Electronic Supplementary Material (ESI) for Energy & Environmental Science.

†Electronic Supplementary Information (ESI): [Introduction, System Description, ReSOC System Cost Analysis, Electricity Arbitrage, Cost Analysis Calculation, Fig. S1-3, supplemental references S62-S68]. See DOI: 10.1039/b000000x/

Introduction

Due to the complex nature of the interplay between energy supply and demand in future non-fossil energy systems, balancing requirements will span time scales from fractions of a second to several months.[1,2,4] Short-time energy balancing can adequately be handled by number of different technologies such as super capacitors, batteries or fly wheels[4] however, energy balancing exceeding several hours leads to prohibitively high costs for those technologies. Pumped hydro, compressed air energy storage (CAES), thermal energy storage or sub-surface hydrogen storage have been proposed to overcome the challenge, but only pumped hydro seem to be economically viable for multi-hour grid-scale electricity storage[4,53] and unfortunately pumped hydro is only available in certain geographical areas. In particular, to meet the seasonal fluctuations in renewable energy supply and demand, power-to-gas seems to be the only solution with sufficient storage capacity.[62]

Unfortunately the round-trip energy efficiency (electrical-to-chemical-to-electrical) offered by conventional hydrogen-based power-to-gas technology is only 25-50%.[57,63] Subsequent catalytic conversion of the produced H_2 mixed with CO and/or CO_2 to produce CH_4 increases the volumetric energy density of the stored gas and enables usage of existing natural gas infrastructure, but unfortunately the catalytic conversion introduces energy losses which decreases the round-trip efficiency even further and thus adds to the cost of storing the energy.

The proposed electricity storage system has a round-trip efficiency exceeding 70% and uses a methane-rich gas as storage media and together with biomass the system could also provide the CO_2-neutral hydrocarbons needed for production of synthetic fuels and chemical commodities, such as benzene, formic acid, polymer pre-cursors, etc.[64]

This Electronic Supplementary Information (ESI) provides additional details to the paper "A Novel Method for Large-Scale Electricity Storage" published in Energy & Environmental Science. The first section of the ESI (System description) outlines the system components not shown in Fig 1. The ReSOC system cost analysis section provides descriptions, arguments and references for the cost items provided in Table 1. Further, a section is provided covering a calculation method for electricity arbitrage on the Danish electricity market. The net expense or future income using the proposed storage system is estimated in the final section.

System Description

The storage system (Fig. 1) is detailed in Fig. S1. The components consist of the two storage caverns, the ReSOC stack module and balance-of-plant (BOP) which includes heat-exchangers, compressors, turbines, condenser, evaporator, air blower, ejector and a recycle split.

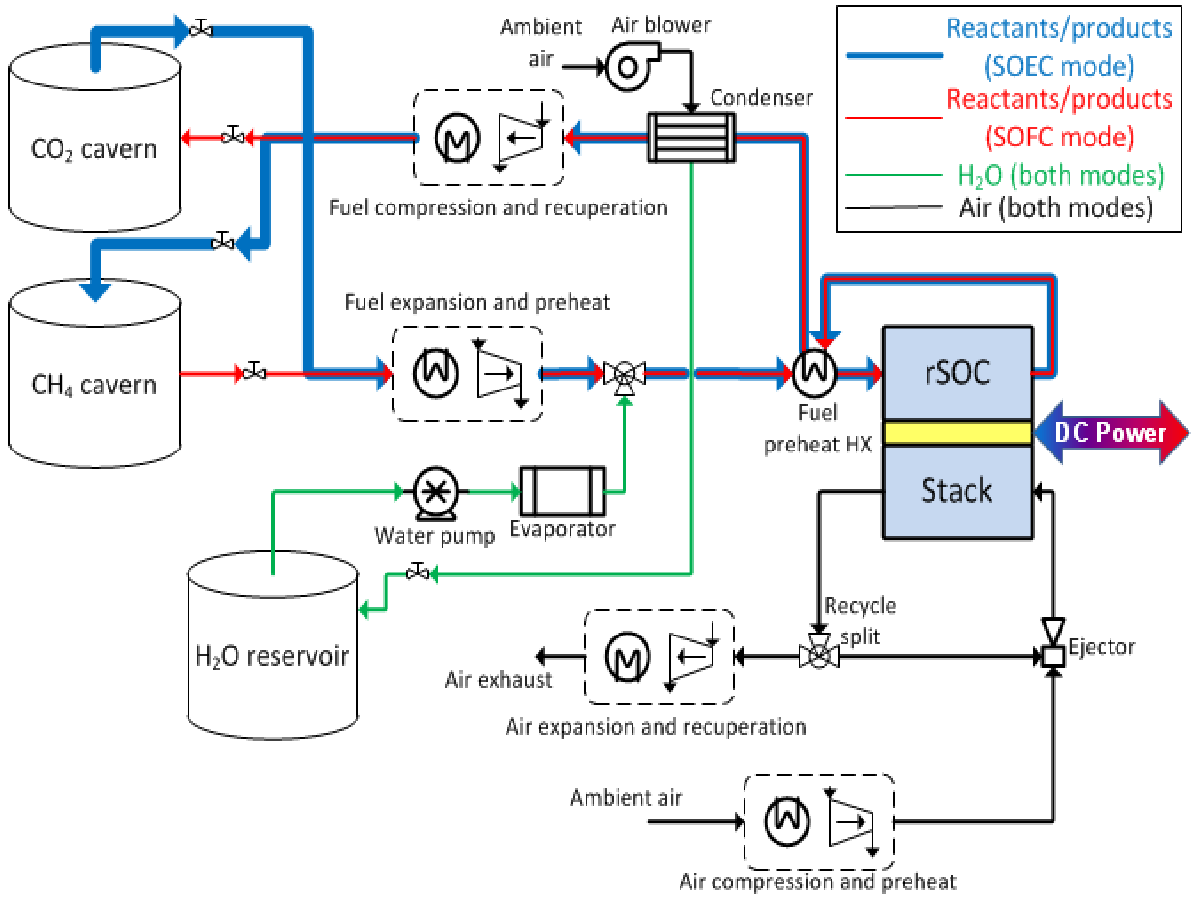

Fig. S1. Detailed ReSOC electrochemical energy storage system design schematic.

The ReSOC stack performance is estimated from a steady-state, one-dimensional, cell-level, counter-flow model adapted from prior work[14,30-32] as previously noted. Importantly, the ReSOC current-voltage characteristic is described by a simplified electrochemical model with constant area specific resistance (ASR) such that the cell voltage is determined by the relatively simple relation:

$$V_{cell} = E_N - j \cdot ASR \tag{3}$$

where j is the current density and E_N is the Nernst voltage. Current is positive in SOFC mode and negative in SOEC mode by convention. Here, we model the stack ASR as fixed (i.e., independent of temperature, pressure, and composition) to isolate the changes in system-level performance irrespective of changes in ReSOC resistance.

The BOP system components are modeled thermodynamically to determine the thermal and electrical energetic requirements under the assumptions of adiabatic, steady-state, and negligible kinetic and potential energy changes. Compressors and turbines are modelled with isentropic efficiencies of 88 and 90%, respectively, and the oxidant gas recycle ejector employs an efficiency of 20%. Heat exchangers employ minimum approach temperature differences between hot and cold streams of at least 15°C, and the temperature of the stored gas injected into the caverns is allowed to vary between 25 and 140°C. The lower storage temperatures (25°C) are achieved in SOEC mode where sub-ambient air (<25°C) from the turbine exhaust is available for cooling. The pressurized condenser has a set outlet temperature of 50°C, allowing cooling with ambient air. For additional details, the reader is referred to Ref. [32].

ReSOC System Cost Analysis

In order to estimate the ReSOC storage system expenses, a detailed analysis of the cost items in the storage system is provided. The values used for the estimation is presented in Table 1 and discussed in detail below.

Total Plant Cost (TPC) assumptions

The guidelines for economy assessment given by DOE[39] was followed in the presented analysis. If not specified, the quoted prices are 2013 prices adjusted for inflation using the US Consumer Price Index.[54] Prices are given for a system including a 250 MW ReSOC stack combined with a 500 GWh salt cavern gas storage system.

CH_4 Cavern: Today natural gas (CH_4) is stored cost-effectively on a seasonal basis in depleted gas reservoirs, aquifers and salt caverns; and CO_2 can be stored in a similar way.[11,12] The caverns in the storage facility at Ll. Torup in Denmark are located 1 km below the surface and are operated at pressures up to 200 bar. The storage volume in each of the caverns is approximately 120 million Nm^3 natural gas of which 70 million Nm^3 can be used for energy balancing. The remaining gas acts as cushion gas, i.e. as a pressure buffer. According to information from DONG Energy and KBB Underground Technologies, the capital expense (CAPEX) of the natural gas storage facility at Lille Torup is approximately 36 M$.[12] At 160 bar and a stack pressure of 20 bar, the volumetric energy density factor (i.e. the volumetric energy density of the gas divided by the volumetric energy density of CH_4) is 0.72 which means the energy density of the stored gas is 4.1 GJ/m^3. This gives an energy balancing capacity of 500 GWh and an energy specific cost of 36 M$ / 500 GWh = 7.2 ¢/kWh. The energy specific cost can also be calculated as 36 M$ / 250 MW = 144 $/kW since the size of the ReSOC stack power is 250 MW.

CO_2 Cavern CAPEX: The overall chemical reaction in the proposed energy storage system is $2H_2O + CO_2 \leftrightarrow CH_4 + O_2$. For this reason the required molar storage of CO_2 equals that of CH_4. At near-ambient temperatures and 150-200 bar, one Nm^3 of CO_2 takes up app. 2.4 times less volume than 1 Nm^3 of CH_4, due to the difference in fugacity between CH_4 and CO_2. However, CO_2 storage in salt caverns is a relatively new and unproven technology, so with 40% process contingency and 50% project contingency,[39] the CO_2 cavern cost is estimated at 32 M$, resulting in an energy specific cost of 6.4 ¢/kWh or 128 $/kW.

H_2O reservoir: A 75,000 m^3 water pit for thermal energy storage (wTES) was constructed in 2011-2012 in Marstal, Denmark at a cost of 50.4 $/m^3 excluding transmission pipes.[16] To produce 70 million Nm^3 of CH_4, 115,000 m^3 of water is required resulting in a H_2O reservoir cost of 5.8 M$, i.e. and energy specific cost of 1.2 ¢/kWh, or 23 $/kW, neglecting that the specific project cost in $/m^3 decreases with storage size and that improved lids for the wTES technology are expected to be developed during the next few years.[16]

ReSOC System. The ReSOC technology is not fully commercialized which means the system cost is still relatively uncertain. In particular, the estimates are highly dependent on the anticipated production volume. Thijssen estimated in 2007, at an annual production of 250 MW/yr, a 3.1 MW stationary hybrid SOFC system with a 2 MW SOFC stack to cost 419 $/kW – 499 $/kW, depending of stack and cell type.[40] Adjusting for inflation, this corresponds to an energy specific cost in 2013 of 477 - 568 $/kW where the 2 MW Stack constitutes 139-203 $/kW. A techno-economic analysis of a 250 MW integrated gasification fuel cell (IGFC) plant was conducted by DOE in 2009.[43] The cost of the 19 bar, 221 MW SOFC converter excluding syngas expander, air blowers and HX was 108 M$. Scaled linearly to 250 MW and adjusted to 2013 prices, this is equal to 122 M$, equivalent to 489 $/kW. At an annual production volume of 500 MW/year, Warren *et al.* estimated the cost of the SOFC stack in a 362 kW combined heat and power stationary fuel cell system to cost 165 $/kW.[42] Adjusted to 2013 prices this is equivalent to 168 $/kW.

The ReSOC is estimated to cost the same as the SOFC converter per kW electrical output, because todays SOFCs can be operated as ReSOCs.[38,59] In this study we choose a cost of 200 $/kW for the ReSOC stack equivalent to 50 M$. Balance of stack and stack assembly is estimated to 2.2 $/kW and 5.7 $/kW, respectively.[40]

Air compression/expansion: Thijssen estimated the turbine system with the for a 3.1 MW SOFC/turbine system to cost 148 $/kW which equals 168 $/kW in 2013.[40] Scaled linearly, the air compression/expansion system for the 250 MW ReSOC stack is estimated to cost 42 M$.

CH_4/CO_2 compression/expansion: The molar flow rate of CO_2 is 31% of the molar air flow rate when the ReSOC is operated in fuel cell mode and 51% when the ReSOC is operated in electrolysis mode. For this reason the cost of the CH_4/CO_2 compression/expansion system is estimated to be 51% of the air compression/expansion system cost, equivalent to 21 M$ or 86 $/kW.

Recuperators: Heat recuperators were estimated to 32 $/kW.[40] Adjusted to 2013 prices this is equivalent to 36 $/kW or 9.1 M$.

Feedwater and miscellaneous balance of plant systems: A 809 ton/hr feed water system including a demineralized water storage tank, condensate pumps, deaerator, intermediate and high pressure feedwater pumps, boiler (28 bar), service air compressors, instrument air dryers, closed cycle cooling heat exchangers, closed cycle cooling water pumps, raw water pumps, filtered water pumps, makeup water demineralizer, water makeup & pretreating was analyzed by DOE in 2007.[41] The cost for the entire system including equipment cost, materials cost and labor cost was 31 M$. The required feed water for the 250 MW ReSOC system is 46 ton/hr. Using a scaling exponent of 0.7 [65] the cost is 4.2 M$, equivalent to 17 $/kW.

Evaporator: A 694 ton/hr heat recovery steam generator system was estimated to have a cost of 51.5 M$ in the 2007 DOE report.[41] A 46 ton/hr steam generator system is required for the 250 MW ReSOC system when it operates in electrolysis mode. Using a scaling exponent of 0.7 this leads to a cost of 7.7 M$, equivalent to 31 $/kW.

Condensator: DOE also estimated the cost of a 298 MW steam turbine system with a 810 ton/hr condenser.[41] The cost including equipment cost, materials cost and labor cost for the condenser unit & auxiliaries was 7.0 M$. Steam piping cost was estimated to be 10.6 M$. The condenser unit for a 250 MW ReSOC system operating in fuel cell mode requires a 122 ton/hr condenser unit which gives a cost of 4.7 M$, equivalent to 19 $/kW.

Accessory Electric Plant: Since the proposed system is used for electricity storage an accessory electric plant is required to interface between the electric grid and the ReSOC system. The accessory electric plant cost for a 250 MW IGFC plant was estimated to be 31 M$,[43] equivalent to 123 $/kW. Thijssen estimated power conditioning to 57 $/kW in 2013 prices for the 3.1 MW SOFC system, equivalent to 14.2 M$.[40] Here we use 80 $/kW equivalent to 20 M$.

Instrumentation and control: The instrumentation and control cost is for the 250 MW IGFC plant[43] was estimated to be 10 M$, equivalent to 41 $/kW. Thijssen estimated instrumentation and control costs to 34.1 $/kW for the 3.1 MW SOFC system.[40] In this study we have chosen 37 $/kW equivalent to 9.25 M$ for the 250 MW ReSOC system.

Piping and Valves: Piping and valves are estimated to cost 34 $/kW equivalent to 8.5 M$.[40]

Improvement to site: The 250 MW IGFC plant analysis[43] estimated the cost of improvement to site to 8.0 M$, equivalent to 32 $/kW.

Building and structures: Based on the IGFC plant analysis,[43] building and structures cost is estimated to be 8.0 M$, equivalent to 32 $/kW.

Fixed Operating and Maintenance cost

Labor expenses: 16 jobs are required to operate a double train 555 MW IGCC plant (case 2).[41] Assuming the required operating jobs scales with plant size, the 250 MW storage system requires 8 operating jobs. Including operating labor burden and labor O-H charge rate the annual labor expense is 0.43 M$ per job[41,43] which gives a total labor expense of 3.4 M$ per year.

Variable Operating and Maintenance cost

Maintenance material, water, chemicals: As with the 250 MW IGFC plant, expenses for makeup water, maintenance materials and chemicals are estimated to be 0.71 ¢/kWh.[43] To obtain the annual expenses, 0.71 ¢/kWh is multiplied with the annual number of hours where electricity is bought and sold times the nominal power of 250 MW. Based on the analysis of arbitrage on the Danish electricity spot market DK-West presented in the next section, the number of selling and buying hours in 2008 required to reach an optimal arbitrage was 5370 hours which gives a total expense of 9.5 M$.

Miscellaneous estimates

ReSOC Lifetime: SOEC operation at high currents is known to accelerate cell degradation.[36,44] Christiansen *et al.*[45] defined the 3 kW SOFC APU lifetime requirement as 12000 hours at current > 0, and 36000 hours at temperature > 100 ˚C.[45] An observed parabolic oxidation behavior and a parabolic rate constant indicated a lifetime around 50,000 h at 650 ºC, i.e. indicating that the requirements were fulfilled.[45] Current SOEC degradation rates are usually higher than SOFC degradation rates.[44,46-49] However, the SOEC R&D is small compared to the SOFC R&D so it is expected that SOEC degradation rates can be reduced to the same level as the SOFC degradation rates.[50,51] SOEC lifetime has been previously estimated as 5 years[52] and reversible cell operation is known to decrease degradation rates.[38,59] For these reasons, a ReSOC lifetime of 5 years is used in this analysis.

System Lifetime: The lifetime of the remaining parts of the ReSOC system is estimated to be 20 years in agreement with other baseline assumptions.[12,39,43]

Interest rate: The interest rate is estimated to be 5%. The annual expenses are calculated as an annuity loan with a 20 year system lifetime for all components except the ReSOC, balance of stack and stack assembly. The ReSOC system annual expense is calculated with an annuity loan with a lifetime of 5 years.

Storage capacity: One of the interesting features of the proposed ReSOC storage system is that it is able to store 500 GWh (see CH_4 Cavern cost estimation for details). Assuming a 250 MW ReSOC system, this energy capacity gives a storage time of 2000 hours or close to 3 months of energy storage.

Volumetric energy density factor: The volumetric energy factor is the ratio between the volumetric energy content in the stored gas composition divided by the energy content in CH_4. When the gas exits the ReSOC during electrolysis mode at 20 bar, it is mainly CH_4 with H_2 and H_2O. After the H_2O is condensed out upon cooling, this latter gas is rich in CH_4 (58%) with substantial H_2 (40%). This mixture has a volumetric energy density factor of 0.72.

Electricity Arbitrage

In order to analyze the economic viability of the proposed electricity storage system the potential income from electricity arbitrage is compared to the estimated annual expenses for the proposed storage system. The western Danish electricity spot market is chosen for the study. Denmark is a convenient choice for the analysis since spot-market prices are available hour-by-hour for the Danish electricity market.[66] Further, large subsurface caverns for natural gas storage with known cost estimates already exist in Denmark[12] and in addition, a forecast of electricity prices in 2050 is available.[67]

Revenue by electricity arbitrage in Denmark

There are three ways for an electricity storage facility to profit in the Danish power system. First, the system can profit from arbitrage (buying and selling power) on the spot market. By far, the dominant part of power production is managed by the market mechanisms on the power spot market and this is also where electricity storage can directly facilitate e.g. higher levels of wind power penetration by shifting load and power production. Secondly, storage systems can profit on the regulating market which maintains energy supply and demand balance in real time. Demand and power supply forecasts are not perfect and the imbalances are handled by the regulating market. Finally, an energy storage system can sell ancillary services to the transmission system operator (TSO), which in the Danish case is Energinet.dk. Here, high prices are paid for primary reserves that can ensure frequency and voltage stability and provide black start. Since the proposed system is aimed for grid-scale energy storage, we only consider arbitrage on the spot market.

A participator on the spot market makes bids on power production and consumption for each hour of the next day (the bids must be submitted the previous day, i.e. 12 to 36 hours before the delivery of the power). The price (and total production) for each hour is then determined by the intersection of the demand and supply curves. When the price on the spot market fluctuates sufficiently an income can be made by buying and selling power.

The annual DK-West spot market electricity prices from 2006 to 2013, sorted hour-by-hour with lowest prices first are shown in Fig. S2A. To estimate the arbitrage given the annual price fluctuations, a simple strategy for buying and selling power is applied. The strategy includes following conditions:

1) The price for buying must be less than the price for selling times the round trip efficiency, ensuring positive income. i.e. that

$$P_{buy} < \eta \cdot P_{sell} \tag{4}$$

where η is the electricity to electricity efficiency, P_{buy} and P_{sell} are the electricity prices when the electricity is bought and sold, respectively.

2) The annual amount of power bought multiplied by the round trip efficiency must equal the annual amount of power sold, ensuring balance of stored energy, i.e. that

$$N_{sell} = \eta \cdot N_{buy} \tag{5}$$

where N_{sell} and N_{buy} is the number of hours where electricity is sold and bought, respectively.

The expressions (4) and (5) define the simple buy-and-sell strategy that yields the maximum annual income. Note that the strategy requires a priori knowledge about the power prices. This is of course unrealistic for a whole year which is why it should be emphasized that the following expression (6) is the *maximum* annual income:

$$\text{annual income} = \int_{P_{sell}}^{\infty} n \cdot p \cdot dp - \int_{0}^{P_{buy}} n \cdot p \cdot dp \tag{6}$$

where n is the number of hours having an electricity price between p and $p + dp$. Using the prices given in Fig. S2A, the dimension of the annual income will be [$/MW], that is the income scales with the power of the energy storage system. The required storage is the difference between the minimum and the maximum of energy in MWh in the storage system divided by the power of the storage system, i.e. the dimension of the required storage is given in hours (or MWh/MW). Similarly the annual number of hours electricity is sold can be found. By dividing the annual income with the number of hours electricity is sold, the annual income can be presented in $/MWh or - equivalently - ¢/kWh.

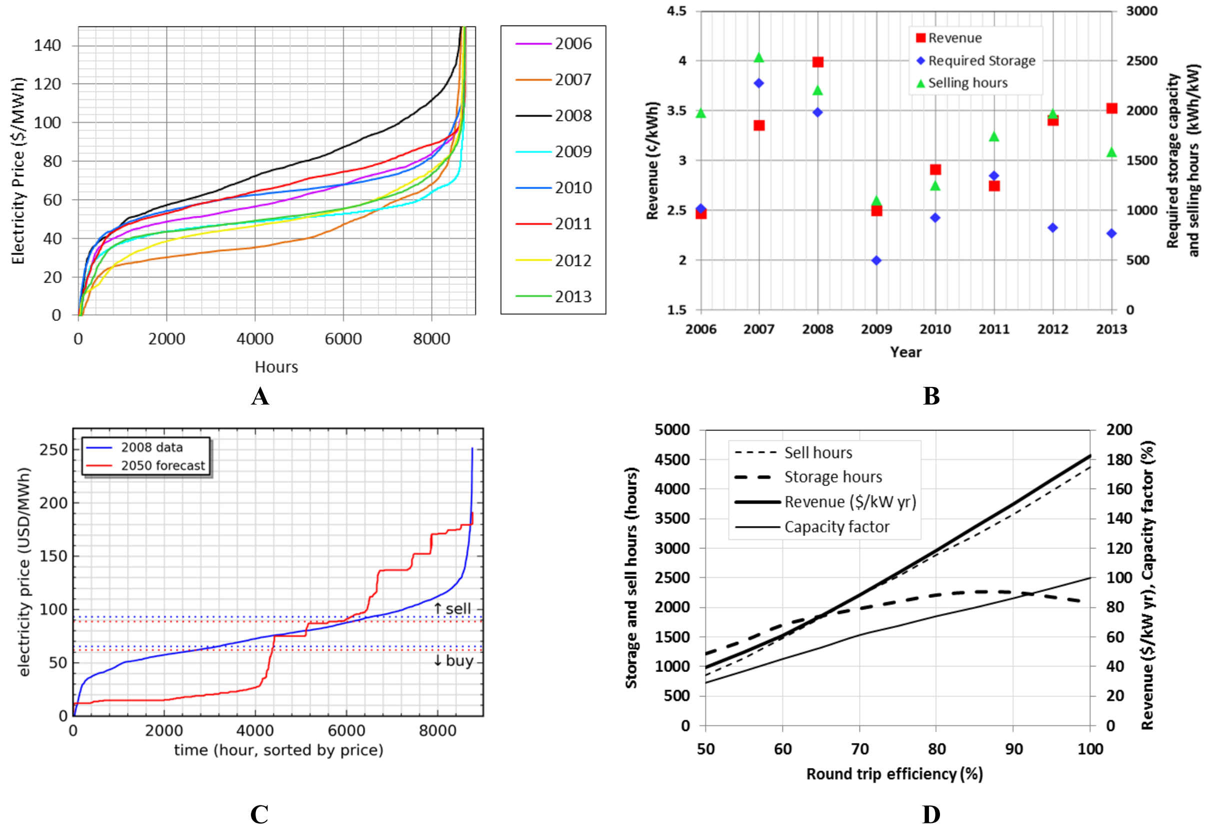

Fig. S2. Danish electricity spot market prices, the implemented arbitrage strategy and resulting profit. (**A**) DK-West spot market electricity prices from 2006 to 2013 sorted hour-by-hour, lowest prices first.[66] (**B**) Maximum revenue which could have been obtained on electricity arbitrage on the Danish electricity spot market during 2006-2013. Also shown is the required storage to obtain the maximum revenue and the number of hours where electricity is sold. Note how the revenue decreased after the 2008 financial crisis and how it subsequently gradually increases. (**C**) Illustration of the buying-selling strategy. The spot market electricity price for western Denmark in 2008 and forecasted in 2050.[67] The dotted lines indicate the price to sell and the price to buy, which gives the highest profit (requiring that the storage is in balance at the end of the year). (**D**) Storage and sell hours, revenue and capacity factor as a function of round trip efficiency based on the 2008 electricity prices.

Using η=70% as the roundtrip efficiency, Fig. S2C provides an illustration of the buying-selling strategy for the spot market electricity price for western Denmark in 2008 and forecasted in 2050.[67] Above the upper dotted lines electricity should be sold, while below the lower dotted line electricity should be bought.

Still with η=70%, the maximum annual income (revenue) for 2006 – 2013 on the western Danish electricity spot market, the required storage hours, and selling hours are presented in Fig. S2B. The maximum annual revenue during 2006 – 2013 occurs in 2008 where it is 88 $/kW with a required storage of 1980 hours. In order to achieve the maximum arbitrage, the ReSOC system should sell electricity for the 2211 hours having the highest electricity prices and buy electricity for the 3159 hours with the lowest electricity prices, which means the ReSOC system capacity factor would be (3159 h + 2211 h)/8760 h = 61% and that the 250 MW ReSOC system would have an income of 22 M$ - equivalent to 4.0 ¢/kWh of electricity sold back to the grid.

Additional arbitrage on the regulating market and income from ancillary services could increase the annual income further but is not considered in this analysis.

Cost Analysis Calculation

A capacity factor of 61% is used in the cost calculation, as suggested by the arbitrage analysis for year 2008 in the above section. The annual expense is calculated as an annuity loan with a 20 year system lifetime for all components except the ReSOC, balance of stack and stack assembly which is calculated using an annuity loan with a lifetime of 5 years. The annuity loans were calculated with an interest rate of 5%.

The total annual expense for the 250 MW ReSOC storage system including fixed and variable O&M (Table 1) is estimated to be 42 M$ in 2008. Using the number of selling hours in 2008 given in the arbitrage section, this is equivalent to 7.7 ¢/kWh sold electricity or an annual expense of 169 $/kW. A detailed annual expense distribution is presented in Fig. 6A.

The maximum annual revenue heavily depends on the round-trip efficiency. The maximum annual revenue for 2008 as a function of roundtrip efficiency is shown in Fig. S2D. The revenue increases from 39 $/kW yr at 50% roundtrip efficiency to 150 $/kW yr at 90%. Further, the required storage hours varies from 1218 hours at 50% round trip efficiency to a maximum of 2263 hours at 85% round trip efficiency. The capacity factor increases from 29% at 50% round trip efficiency to 100% at 100% round trip efficiency. Finally, the number of sell hours increases from 850 hours at 50% round trip efficiency to 4379 hours at 100% round trip efficiency.

An earlier analysis of the western Danish electricity spot market data for 2000 – 2008[53] found that with a round-trip efficiency of 70%, around 70% of the income can be obtained if the analyzed time interval is 24 hours instead of one year (i.e. that the energy balance requirement * MERGEFORMAT (4) must be met every 24 hours instead of every year) and that this would reduce the storage requirement to 3-4% of the storage required for annual storage. This indicates that the storage media needs to be extremely cheap in order to make additional profit when storing electricity beyond 24 hours. Although it is challenging to make profitable, electricity storage on a seasonal basis, it is highly required for future non-fossil based energy infrastructures.[3] Fig. S3 indicates the feasibility for seasonal storage for various electricity storage technologies. The Figure displays the potential storage size and maximum discharge hours of the proposed ReSOC system (CH_4) along with other storage technologies.

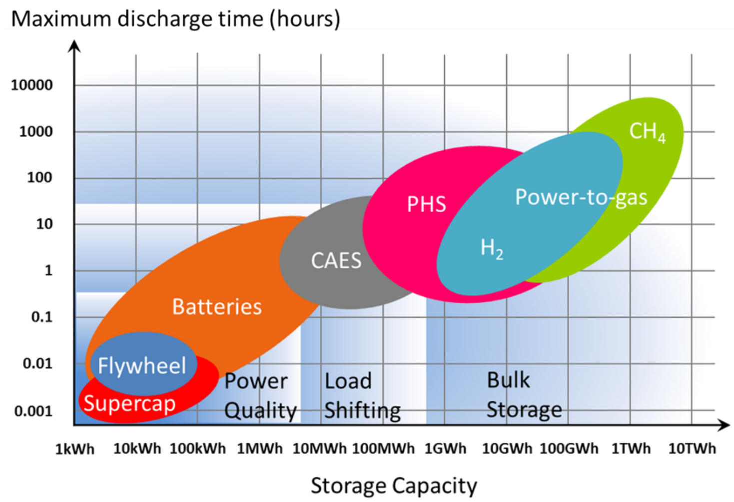

Fig. S3. Grid requirements and energy storage technologies amended from literature[1,4,68] as function of size and maximum storage hours.

Electricity arbitrage in the future

Although the proposed ReSOC storage system does not seem profitable today, it may become profitable in the future. It is the ambition of the Danish Government to reach 100% renewable energy production in 2050 and 100% renewable electricity and heat production in 2035.[17] This will lead to larger electricity price fluctuations which will increase the electricity arbitrage revenue. As mentioned, the electricity prices at the DK West market in 2008 along with a 2050 forecast[67] is presented in Fig. S2C.

The figure indicates that significantly larger price fluctuations will be generated in 2050 relative to what was present in 2008. From the forecast, the maximum possible arbitrage in 2050 is 369 $/kW yr which corresponds to 92 M$ with the 250 MW system – equivalent to 17 ¢/kWh assuming the same number of selling hours and expenses as in 2008. This means the system would have a net *income* of 17 ¢/kWh – 7.7 ¢/kWh = 9.3 ¢/kWh.

It is important to note that if sufficiently large storage systems enter the electricity market, this will decrease price fluctuations. The current annual electricity production in DK is slightly less than 30 TWh. Based on the 2008 electricity prices (Fig. S2A) the 250 MW ReSOC system would store 0.6 TWh or about ~2 % of the production and therefore have limited impact on the electricity price. Assuming the Danish targets of 100% renewable electricity production in 2035 is met and that the storage requirement is 10-20% of the electricity production[3] it would require five to ten 250 MW ReSOC systems to fully cover the future Danish storage need. In this case the storage systems would have some impact on the electricity price fluctuations but it is beyond the scope of this study to quantify this impact.

There are, of course, a number of uncertainties in the presented cost assumptions and economy calculations, but it seems realistic that electricity arbitrage by use of the proposed ReSOC storage system could become profitable or at least provide a seamless transition towards fossil-free energy systems well before 2050.

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Additional Author notes: S.H.J. described the operating concept. S.B and G.H. did the thermodynamic calculations. Z.G provided the cell test data. C.H.W. and R.J.B. conducted the system modelling. S.H.J and C.G made the economic estimation. M.M contributed to the overall scientific consistency and interpretation. Figures and text were edited and improved by all authors.