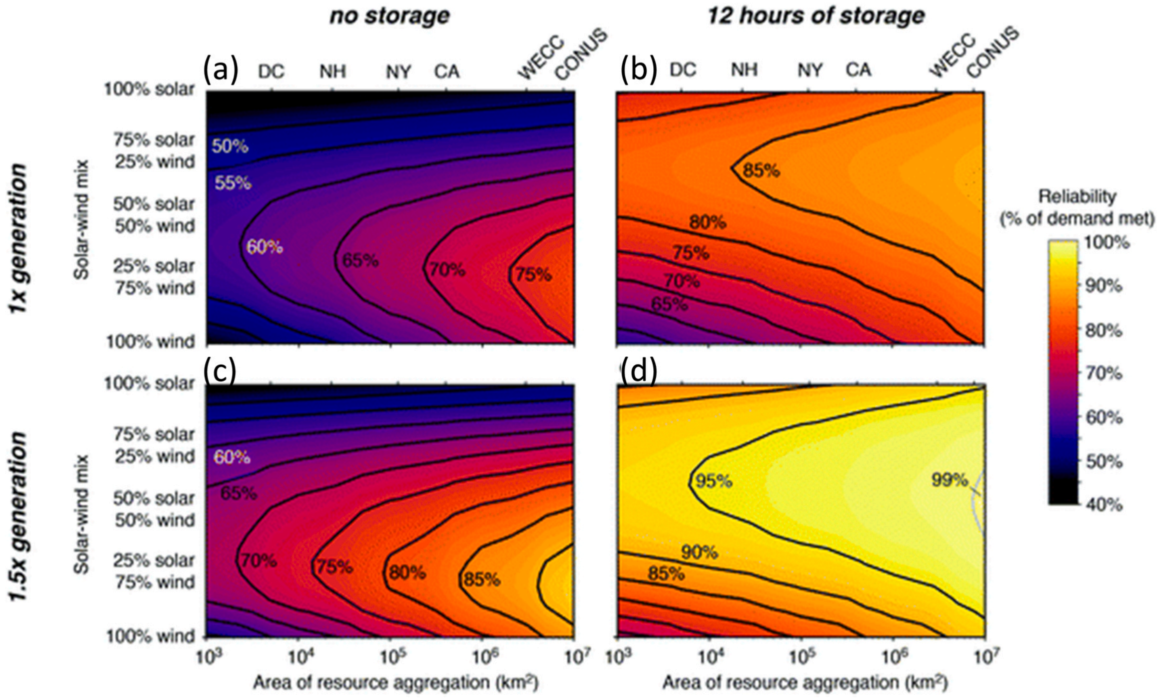

Fig. 6. Improved grid reliability with 1.5x solar and wind generation capacity and 12-h storage capacity (5.5 TWh). (a) 1x generation without storage showing poor reliability. (b) 1x generation with improved reliability using storage. (c) 1.5x generation without storage. (d) 1.5x generation with storage showing significantly improved reliability over large areas (reproduced from Ref. [26] with permission) [26].

capacity. Different storage concepts or grid operation ideas should be developed.

These results suggest that to meet ~80 % reliability, solar-biased, mixed generations can use energy storage to overcome the daily solar cycle, but wind-biased, mixed generation is more difficult. The results also suggest that the mixed generation can meet more than 80 % of electricity demand with modest energy storage capability in the US, but meeting 80–100 % electricity demand requires either long-duration storage or other measures to overcome the large, long-duration variations or unpredicted events.

Figs. 6 and 7 provide important insights for practical energy storage deployment: The deployment of renewable energy and energy storage should be considered in a synergistic way. The solar and wind generation should be aggregated over large areas to reduce the effect of

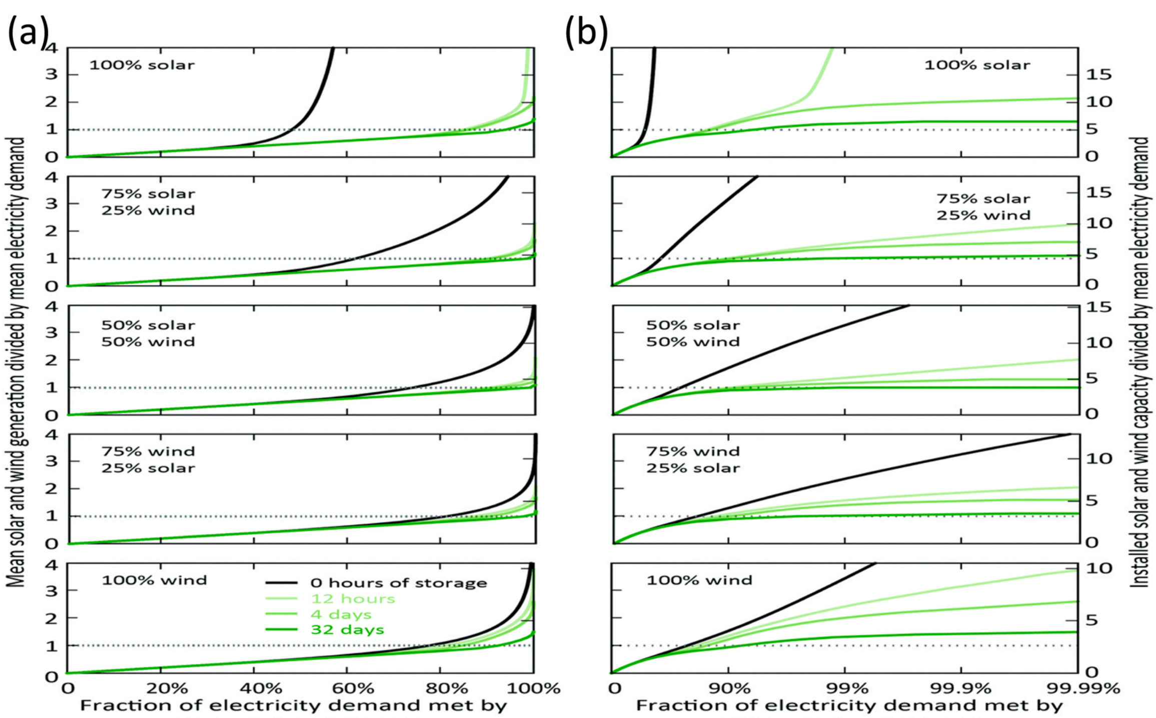

Fig. 7. Fraction of electricity demand met by different renewable generation [26]. (a) Plotted for 0–100 % electricity demand. (b) Plotted on logarithmic scale. More than 80 % electricity demand can be met using 12-h storage and combined solar and wind (reproduced from Ref. [26] with permission).

generation fluctuation. Under these conditions, a 12-h storage can play a critical role in the renewable energy meeting the majority of the electricity demand, but long duration and large capacity are still needed to meet 100 % demand and reliability. Annual electricity consumption in the US is 4000 TWh. A 12-h storage capacity equals 5.5 TWh. This estimate is close to some other estimates reported [5].

However, it is important to point out that this analysis only provides an overview of the larger picture. The generation, transmission and storage needs could differ from region to region, and the exact approach needs to be analyzed case by case and from year to year. The actual storage capacity needed to meet 100 % reliability and demand all the time can be much larger than a few TWh.

4. Getting the TWh storage capacity

The question is how to meet the 5.5 TWh storage challenge. Parallel approaches should be followed to maximize the benefits of all resources, including high renewable generation [26], batteries, pumped hydro, and compressed air if available. This article will focus on the potential and limitations of battery storage because of its flexibility for deployment at different scales.

Roughly about 300 million vehicles will be on the road in the US in the future. Many companies like General Motors, and some states, have already announced plan to go 100 % electric by 2030. The government is making it a high priority to achieve a goal so that more than 60 % of new car sales in the future is electric although the estimates differ from different sources (Fig. 8a). With a very aggressive strategy, one hopes to reach more than 120 million EVs in the fleet in 2050. Fig. 8b shows the predicted cumulative battery utilization capacity in the US. The EV needs are estimated to be 8 TWh in 2030, which should surpass 15 TWh in 2050.

Using EVs for energy storage has been discussed in the literature. Vehicles like the Ford F150 Lightning are designed to provide power to buildings. 120 million EVs will provide 12 TWh battery capacity. If 25 % of the capacity can be used for storage, the 120 million fleet will provide 3.75 TWh capacity, which represents a large fraction of the 5.5 TWh capacity needed. In addition, industry is ramping up battery manufacturing just for stationary and mobile storage applications. Some large manufacturers like Tesla's Gigafactory already have more battery sales for storage than for EVs. More than 2 TWh of batteries should be deployed for storage by 2050 (Fig. 8b). Under such conditions, 5.5 TWh storage capacity could be met by adding the capacities from EVs and stationary/mobile storage facilities.

However, using EVs for storage face serious challenges. One large concern is that frequent use for storage could cause degradation of battery life. To address this concern, the battery cycle stability needs to be greatly improved and the charge-discharge processes need to be carefully monitored and controlled. Modern EVs have a large battery pack, from 70 to 120 kWh nowadays for personal vehicles, which enables a range of more than 300 miles per charge. More than 90 % of people drive less than 100 miles a day. This implies that less than 1/3 of the EV battery capacity is being used daily. For an average household in the US, the electricity consumption is less than 30 kWh. A 100 kWh EV battery pack can easily provide storage capacity for 12 h, which exceeds the capacity of most standalone household energy storage devices on the market already. For the degradation, current EV batteries normally have a cycle life for more than 1000 cycles for deep charge and discharge, and a much longer cycle life for less than 100 % charge and discharge (Fig. 8c) [29]. For most storage applications over 1 day, one needs to ensure a shallow charge-discharge protocol is followed. If the charge and discharge processes can be automatically controlled so that the storage use does not deplete the battery capacity beyond a certain a threshold (50 %, for example), the impact on the battery life should be reduced [30]. Nonetheless, it is still critical to develop strategies to further improve stability and cycle life for batteries, a key point to be discussed later.

Estimating the exact cost of using EV as storage needs a careful analysis of the service provided by the battery and the impact on the battery life, and thus it is beyond the scope of this article. As discussed later, future batteries should achieve a cycle life of more than 10,000 cycles. A modern EV should last more than 10 years. It should also be noted that a cycle life of more than 10,000 cycles is already achievable for the shallow charge and discharge [28,29]. The cost of the battery needs to be reduced to less than \$100 kWh^{-1} and the cost of the whole battery system (including the battery management system, BMS) reduced to less than \$150 kWh^{-1}. The total battery system cost will be \$15,000 for a 100 kWh vehicle. For battery degradation, an arbitrary depreciation (20 % capacity degradation) value is assigned to the storage use (20 % of the battery cost) for 10 years, or \$3000. Another significant cost is the bi-directional charger, which is expensive today (up to \$5000 for an average household). The industry is already claiming less than \$1500 now, with the potential to further decline in the future [31]. For a typical household with a daily electricity use of 30 kWh, the amount of electricity dispatched to the home and to the grid will be limited to less than 50 % of the total battery capacity at any time (50 kWh), and will be controlled to a threshold value over certain period of time (for example, 50 kWh in a week). Then the total electricity dispatched is 2600 kWh in a year and 26,000 kWh over 10 years. The net revenue from the electricity generation will be \$2600 at \$0.10 kWh^{-1}. The net cost is \$1900. The final electricity cost will be the net cost divided by the electricity dispatched, which is \$0.07 kWh^{-1}. If the service life is extended to 15 years, the electricity cost from the battery storage will be only \$0.05 kWh^{-1}. Although this estimate is not accurate, it is a rough indication of the cost effectiveness of EV storage.

EV storage also needs a new ecosystem for operation because the EV users as well as the renewable sources are not synchronized or coordinated. There have been extensive discussions of the principles of vehicle-to-home and vehicle-to-grid operation [32,33]. A recent

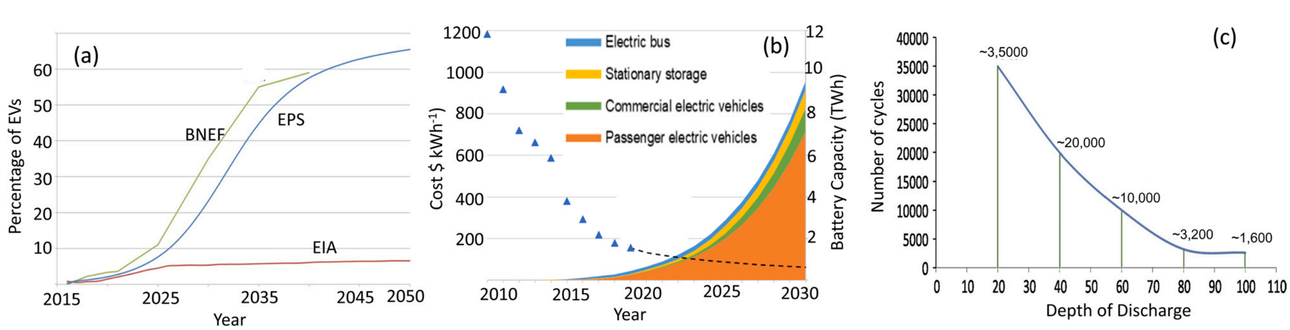

Fig. 8. EVs and batteries as assets for energy storage. (a) Predicted percentage of new car sales in the US (EIP: Energy Information Administration; EPS: Energy Policy Simulator; BNEF: Bloomberg New Energy Finance) Reproduced from Ref. [27] with permission from Energy Innovation Policy & Technology LLC) [27]. (b) Predicted cumulative battery capacity in the US. The blue triangles are the cost trend (Reproduced from Ref. [27] with permission) [5]. (c) Lithium iron phosphate battery cycle life as a function of depth of discharge (reproduced from Ref. [28] with permission) [28].

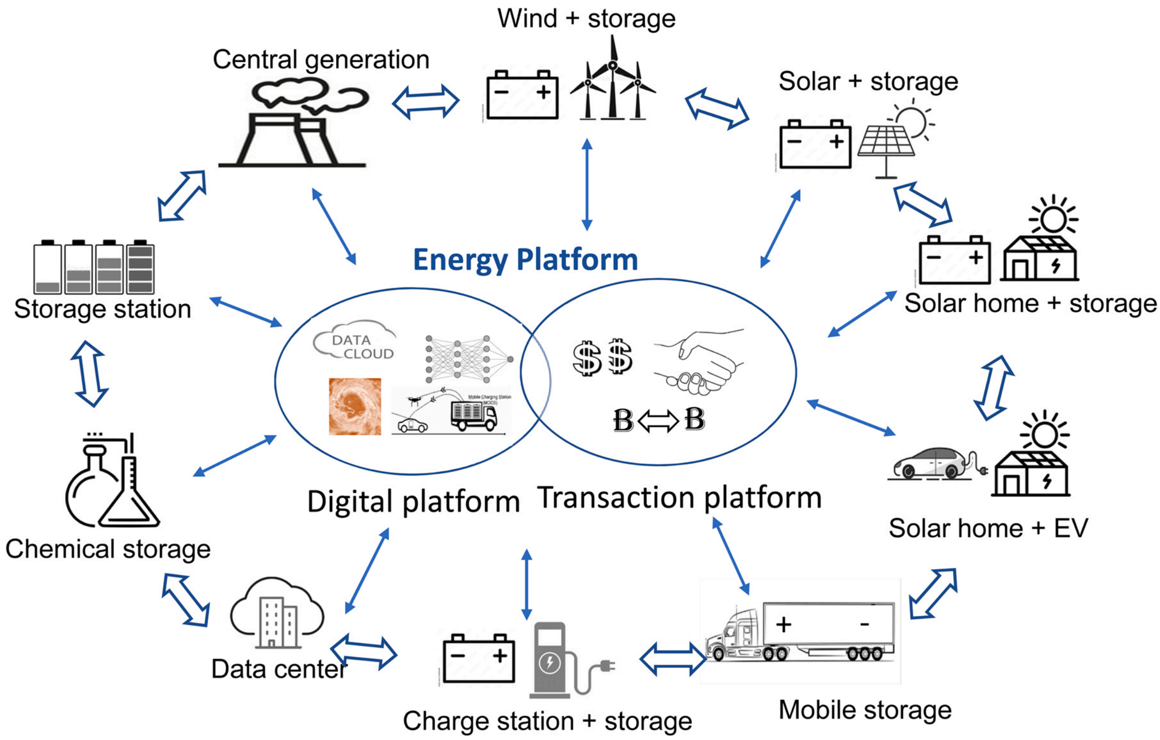

Fig. 9. Energy network to enable EV and other storage technologies. New energy platforms need to be developed to manage the generation, storage and demand at the same time [4].

analysis illustrated that the use of EVs for energy storage is more than an order of magnitude cheaper than building new storage capacities [34]. The EVs can be viewed as a collection of many small generation sources. A new ecosystem needs to be developed to integrate and connect the distributed sources including the mechanisms to connect to the grid and control the charge-discharge process, cost-effective intelligent bi-direction chargers, and the transaction mechanisms and business model. It is proposed that the energy infrastructure can be controlled by a platform-based approach, the energy platform [4]. The energy platform connects and controls the real (hard assets) and virtual network (digital assets) (Fig. 9). The hard assets include the energy production, transmission and distribution infrastructure, energy storage facilities, EVs, smart meters, controls, bi-directional chargers, etc. The digital assets include energy production data, energy consumption data, weather and climate data, data management and cloud services, and computational and mathematical tools. The energy platform can be used to gather real time data of the generation sources, custom demand, and the status of the vehicles, then make decisions to allow either the charging or dispatching process (discharge) to take place. The energy platform should also provide the capability for customers to participate in peer-to-peer electricity trading and monetize their investments. A new service model should be provided to the customer, like packaged service depending on whether the customer is a pure consumer or has the capability for storage (batteries, EVs), generation (solar) and dispatch. Different business models have been discussed in the literature [4,35]. In addition, cost-effective power electronics like intelligent bi-directional charger systems should be developed and implemented [36].

5. Long cycle life batteries

Based on the discussion in this paper, a high priority for storage applications is to significantly increase the cycle life of the batteries. There are several strategies to increase the cycle life, i.e., improving the electrode materials and electrolytes, choosing different battery chemistries, and improving cell design, manufacturing, quality control and battery management. Currently, widely investigated approaches for electrode materials include the development and deployment of single crystalline electrode materials [37,38], concentration gradient electrode materials [39,40] and surface coatings [41,42] to reduce interfacial reactions and cracking during long cycling. Much longer cycle life can also be achieved by improving the environment of the batteries and by better control of the charge and discharge processes [30].

For different applications, it might be necessary to have different designs for high-energy cells and long cycle cells. For example, lithium iron phosphate (LFP) batteries are more stable and have a longer cycle life than other transition metal oxide-based batteries (Fig. 10a) [43]. It has been demonstrated that LFP batteries can achieve more than 10,000 stable deep cycles on the cell level. If such technologies can be optimized to obtain even longer cycle life, and if the technology can be scaled up for large commercial applications, the energy storage cost could be reduced significantly for long cycle applications. The LFP battery also reduces the pressure on the supply chain in transition metals.

Because LFP has been commercially used for some time, currently there is less attention to the fundamental degradation mechanism in such materials and cells. Some reports suggest that the loss of active lithium from the cathode is the main degradation mechanism [44], but this finding needs to be verified for long cycle life applications because the degradation strongly depends on the operation conditions, temperature and charge-discharge rate, among other factors. Prelithiation to mitigate the loss of lithium has been investigated, but most processes investigated are difficult to implement and scale up [45]. The batteries can also suffer from other degradation processes like conversion of active materials into inactive materials, including the dissolution of the metallic iron species [46]. Many new in situ techniques developed for lithium metal batteries [47], particularly those developed to quantify active materials loss, should be applied to investigate the fundamental degradation processes in LFP systems such as lithium loss and iron dissolution. Scalable processes for treating the electrode materials and for cell design should be developed to prevent long-term degradation.

The cycle life is also connected with the charge-discharge rate. Fast charging (the ability to achieve 40 % of the charge state-of-charge in

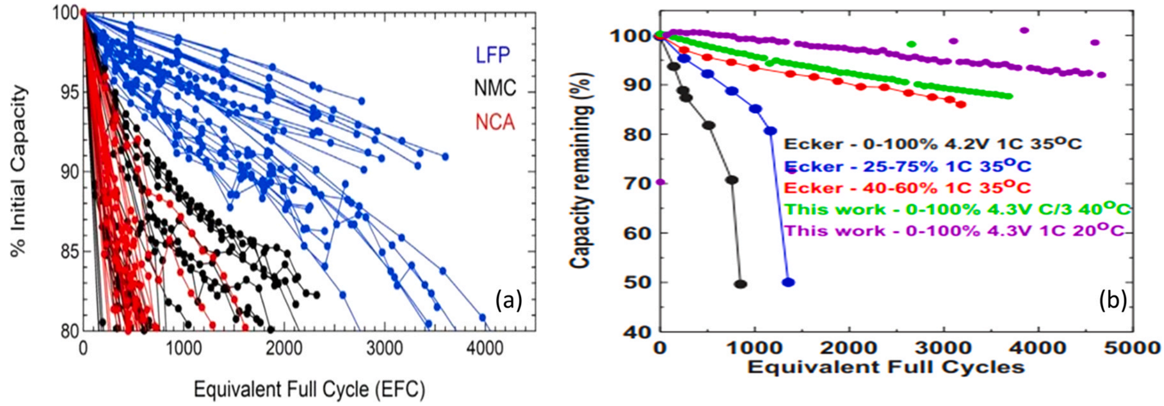

Fig. 10. Cycle life of different batteries. (a) Comparison of LFP and NMC batteries with different cycle lives (reproduced from Ref. [50] with permission). (b) Optimized NMC cells for extend cycle life (reproduced from Ref. [29] with permission).

15 min) and discharging is desirable, but may also cause degradation of the cycle life [48,49]. This effect must be included when considering long cycle life cells. Many strategies investigated for long cycle life, such as developing materials and electrolytes with fast charge transport, stable architectures, and stable and highly conductive solid electrolyte interphases (SEIs), are also applicable to fast charge cells [48].

There is also significant potential to improve the longevity and cycle life of Li-ion batteries by changing the battery design and operating conditions. For example, recent studies prepared and tested batteries made of single crystalline lithium manganese cobalt oxide (NMC) cathode and graphite (Fig. 10b) [29]. This study concluded that by modifying the electrolyte additives and optimizing the maximum voltage the cell is charged to, the battery life can be improved by more than one order of magnitude. Such studies provide good lessons on developing principles for batteries for energy storage with exceptionally long lives.

6. Next generation high-energy and low-cost batteries

Over the last two decades, the specific energy of Li-ion batteries has been significantly increased while the cost has dramatically decreased. With better electrode materials such as high-nickel lithium nickel manganese cobalt oxide (high-Ni NMC) and carbon/silicon composite anodes, Li-ion batteries are reaching a cell-level specific energy higher than 300 Wh kg^{-1} [51,52]. At the same time, the cost has been reduced to close to $100 kWh^{-1}. The scientific community and industry are actively pursuing next generation materials and cell designs that will overcome the limits of current Li-ion batteries in terms of materials supply, energy density and cost. Notable examples include the pursuit of methods to replace the graphite anode with lithium alloys [53] or lithium metals [54], and solid electrolytes and solid state batteries [55]. Among these approaches, lithium metal anode with liquid electrolytes or solid electrolytes has attracted the widest attention (Fig. 11a) [54,56]. Rechargeable lithium batteries have the potential to reach the 500 Wh kg^{-1}, and less than $100 kWh^{-1} goal. In the last several years, good progress has been made in the fabrication of high-energy lithium cells and good cycle life has been achieved using liquid electrolytes [57]. The industry and the scientific community are also working on solid state batteries with lithium metal [58,59], but in most cases a small amount of liquid is used. Pure solid state batteries have not been demonstrated for practical cells except for polyethylene oxide cells that operate at more than 60 °C [60].

For lithium metal batteries, a long-standing problem is dendrite formation [61] related to SEI structure and reaction mechanisms. [62] Dendrite formation is regarded as the most important cell failure mechanism. However, there are many pathways for the lithium metal batteries to fail, depending on the cell parameters. For example, in a typical high-energy pouch cell, the cell is more susceptible to other failure mechanisms, particularly through depletion of electrolytes and the formation of a dry SEI layer, causing sudden cell death (Fig. 11b) [57]. To make things worse, the distribution and depletion of electrolytes may not be uniform in the real cells, and the local drying accelerates the cell degradation (Fig. 11c) [63].

Another widely explored topic is three-dimensional (3D) architectures to stabilize the anode [64]. However, there have been few studies actually implementing 3D architectures into real cells and demonstrating the effectiveness [64]. In many cases, the advantages or limitations of 3D architecture may be masked by other, more important factors. Fig. 12a–d shows an example of the lithium metal cell properties with different carbon hosts and different electrolytes [65]. Three different carbon hosts were used: hard carbon, mesoporous carbon and surface functionalized mesoporous carbon. Based on results reported in the literature, these different hosts should produce very different behaviors. However, under realistic conditions for more than 300 Wh kg^{-1} cells, all three carbon materials showed very poor coulombic efficiency. When a good electrolyte is used, all three carbon materials demonstrate high efficiency, and there is little difference between the different materials. These results suggest that the true material properties cannot be revealed unless realistic conditions are used. The true cycle stability can be only observed in real cells (Fig. 12e).

Finally, solid electrolytes are widely discussed as means to suppress dendrite growth or to make solid state batteries. This is a very promising direction for lithium batteries [66], but so far there have been few reports on how the solid electrolytes can be incorporated into practical cells [67]. Besides limitations from the intrinsic properties, there is an urgent need to develop manufacturing techniques to make large and ultrathin (< 50 μm) solid electrolyte materials with high uniformity, robust mechanical property and flexibility, good chemical stability, and stable and conformal interfaces with both the cathode and anode materials. Without breakthroughs in this area, it is difficult to validate many of the hypotheses about solid electrolyte batteries, not to mention the scaling up and manufacturing of the materials and devices in a practical way.

7. The supply chain

Finally, one needs to answer the question of whether the large battery capacity can be constrained by the limitations of the supply chain. There have been extensive analysis and numerous reports on the supply chain problem in lithium-based batteries, but the conclusions vary greatly. It has been widely reported in the news media that there will be a large gap between the demand and supply by 2025 or so. However, rigorous analysis in peer referred literature is more indicative of the real challenges in the supply chain. A recent analysis predicts that there will be a very tight and delicate balance between the supply and demand for a long time (Fig. 13a) [69]. With new mining, extraction and processing technologies, the lithium itself may not be the

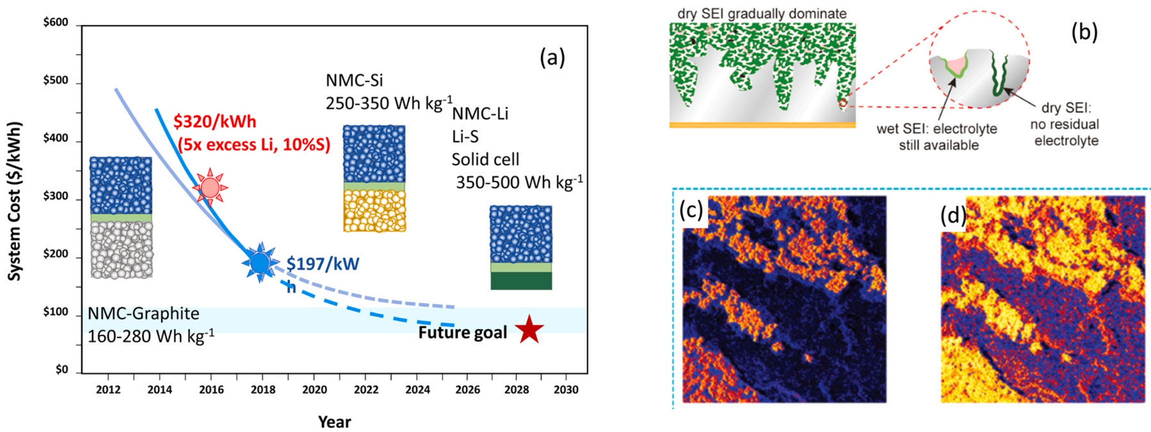

Fig. 11. Next generation high-energy low-cost batteries. (a) The roadmap for the Department of Energy (modified with permission from Tien Duong and David Howell, DOE VTO Office). (b) SEI structure in Li metal pouch cells. The SEI is different with different cell designs. Electrolyte drying and formation of dead SEI are often the main degradation mechanism [57] (c) and (d) Uneven distribution of electrolyte and reaction species in lithium-sulfur (Li-S) pouch cells (reproduced from Ref. [63] with permission) [63].

bottleneck even with a much accelerated deployment of EVs up to 2 billion units. Another study suggests that the lithium supply may be sufficient for up to 1 billion EVs but reaching 2 billion will be difficult [70]. For transition metals like nickel, cobalt and manganese oxide cathodes, nickel and manganese may not be a serious problem, but the cobalt demand could exceed the available supply. However, the real challenge is what happens in the marketplace in the world. In the last decade, the lithium prices (lithium carbonate and lithium hydroxide) have fluctuated over a wide range, from a few thousand dollars per ton to more than twenty thousand dollars per ton. Similar price fluctuations have also occurred with other transition metals. Such wide fluctuations in prices and supply chain are detrimental to the manufacturing industry. In addition, it is widely recognized that the resources and the production capacities are not evenly distributed in the world [71,72]. Chile, Argentina, Australia and China produce the majority of the lithium in the world. Most manufacturing capabilities are located in Asia (Fig. 13b). Such uneven distribution causes serious stress on the materials manufacturing and supply chain. The problems in the supply chain makes it important for the scientific community and industry to pursue alternate battery chemistries like LFP or sulfur (S) cathodes (Li-S batteries), as well as non-lithium based batteries and recycling [73].

8. Alternate battery chemistries and challenges beyond batteries

Currently, a wide range of battery chemistries are being investigated to improve the energy density and safety of batteries, reduce cost and improve supply chain resilience. Table 1 summarizes the key attributes of these batteries. Notably, Li-ion batteries still provide the best balance of performance and cost, but some different battery forms like redox flow batteries (RFBs) are also being deployed at MWh scales. For comparison, Table 1 also includes thermal storage. Thermal storage can be deployed at large scales and the storage materials are inexpensive (less than \$15 kWh^{-1}, over 10,000 cycles, with a low energy density), but energy conversion between thermal energy and electricity is inefficient and expensive [75].

The three most studied battery chemistries are sodium-ion (Na-ion) batteries, RFBs and Zn-ion batteries (Fig. 14). The different attributes of these batteries are compared with Li-ion batteries (Fig. 14a). Currently, Na-ion batteries have attracted wide attention because they essentially work based on the same principles as Li-ion batteries but replace lithium with sodium to eliminate lithium dependance [2,76]. Such batteries are also manufactured in the same way as their lithium counterpart, and therefore can be a true drop-in replacement for Li-ion batteries. However, Na-ion batteries inherently have a low energy density, which usually leads to a higher cost just because more materials and batteries need to be manufactured to meet the same demand. In addition, the stability of the cathodes, anodes, electrolytes and separators is still behind those of Li-ion batteries, leading to poorer stability and shorter cycle life. Sodium intercalation materials are also less stable than lithium intercalation materials [77]. The ideal anode material graphite in Li-ion batteries does not work with sodium chemistry. Instead, hard carbon, which is a disordered form of graphite, is mostly

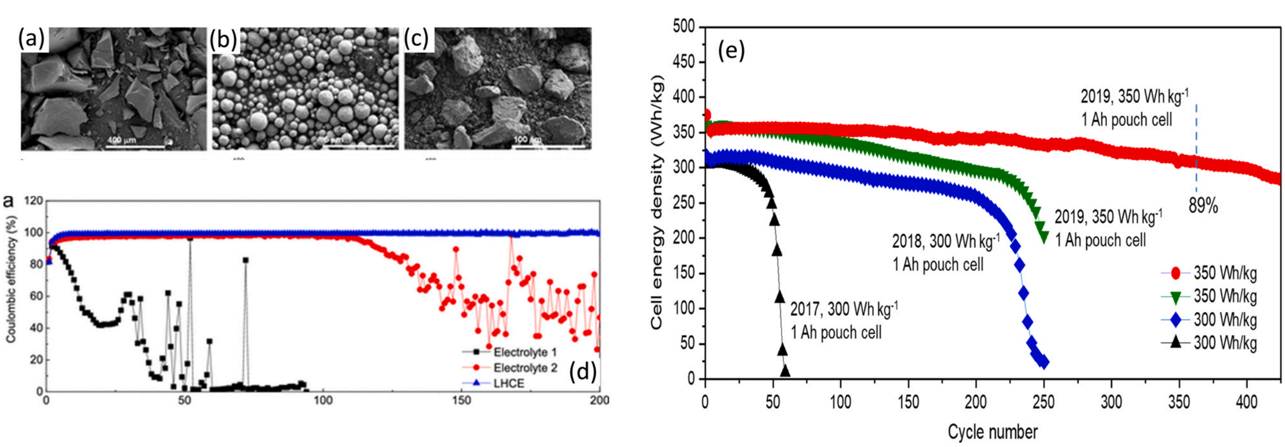

Fig. 12. Electrochemical properties of NMC-Li batteries (reproduced with permission from Ref. [65] copyright ACS). (a) to (c) Different carbon hosts, hard carbon, mesoporous carbon and functionalized mesoporous carbon. (d) Coulombic efficiencies for Li depositions. All three carbon materials showed poor stability in poor electrolytes and good stability in good electrolytes. (e) Progress of cycle behavior of pouch cells supported by Battery500 [68].

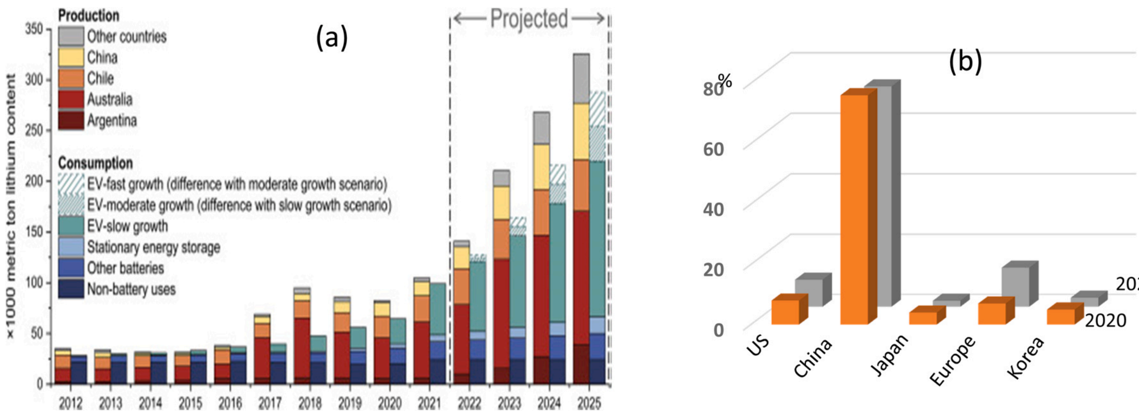

Fig. 13. The manufacturing and supply chain problem. (a) Tight balance between lithium supply and demand (reproduced from Ref [69] with permission). (b) Cell manufacturing capacity by country and region (replotted from Ref [74]).

Table 1
Comparison of the properties of different batteries.

Battery	Energy	Cycle	Supply	Cost	Safety	Scale-up	Comments
LFP	★★★★	★★★★★	★★★	★★★★★	★★★★★	★★★★	Great for energy storage.
LIB	★★★★★	★★★★	★★	★★★★	★★★★	★★★★	Good balance of performance.
NIB	★★★	★★	★★★★★	★★★	★★★	★★★★	Property and cost not as good as LIB, but helps supply chains.
VRB	★★	★★★★★	★★	★★★★	★★★★★	★★★★★	Large scale storage. V is problem. Long stability needs to be validated.
Fe-CR RFB	★★	★★★	★★★★★	★★★	★★	★★★★★	Need careful control of reactions.
ORB	★★★	★	★★★★★	★★★★	★★	★★★★	High energy redox flow system, but most agents are not stable.
H_2O ORB	★★	★★	★★★★★	★★★★★	★★★★★	★★★★★	Need to find stable redox agents.
ZIB	★★	★★	★★★★★	★★★	★★★★★	★★★★	Long time stability not demonstrated in large systems.
SSB	★★★★★	★★	★★★	★★	★★★★★	★	Manufacturing is hard.
Li-S	★★★★★	★★	★★★★	★★★	★★★	★★★	Cycle stability needs to be improved.
MIB	★★★	★★	★★★★	★★★	★★★★	★★★	No good intercalation cathode is found.
Li-O	★★★	★	★★★	★★	★★	★	Stable cells not demonstrated.
Pb-acid	★★	★★	★★★★★	★★★★	★★★★★	★★★★★	Cheap but not long lasting. Pb is not welcome.
Pb-C	★★★	★★★	★★★★★	★★	★★★★★	★★★★	Good cycle life but becomes expensive.
Na-S	★★★	★★★	★★★★★	★★★	★	★★	High temperature operation is a problem.
Thermal	★	★★★★★	★★★★	★★	★★★★★	★★★★★	Low cost and scalable, but energy conversion is not efficient.

(LFP: lithium iron phosphate cells. LIB: Li-ion batteries with lithium nickel manganese cobalt oxide (NMC) or lithium nickel cobalt aluminum oxide (NCA). NIB, sodium-ion batteries. VRB: vanadium redox flow batteries. Fe-Cr VRB: iron chromium redox flow batteries. ORB: organic redox flow batteries. H_2O ORB: aqueous redox flow batteries. ZIB, zinc-ion batteries. SSB: solid state batteries. Li-S: lithium-sulfur batteries. MIB: magnesium-ion batteries. Li-O: lithium-oxygen batteries. Pb-acid: lead acid batteries. Pb-C: lead carbon batteries. Na-S: sodium sulfur batteries. Thermal: thermal storage.)

used. The hard carbon is currently more expensive, and is tied to the poor stability with the electrolyte [78]. Still, many of the problems with Na-ion batteries are not fundamental. There should be steady progress, improvement and gradual market adaption in the next few years, but it will be hard for Na-ion batteries to directly compete with Li-ion batteries in terms of cost and performance for now.

RFBs are another widely studied technology [80]. RFBs are made of electrolytes (a catholyte - a liquid cathode material and an anolyte - a liquid anode material) stored in large tanks rather than the solid cathode and anode materials used in traditional batteries. The electrolytes are pumped into the stack, where the redox reactions produce electricity. There are several benefits of RFBs. First, the active materials in the electrolyte can be either organic or inorganic species, and the numerous options can provide real solutions to the materials supply chain problem. Second, the amount of energy stored depends on the amount of the electrolyte stored in the tanks, and therefore the technology is readily scalable. It is much easier to construct megawatt-size RFBs compared to other options, particularly for longer duration storage. The energy density is separate from the power of the device, which is determined by the stack size, or the contact area determined by the separation membranes. Third, the electrolytes can be aqueous. Aqueous electrolytes are non-flammable, and largely reduce the safety concern. The history of RFBs is as long as that of Li-ion batteries, and there have been many demonstration projects with MWh systems for

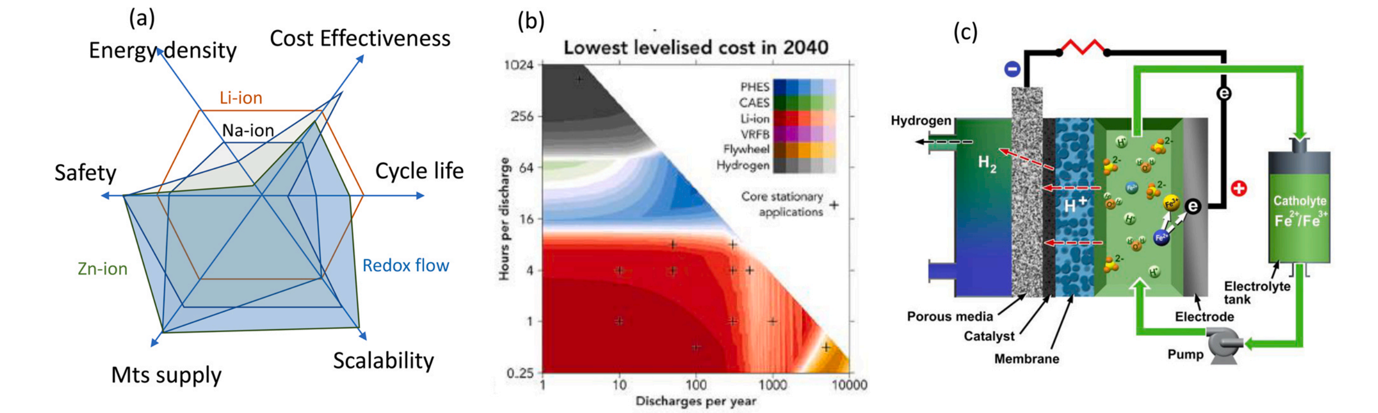

Fig. 14. Alternate approaches for storage. (a) Comparing attributes of Li-ion, Na-ion, redox flow and Zn-ion batteries. (d) Benefit of hydrogen storage for long duration (reproduced from Ref. [79] with permission). (c) Conceptional sketch of a dual use battery. The dual use device can produce large amount of hydrogen and store it for long-duration discharge when needed, and functions as a battery for short duration energy storage.

energy storage. Overall, RFBs have a much lower energy density than Li-ion batteries (about 1 order of magnitude lower) because the energy density is limited by the solubility of the active species in the electrolytes. Although the cost has been significantly reduced, and an excellent cycle life has been reported, RFB systems are still more expensive than Li-ion batteries based on the best available data reported, and their long-term reliability has yet to be proven. The longevity of this technology needs to be carefully considered because it contains large amounts of corrosive, super-concentrated electrolytes and complicated plumbing and pumping systems. If not carefully maintained, the system can crash or leak due to small fluctuations in temperature and other operation conditions.

Currently, aqueous vanadium redox flow batteries (VRBs) are the most mature type of RFB, and very large storage projects are under construction in different parts of the world [81]. Therefore, VRBs are a good candidate for energy storage, in particular for large storage systems, but vanadium is not exactly environmentally friendly, and vanadium supply and prices have been subject to large fluctuations in the past. Other redox battery chemistries such as iron chromium and pure iron systems have also been explored and commercialization efforts are also underway. These systems do not depend on vanadium, but the reactivities with iron or chromium species are much more difficult to control [82]. The long-term promise may be in pure organic RFBs. The choices of organic molecules are almost endless, and it is easy to perform molecular-level surgery on the molecular structure and properties [83]. There have been very encouraging reports on using advanced computational tools to discover new and promising organic materials for RFB applications, but so far mutually stable organic cathode and anode materials to enable stable long cycling have not emerged.

Alkaline zinc batteries have been in the marketplace for a long time, and they are very inexpensive because of their materials abundance and easy fabrication. In the last few years, there has been significant interest in making alkaline zinc batteries rechargeable (Zn-ion batteries) and using them for energy storage [84]. The zinc battery system is aqueous and somewhat resembles what happens in lead-acid batteries [85,86]. In addition to the movement of Zn ions, the electrode materials also go through a series of complicated dissolution and precipitation reactions, particularly with a large current [86]. The aqueous electrolyte (water) can also be decomposed to generate hydrogen (electrolysis). Therefore, although there have been numerous publications on Zn-ion batteries, these results are mostly realized under laboratory environments and commercially viable products are still underway.

The final challenge for renewable energy is meeting the last 10–20 % of the electricity demand with more than 90 % reliability. As discussed already, this is a very difficult goal and cannot be met by storage alone. For example, if more renewable generation can be realized, and other resources like pumped hydro and tools to control demand and response are available, the need for storage will be largely reduced. In fact, the renewable generation cost is already approaching \$0.03 kWh^{-1} [87], much cheaper than long-duration storage, so building excess generation capacity can be economically viable. Furthermore, if the generation sources can be coordinated over long distances (from east to west, for example) with ultra-high voltage transmission lines, the need for storage can also be substantially reduced [88]. Still, large-scale long-duration storage will be needed to certain levels.

At this time, all the battery technologies investigated for large-scale applications are based on the assumption that the materials are inexpensive and abundant, but none of these battery technologies have demonstrated the performance needed for long-duration storage. Other resources need to be considered. Among those resources, pumped hydro is obvious [89], but it is difficult to build new capabilities. Hydrogen storage has been widely reported [90]. Several studies indicate that hydrogen storage and conversion could be economical for storage over many days (Fig. 14d) [79]. However, the challenges have also been known for decades, including difficulty in generation, efficiency, transportation and storage. Current reports that demonstrate exactly how hydrogen storage will work for long durations remain scarce.

In addition, as discussed, it will be very difficult and very expensive to make a large investment that is not being used daily. New concepts that will enable dual purpose should be developed. It will be desirable to develop a system integrating different batteries that can be used on a daily basis for short duration storage, and when needed, can also be used to storage and deliver electricity over long durations. The dual use technology could also integrate energy conversion, chemical conversion and storage together. It can be used for energy storage when needed, and can be also used to produce other benefits for different applications when the storage is not needed.

Fig. 14c shows a conceptional design of a dual use an energy conversion and storage device, the H_2-Fe flow cell. The new flow cell enables two operating modes: as a pseudo-electrolyzer, it produces H_2 gas for industrial or energy capture applications; and as a hydrogen-iron redox flow cell, it is capable of high efficiency and low-cost grid scale energy storage. The device combines two highly reversible single electron reactions: the $H+/H_2$ half-cell from fuel cell/electrolyzers; and the Fe^{3+}/Fe^{2+} half-cell from Fe-Cr redox batteries. Capitalizing on the redox reactions between $H+/H^2$ and Fe^{3+}/Fe^{2+}, the new H_2Fe flow cell will be able to produce H_2 at a much lower voltage and improved efficiency than that of typical water electrolysis systems. The continuous production of H_2 for long-duration application can be achieved by various regeneration technologies on the Fe^{3+}/Fe^{2+} electrode. Meanwhile, the H_2Fe flow cell can also function as a redox flow battery utilizing H^+/H_2 and $Fe^{3+/2+}$ as redox couples in the anolyte and catholyte [91]. As such the H_2Fe flow cell has multi-role capability: it continuously produces and stores hydrogen for industrial gas and fueling applications, and functions as a redox flow battery when needed.

9. Conclusions

This paper analyzes data reported in the literature for both short- and long-term storage for renewable energy. The analysis suggests that a 12-h storage, totaling 5.5 TWh capacity, can meet more than 80 % of the electricity demand in the US with a proper mixture of solar and wind generation. Accelerated deployment of EVs and battery storage has the potential to meet this TWh challenge. It is critical to develop new mechanisms to manage and control the whole energy infrastructure, including the charging and discharging of EVs. It is also critical to further reduce the cost and increase the cycle life of the batteries to meet the cost target for both transportation and grid applications. Many new approaches are being investigated currently, including developing next generation high-energy and low-cost lithium metal batteries. The key scientific problems in SEI and dendrite reactions, stable electrode architectures and solid electrolyte materials have been intensely studied in the literature, but there is an urgent need to investigate these phenomena and material properties under realistic conditions so that the discoveries can be incorporated in practical high-energy cells. There is also a significant need to systematically investigate the degradation processes of important Li-ion battery systems over long cycling and develop new design rules for batteries with exceptionally long cycle lives for EVs and storage applications. Finally, meeting 100 % electricity demand with renewable energy requires new resources on the grid as well as long-duration storage. Many approaches are being evaluated or investigated for long-duration storage, but most of the battery chemistries cannot meet the cost requirement for this application. Breakthroughs in storage concepts like dual use technologies and new grid operation principles are needed. Finally, although this article only analyzed the date in the US, the principles should be applicable to other developed and developing regions.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jun Liu reports financial support was provided by US Department of Energy and University of Washington. Jie Xiao reports financial support was provided by US Department of Energy. Wei Wang reports financial support was provided by US Department of Energy. Yuyan Shao reports financial support was provided by US Department of Energy. Ping Liu reports financial support was provided by US Department of Energy. Stan Whittingham reports financial support was provided by US Department of Energy. Stan Whittingham reports financial support was provided by US Department of Energy. Jun Liu reports financial support was provided by US Department of Energy.

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