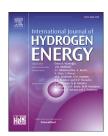


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Hybrid Hydrogen Home Storage for Decentralized Energy Autonomy



Kevin Knosala ^{a,*}, Leander Kotzur ^a, Fritz T.C. Röben ^a, Peter Stenzel ^a, Ludger Blum ^b, Martin Robinius ^a, Detlef Stolten ^{a,c}

- ^a Institute of Energy and Climate Research, Techno-economic Systems Analysis (IEK-3), Forschungszentrum Jülich GmbH, Wilhelm-Johnen-Str., D-52428, Germany
- ^b Institute of Energy and Climate Research, Electrochemical Process Engineering (IEK-14), Forschungszentrum Jülich GmbH, Wilhelm-Johnen-Str., D-52428, Germany
- ^c Chair for Fuel Cells, RWTH Aachen University, c/o Institute of Energy and Climate Research, Techno-economic Systems Analysis (IEK-3), Forschungszentrum Jülich GmbH, Wilhelm-Johnen-Str., D-52428, Germany

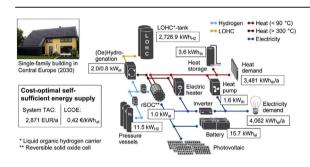
HIGHLIGHTS

- Hybrid hydrogen storage enables energy self-sufficient residential buildings.
- Different technology supply and storage configurations are comparatively assessed.
- RSOC and LOHC show high potential in self-sufficient building energy systems.
- Heat integration between rSOC and LOHC systems reduces hydrogen storage needs.
- Levelized cost of electricity for self-sufficient supply can reach 0.42 €/kWh by 2030.

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GRAPHICAL ABSTRACT



ABSTRACT

As the share of distributed renewable power generation increases, high electricity prices and low feed-in tariff rates encourage the generation of electricity for personal use. In the building sector, this has led to growing interest in energy self-sufficient buildings that feature battery and hydrogen storage capacities. In this study, we compare potential technology pathways for residential energy storage in terms of their economic performance by means of a temporal optimization model of the fully self-sufficient energy system of a single-family building, taking into account its residential occupancy patterns and thermal equipment. We show for the first time how heat integration with reversible solid

E-mail addresses: k.knosala@fz-juelich.de (K. Knosala), l.kotzur@fz-juelich.de (L. Kotzur), f.roeben@fz-juelich.de (F.T.C. Röben), p. stenzel@fz-juelich.de (P. Stenzel), l.blum@fz-juelich.de (L. Blum), m.robinius@fz-juelich.de (M. Robinius), d.stolten@fz-juelich.de (D. Stolten).

^{*} Corresponding author.

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oxide cells (rSOCs) and liquid organic hydrogen carriers (LOHCs) in high-efficiency, single-family buildings could, by 2030, enable the self-sufficient supply of electricity and heat at a yearly premium of 52% against electricity supplied by the grid. Compared to lithium-ion battery systems, the total annualized cost of a self-sufficient energy supply can be reduced by 80% through the thermal integration of LOHC reactors and rSOC systems.

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Abbreviations

AC alternating current
CAPEX capital expenditures
COP coefficient of performance

DBT dibenzyltoluene
DC direct current
HT high temperature

LOHC liquid organic hydrogen carriers

LIB lithium-ion battery LT low temperature

OPEX operational expenditures
PEM proton-exchange membrane

RFB redox-flow battery rSOC reversible solid oxide cell

SO solid oxide

TAC total annualized cost

Introduction

The European Union recently updated its Clean Energy for all Europeans package to facilitate the transition towards clean energy and the reduction of greenhouse gas emissions [1]. Its Renewable Energy Directive II increases the target for renewable energy sources consumption to 32% by 2030 and concepts for net-zero, energy-positive, and fully renewable districts characterized by low resource consumption and the high integration of local renewable energy sources are avidly promoted [2]. Around 40% of global energy consumption is accounted for by the building sector for the purposes of heating, cooling, ventilation, hot water, and lighting [3]. In the coming years, charging stations for battery-electric vehicles placed on buildings will significantly add to building energy demand. Therefore, measures for sustainable development and the reduction of greenhouse gases are highly relevant to the sector. It has been acknowledged that increased self-consumption is beneficial in the context of large shares of distributed renewable generation and falling feed-in tariffs [4], and therefore public and academic interest in – at least partly – "leaving the grid" or "living offgrid" is growing [5]. The levelized costs of self-generation have already dropped below the electricity retail price for end consumers in some parts of the world [6]. In 2020, the globallyinstalled photovoltaic capacity exceeded 700 GW, and additions are expected to accelerate through the early 2020s [7]. The costs of electricity from utility-scale photovoltaic (PV) will drop below 0.02 \$/kWh for projects in Australia, China, Chile, and the United Arab Emirates in this time [8]. In the long term, it can be assumed that increased self-consumption will lead to higher distribution costs, as grid costs are distributed over the lower amounts of energy provided [9,10], resulting in changing energy tariff structures [11]. Especially for new buildings, high shares of self-consumption will become economically interesting, as investments in building energy supply systems are less prone to future price changes for energy supply and will therefore be more predictable in terms of future costs.

Collective prosumer¹ business models and regulations for self-generation and self-consumption are on the rise and challenge current energy market structures and institutions [12]. Aside from economic motivations, environmental awareness and increased political participation are further drivers for households and municipalities towards increased energy self-sufficiency [13–15]. Citizens may wish to play an active role in supplying their own energy via self-governance and community ownership to be less dependent on utilities, centralized markets, and other structures [12] and be willing to pay extra for increased resilience [16]. Furthermore, small-scale residential solutions could lead to an energy transition that is characterized by heterogeneous interests, social acceptance issues, and bureaucratic barriers on a large scale.

Battery-electric systems are widely installed in residential buildings in order to increase self-consumption from rooftop PV [4]. Advancements in battery technology seek to lower capacity costs and reduce environmental impacts [17]. The development of flow batteries is a promising solution for reducing cost per storage by separating power and storage units (electrolyte), which makes them especially interesting for longer storage durations [18]. However, it has been shown that relying on battery storage alone for balancing PV and wind generation across seasons results in significant overcapacities alongside efforts to overcome periods of little or no renewable generation [19]. In particular, regions subject to abrupt changes in wind and solar availability during autumn and winter are prone to this problem when striving for a system based solely on renewable generation [19,20]. The necessary overcapacities, considering energy supply by

¹ A prosumer in this context is defined as an actor who produces and consumes renewable power.

rooftop PV alone, are cost-intensive, technically-limited (e.g., due to a lack of available roof area for photovoltaic installations) and unsustainable [5,21]. In contrast, chemical energy storage exhibits lower storage capacity costs for long-term seasonal storage and high storage density [22]. Colbertaldo et al. showed in their analysis on hydrogen energy storage for a fully renewable Californian electric power system that a power-to-power hydrogen storage system results in substantially lower system costs as battery storage [23].

A life cycle analysis of a self-sustained trigeneration energy system based on renewable hydrogen for an office building in Greece showed substantial reduction in environmental load compared to the exclusive use of the Greek energy mix [24]. Sorgulu and Dincer showed in their study how a hydrogen energy system based on concentrated solar power and wind energy can efficiently supply electricity, hot water and cooling demands of a group of residential buildings [25]. A techno-economic analysis of an off-grid residential community in a desert region demonstrates the economic viability of a system powered by solar energy and equipped with hydrogen storage for a renewable fraction of 40% in 2020 [26]. Lokar and Virtič showed, in a pilot residential building located in Slovenia with a limited PV generation capacity of 6.72 kW_p and annual energy demand of 9015 kWh, that the operation of a fuel cell for energy supply requires 144 kg of additional hydrogen per year. They concluded that a fully self-sufficient supply would be possible with a larger PV generation capacity [27]. In a thermoeconomic analysis for a residential complex in Tabriz, Iran, a hybrid hydrogen system for 8 days of energy autonomy with PV thermal, proton-exchange membrane (PEM) fuel cells, hydrogen pressure storage and a battery resulted in an LCOE of 0.286 \$/kWh [28]. Gstohl and Pfenninger showed that energy self-sufficient households with PV and electric vehicle would be feasible in the temperate climate predicted for 2050 [29]. They identify PV efficiency and available module area as being the most critical parameters for the feasibility of such systems and conclude that self-sufficiency may become cost-competitive, depending on storage and fossil fuel prices. In a technoeconomic analysis, Kleinebrahm et al. demonstrated important cost reductions for self-sufficient building energy systems with an electric vehicle with the use of highpressure hydrogen storage [30].

A self-sufficient energy supply with hydrogen storage has already been realized for single- and multi-family dwellings [31,32], as well as for residential districts [33], and there are commercial suppliers that offer all-in-one hydrogen solutions for residential storage.² These implementations show that a viable degree of autarky³ for energy self-sufficient buildings strongly depends on such buildings' heating requirements. The lower the building's energy demand, the easier a self-sufficient supply can be realized. In order to reduce heat demand to a minimum, good insulation is a key requirement. Furthermore, relatively cheap

latent or sensible heat storage can serve as a source of flexibility for the entire system in an efficiently-coupled configuration [34]. Therefore, cogeneration from fuel cells is seen as a promising solution for off-grid residential energy supply [35].

Many studies have analyzed photovoltaic-battery and hydrogen storage systems for residential buildings, focusing only on electricity self-sufficiency [13]. Herein, we present a building-level techno-economic study of an energy self-sufficient single-family house for the year 2030. We show for the first time how a heat-integrated hydrogen storage unit equipped with a liquid organic hydrogen carrier (LOHC) storage system and reversible solid oxide cells (rSOCs) enables cost-effective, self-sufficient residential buildings with only rooftop PV installed. In contrast to previous studies of residential hydrogen storage, energy systems with LOHC storage achieve cost-efficient supply with comparatively low PV generation capacity. They are therefore highly suitable for renewable energy generation applications in residential buildings with significant space restrictions.

Hydrogen applications for residential buildings

Fuel cells and electrolysis

Fuel cell-combined heat and power systems for residential buildings based on PEM fuel cells and solid oxide (SO) fuel cells are state-of-the-art and commercially available from different suppliers. They process natural gas (from the gas grid) into hydrogen in fuel processor units based on steam or auto-thermal reforming, operating at temperatures above 800 °C [36]. Electricity self-sufficiency can be achieved when a battery is integrated. However, such systems usually aim to maximize the running time and are therefore connected to the electricity grid in order to feed-in excess generated power. For a fully self-sufficient supply, an electrolysis unit is needed; a reforming unit is no longer necessary. Without a grid connection, operation is determined by seasonal fluctuations of energy supply and demand, and so high operational flexibility is required from fuel cells and electrolysis.

PEM cells are based on polymer materials and operate at temperatures of 60-160 °C [36] and exhibit electrical system efficiencies of 50% [37]. Mostly driven by applications in the automotive sector, PEM fuel cells are projected to reach electrical peak efficiencies of up to 65% by 2030 [37]. SO cells, meanwhile, are based on ceramic materials and operate at high temperatures (600-850 °C). Compared to PEM systems, SO cells show lower activation losses at lower current densities, resulting in higher theoretical efficiencies [38]. Reviews have shown that the overall efficiencies for residential SO fuel cell systems in the range of 1-10 kW can reach 90% [39] showing electrical efficiencies of 45-60% (against lower heating values of hydrogen) [40]. SO electrolysis cells can achieve electrical efficiencies of 70-76%, with partial heat recovery for water evaporation and heating of the stack [41]; however, their technology readiness level is still at the laboratory scale [38].

² See, e.g., https://www.homepowersolutions.de/en, https://lavo.com.au/.

 $^{^{\}rm 3}$ The terms energy autarky and self-sufficiency are used as synonyms here.

In recent years, cell technologies that combine fuel cells and electrolysis into single units have been developed for both PEM and SO cells [38,42]. Such systems potentially prolong component lifetimes by reducing thermal stress, raising the operation rate and reducing installation space, as well as the combined device cost [38]. Especially for the ceramic membranes of SO cells, which are highly sensitive to thermo-mechanical tensions [43], it is beneficial to reduce thermic cycles by switching between electrolysis and fuel cell modes at the operating temperature. The long-term operation of a 5 kW-class rSOC at our facilities has demonstrated good functionality [44], but questions regarding long-term stability remain and are subject to ongoing research activities. To take into account the minimum load constraints on the components in our model, we imposed a restriction on rSOC operation of over 20% at all times. The techno-economic parameters for the technology must be derived on the basis of conventional SO fuel cell systems due to missing commercial experience with reversible fuel cells [38]. The aforementioned combination of conventionally two separate units (for fuel cells and electrolysis) into one allows an optimistic price prognosis for the year 2030 if commercialization is achieved.

Hydrogen storage

Hydrogen storage with pressure vessels⁴ and metal hydride storage systems⁵ are already commercially-available for residential applications. Recent technological developments have aimed to improve the efficiency, handling, and space requirements of hydrogen storage units [45]. Promising solid-state storage technologies use hydrogen-absorbing compounds such as metal hydrides that allow for the compression and storage of hydrogen without the use of moving components [45,46]. Another storage technology uses LOHCs as a carrier fluid that can be enriched with hydrogen in a catalytic, exothermic, hydrogenation reaction [47]. Although both technologies allow for the efficient integration of residential heating and cooling with the storage process, this study focuses on LOHCs as an alternative storage solution.

Most applications of LOHC storage solutions utilize dibenzyltoluene (DBT) as a non-flammable carrier fluid. DBT can be enriched in the hydrogenation process to a capacity of 6.4 wt% hydrogen (1.75 kWh_{H2}/kg_{DBT}) [47] and easily recovered after dehydrogenation [48]. However, the dehydrogenation of DBT can be challenging because of the strong bonds between the carbon and the hydrogen atoms. Advanced catalysts are researched for a more efficient dehydrogenation [45,49,50,51]. Studies by Jorschick et al. have demonstrated the operation of a single LOHC reactor with a 0.3 mass% Pt on alumina catalyst for both LOHC hydrogenation (exothermic at 301 °C, 3 MPa) and dehydrogenation (endothermic at 291 °C, 0.1 MPa), offering high dynamics when switching between the modes [52]. Peters et al. demonstrated for a combined system of LOHC

dehydrogenation and an SOFC that the maximum efficiency of LOHC-bound hydrogen-to-electricity is 45% at full load, avoiding any critical conditions for the system components [51]. Further, roundtrip efficiencies (electricity-to-electricity) for LOHC storage systems of 30–40% without heat recovery have been reported [47]. Up to 70% of electrical energy is converted into heat in this process. Residential buildings offer the opportunity to utilize this large share of energy for room heating or adsorption cooling. Further, the combination of rSOCs with LOHCs shows promising operational advantages through the possibilities of heat integration [51]. The integration of fuel cell heat into the LOHC reactor can contribute to the solution of the dehydrogenation challenge.

Fig. 1 shows the energy flows for an rSOC combined with LOHC storage. Heat from the exothermic hydrogenation of the carrier fluid can be extracted and used to preheat and evaporate the electrolysis feed water or transferred to a residential heating system. Heat for the endothermic dehydrogenation of the carrier fluid can be supplied by excess heat from the rSOC that operates in fuel cell mode or by an electric heater. We assume that the rSOC in fuel cell mode runs at an electrical efficiency of 45% and heat efficiency of 35% to decouple enough heat for the dehydrogenation process. The heat integration increases the roundtrip electrical efficiency to around 40%, discounting heat usage in the residential heating subsystem [47].

Integration into building energy systems

The efficient integration of hydrogen components into residential energy systems requires a management system and safety controlling [53]. Multi-objective energy management systems based on fuzzy logic are developed for optimal operation of such systems [53]. Advanced controlling and monitoring aims at reducing peak demands, minimizing component degradation, and improving overall system performance with demand response [54]. Further, the installation in residential communities needs specific safety features like gas escape detection [55]. The necessary management and security systems are beyond the scope of this analysis and are not explicitly considered.

Methodology

Optimization model of self-sufficient residential supply system for 2030

A hypothetical residential energy system has been developed as a mixed-integer linear program within the Framework for Integrated Energy System Assessment (FINE) to determine the cost-minimal supply system and its operation for self-sufficient buildings. It is built on the optimization framework Pyomo [56] and is available open source.⁶ The model represents decisions on the buildings' energy system structure (technology selection), sizing

⁴ https://www.homepowersolutions.de.

⁵ https://lavo.com.au/.

⁶ https://github.com/FZJ-IEK3-VSA/FINE.

⁷ with a 90% degree of loading.

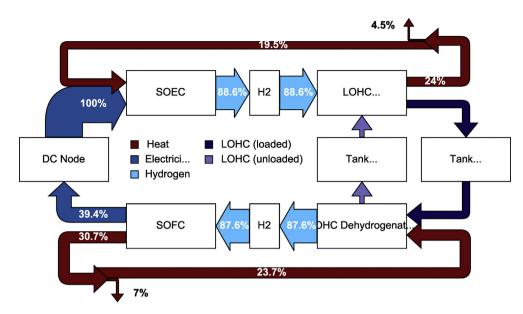


Fig. 1 – Energy flows of a solid oxide electrolysis (SOEC) and fuel cell (SOFC) combined with a liquid organic hydrogen carrier (LOHC) system, with scaling of the SOEC parameters based on Peters et al. [41]. Heat generated in fuel cell mode is employed for the dehydrogenation of the carrier fluid, whereas excess heat from hydrogenation is used to preheat and evaporate the electrolysis feed water.

(capacity of the energy supply units), and operation for one representative year within a set of linear equations with continuous and binary variables. The linear objective function minimizes the total annualized costs (TACs). Therefore, we assume that the annual economic interest rate (i) over the component lifetime (n) for the house owner is 3%. The TAC is calculated as in:

$$TAC = CAPEX*\left(\frac{i}{1-\left(1+i\right)^{-n}} + OPEX\right)$$

Capital expenditures (CAPEX) are the sum of fixed and capacity-variable investment costs. The operational and maintenance expenditures (OPEX) are expressed as a fraction of the overall capital costs.

In the following, we describe a modeling framework for the component groups. All of the techno-economic parameters employed are listed in Tables 1 and 2. The structure of the modeled energy system is displayed in Fig. 2. A Python notebook of the model with the underlying data to reproduce the results is available online.⁸

DC power subsystem

DC power provided from PV is transmitted to the DC node of the system, connected to which is a lithium-ion battery, the hydrogen subsystem (the fuel cell and electrolysis cell, respectively, and the rSOC), an electric water heater, and an inverter. The inverter connected to the DC and AC nodes converts DC power from PV or the fuel cell into AC power to serve the electrical demands of the household or heat pump.

Hydrogen storage subsystem

The selection of technologies includes pressurized hydrogen storage at a pressure level of 16 MPa or a LOHC system. Fig. 3 displays an illustration of the hydrogen subsystem. Hydrogen in the system is available at three pressure levels: 16 MPa, 2 MPa, and atmospheric pressure. Compressors achieve an isentropic efficiency of 75% and mechanical efficiency of 92%. As the compressors are separately modeled, the pressurized hydrogen storage tank is not fitted with further charge or discharge efficiencies. The compressors' energy demand is then calculated using data from CoolProp, which was developed by Bell et al. for thermo-physical fluid properties [69]. It is assumed that no notable loss of hydrogen occurs in the compressors. Compressor 1 (2 MPa) has a ratio of 17.51, based on the lower heating value of hydrogen compressed per unit of electric power consumed (H₂LHV/power). The compressor consists of two compression stages with an intercooler. These two stages compress the hydrogen to 2 MPa, which can subsequently be compressed further or used for the LOHC's hydrogenation. The second compressor reaches a pressure level of 16 MPa with an H₂LHV/power ratio of 22.97.

Alternatively, an LOHC storage system can be used. The dibenzyltoluene (DBT, $C_{21}H_{20}$) is held in two separate tanks for unloaded (H_0 -DBT) and loaded (H_{18} -DBT) states. These tanks can be conventional heating oil tanks [47]. The exothermic hydrogenation and endothermic dehydrogenation, in this long-term storage approach, would cause a major loss of efficiency. The specific energy demand is 65.4 kJ/mol $_{H2}$, which is 27.06% and relates to the lower heating value of the stored hydrogen [70]. Therefore, the efficiency is enhanced via heat

⁸ Knosala, Kevin; Kotzur, Leander; Röben, Fritz T.C.; Stenzel, Peter; Blum, Ludger; Robinius, Martin; Stolten, Detlef (2021), "Hybrid Hydrogen Home Storage for Decentralized Energy Autonomy", Mendeley Data, https://doi.org/10.17632/zhwkrc6k93.1.

Table 1 — Economic parameters. Costs for balance of plant, energy management and safety controlling systems are included in the component costs. We assume an annual economic interest rate for the house owner of 3%. CAPEX: capital expenditures, OPEX: operational expenditures; PEM: proton-exchange membrane; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier; DBT: dibenzyltoluene.

Component			CAPEX			OPEX	Lifetime		Source	
	Fix		Variable		Fix					
Photovoltaic system	_	-	769	€/kW _p	1	% Inv./a	20	years	[57]	
Battery-electric system	_	_	301	€/kWh _{el}	_	_	15	years	[57]	
Redox flow system	3000	€	835	€/kW _{el}	2	% Inv./a	15	years	own assumptions	
			190	€/kWh _{el}						
PEM fuel cell	2000	€	1000	€/kW _{el}	1	% Inv./a	15	years	[29,37,58]	
PEM electrolysis	2000	€	1000	€/kW _{el}	1	% Inv./a	15	years	own assumptions	
rSOC	5000	€	2400	€/kW _{el}	1	% Inv./a	15	years	own assumptions	
Heat pump	4230	€	504.9	€/kW _{th}	1.5	% Inv./a	20	years	[59]	
Thermal energy storage	_	_	90	€/kWh _{th}	-	_	25	years	[60]	
Electric heater	_	_	60	€/kW _{th}	2	% Inv./a	30	years	[60]	
Compressor 2 MPa	_	_	1717	€/kW _{el}	_	_	20	years	[61]	
Compressor 16 MPa	560	€	1330	€/kW _{el}	_	_	20	years	[61]	
Pressurized hydrogen storage	_	_	15	€/kWh _{H2}	_	_	25	years	[37,62]	
LOHC fluid (DBT)	_	_	1.25 ⁷	€/kWh _{H2}	_	_	25	years	[34,63]	
LOHC tank	_	_	0.79	€/kWh _{H2}	_	_	25	years	[64,65]	
LOHC hydrogenation unit	2123	€	761	€/kW _{H2}	1	% Inv./a	20	years	[34]	
LOHC dehydrogenation unit	1140	€	409	€/kW _{H2}	1	% Inv./a	20	years	[34]	

Table 2 — Technical parameters. PEM: proton-exchange membrane; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier; DBT: dibenzyltoluene; HT: high temperature; LT: low temperature.

Component	су	Source	
Inverter	η_{el}	97%	own assumptions
Lithium-ion battery	η_{charge}	95%	own assumptions
	η _{discharge}	95%	own assumptions
	Self-discharge	0.01%/h	own assumptions
Redox flow battery	η_{charge}	86.6%	own assumptions
	η _{discharge}	86.6%	own assumptions
	Self-discharge	_	own assumptions
PEM fuel cell	η_{el}	55%	[37]
PEM electrolysis	η_{el}	70%	[66]
rSOC electrolysis mode (w/o heat integration)	η_{H2}	73.2%	[41]
rSOC fuel cell mode	η_{el}	45%	[66,67]
	η_{th}	35%	[66,67]
Heat pump	COP_{\min}	2.86	[68]
	COP_{max}	7	[68]
Thermal storage	η_{charge}	99%	own assumptions
	$\eta_{ m discharge}$	99%	own assumptions
	Self-discharge	0.1%/h	own assumptions
Electrical heater LT	η_{el}	98%	own assumptions
Electrical heater HT	η_{el}	98%	own assumptions
Heat exchanger	η	99.5%	own assumptions
Hydrogenation	η	99.5%	own assumptions
Dehydrogenation	η	99.5%	own assumptions
LOHC fluid	Self-discharge	-	own assumptions
Compressor 2 MPa	η_{el}	94.3%	own assumptions
Compressor 16 MPa	η_{el}	95.6%	own assumptions

integration between the LOHC reactor and rSOC system. For the hydrogenation process, 0.3 kWh heat is generated for the storage of 1 kWh of hydrogen. For the dehydrogenation process the same amount of heat is required, and therefore it is considered separately from the efficiencies. Based on the hydrogenation reaction of DBT with hydrogen [63], the specific

amount of DBT necessary per mass of hydrogen stored is calculated by the following formula:

$$\frac{m_{\rm DBT}}{m_{\rm H2}} = \frac{1}{\rm LF} * \frac{M_{\rm DBT}}{9*M_{\rm H2}} = 16.68 \frac{kg_{\rm DBT}}{kg_{\rm H2}} \tag{2}$$

where m_{DBT} is the mass of DBT necessary to store a mass of

Table 3 — Built storage capacity and conversion-rated power for different technology configurations as a result of design and operation optimization for one representative year. LIB: lithium-ion battery; RFB: redox-flow battery; PEM: proton-exchange membrane; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier. Photovoltaic orientations: NW: northwest; SE: southeast; electric heater classes; HT: high temperature; LT: low temperature.

	Photovoltaic NW [kW _p]	Photovoltaic SE [kWp]	Inverter [kW _{el}]	Heat pump [kW _{th}]	Electric heater LT [kW _{th}]	Electric heater HT [kW _{th}]	Redox flow battery [kWel]	PEM fuel cell [kW _{el}]	PEM electrolysis [kW _{el}]	rSOC [kWel]	LOHC dehydrogenation [kW _{el}]	LOHC hydrogenation [kW _{el}]	Thermal storage [kWh _{th}]	Lithium-ion battery [kWh _{el}]	Redox flow battery[kWh _{ei}]	Hydrogen storage[kWh _{H2}]	LOHC storage[kWh _{H2}]
LIB	13.4		6.0	6.8	0.0	0.0	-	-	-	-	-	-		458.1	-	-	-
LIB RFB	13.4	13.4	6.0	2.2	0.2	0.0	2.9	-	-	-	-	-	0.0	66.6	504.1	-	-
LIB PEM	0.0	13.4	6.0	1.7	0.6	0.0	-	0.9	0.7	-	-	-	9.5	17.4	-	2014.8	-
LIB rSOC	0.0	13.4	6.0	1.3	0.6	0.2	-	-	-	0.7	-	-	8.0	17.1	-	2078.7	-
LIB PEM LOHC	0.0	13.4	6.0	1.7	0.0	0.7	-	1.4	2.9	-	2.4	0.9	5.1	14.7	-	178.4	3917.7
LIB rSOC LOHC	0.0	11.5	6.0	1.6	0.0	0.0	-	-	-	0.9	2.0	0.8	3.6	15.7	-	0.0	2726.9

hydrogen $m_{\rm H2}$, LF is a load factor of 0.9, which defines that not all of the DBT can be actively utilized and remains in the respective tanks, $M_{\rm DBT}$ is the molar mass of the DBT, and 9* $M_{\rm H2}$ are the molar masses of the 9 bound hydrogen molecules that react with unloaded DBT during the hydrogenation reaction. The resulting ratio is 16.68 kg_{DBT}/kg_{H2}.

Hydrogen conversion subsystem

As rSOC systems are only used in laboratory environments, assumptions regarding their performance in residential buildings will be made based on experience with separate SO fuel and electrolysis cells. The rSOC electrolysis process requires high-temperature heat to run. A power input of 1 kW $_{\rm el}$ and 0.21 kW $_{\rm th}$ results in 0.886 kW $_{\rm H2}$. The high-temperature heat can either be provided by a high-temperature electrical heater or via heat integration with the hydrogenation process. The rSOC in fuel cell mode produces high-temperature heat. A power output of 1 kW $_{\rm el}$ produces 0.78 kW $_{\rm th}$ of high-temperature heat and consumes 2.22 kW $_{\rm H2}$, resulting in an electric efficiency of 45% and a thermal efficiency of 35%. The PEM electrolysis heat is provided by the fuel cell itself and included in its efficiency measure. A power input of 1 kW $_{\rm el}$ results in 0.7 kW $_{\rm H2}$. The PEM fuel cell

cannot provide any exhaust heat to other components, and a power output of 1 kW_{el} requires 1.82 kW_{H2} , resulting in an electrical efficiency of 55%.

Heating subsystem

Heat can be supplied by means of two conversion units: Either by an electric heater with an efficiency of 99% or a heat pump with a time-dependent coefficient of performance (COP) that

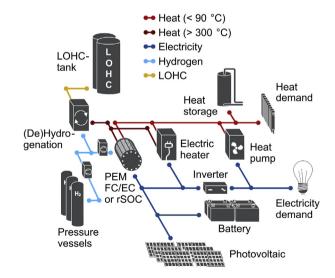


Fig. 2 — Model of a self-sufficient residential energy system. If an rSOC is installed, high-temperature process heat (c. 300 $^{\circ}$ C) can be exchanged between the LOHC reactor and the rSOC. EC: electrolysis cell, FC: fuel cell, LOHC: liquid organic hydrogen carrier, PEM: proton-exchange membrane, rSOC: reversible solid oxide cell.

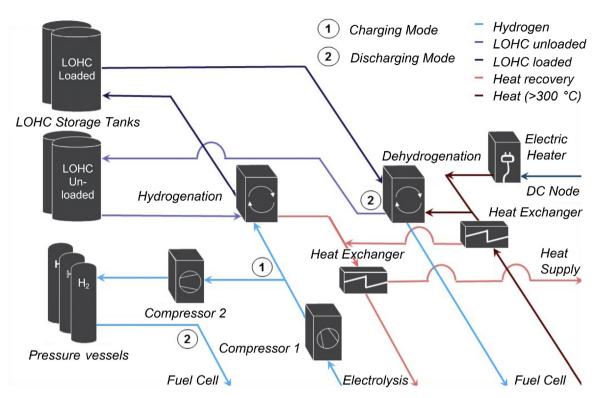


Fig. 3 - Model of a hydrogen subsystem. LOHC: liquid organic hydrogen carrier.

is modeled according to Lindberg et al. [60]. A supply temperature T^{sup} of 40 °C was considered, with a quality grade η_C of 45%, and ambient air temperature T_t^{amb} being drawn from the representative year weather data for every time-step t. The COP was then calculated thus:

$$ext{COP}_{t} = \eta_{C} \; rac{ ext{C}}{ ext{T}^{ ext{sup}} - ext{T}^{ ext{amb}}_{t}}$$

Case study

PV generation profiles

Solar irradiation was determined from weighted average temperature and irradiance data for the typical meteorological year (TRY-6) of 2010 from the German Meteorological Service [71]. The simulation of one weather year was suitable to depict tendencies among residential energy supply systems at Central European latitudes, and does not permit conclusions of the exact dimensioning for the edge cases of such systems. The data for this location reveals periods of radiation under 20% (compared to peak levels) for up to two weeks in the months of January and February. The PV system is simulated with PV-Lib for 300 W Silevo Triex U300 Black⁹ modules. The roof tilt was 37°. Under standard test conditions, these modules attained an efficiency of 17.8%. A maximum of 26.8 kW_p at standard conditions could be installed on the 131.9 m² roof area (window areas were not considered). Ground-mounted

Load profiles

Electricity and heat load profiles were calculated at an hourly resolution using the openly available Python module TSIB [72]. Based on a simulation of the occupants' behavior with the CREST¹⁰ module, the electric device load and demand for thermal comfort were derived. The considered thermal heat flows include heat transfer from irradiation, ventilation, ingress air, transmission, and internal heat gains. These were obtained by combining the simulated building occupancy profiles with the heat transfer coefficients of the building hull in a simplified 5R1C thermal building model [64]. The weather data for the test reference year was used for outside irradiance and temperature [71]. The house envelope was modeled assuming a detached, modern, energy-efficient building with a room heating demand of 18.6 kWh/m² per year. Drinking water was heated by an electrical water heater and included in the overall electricity demand. Energy demand for cooling was not considered. The simulated annual energy demand for a 187 m² living area and four residents is 4062 kWh of electricity and 3481 kWh of heat. The load profiles for electricity and heat can be found in the Appendix in Figure A-2.

PV modules were not considered. The PV capacity factor profiles (kW/kW_p) for the south-east and north-west orientation can be found in the Appendix in Figure A-1.

⁹ http://www.solardesigntool.com/components/module-panelsolar/Silevo/2272/Triex-U300-Black/specification-data-sheet. html.

¹⁰ https://www.lboro.ac.uk/research/crest/demand-model/.

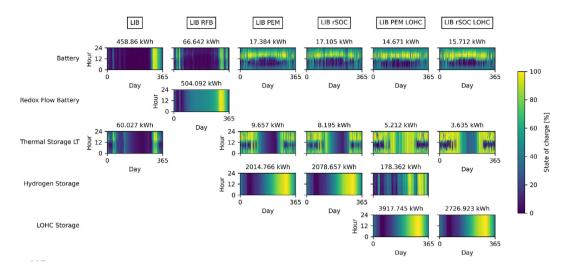


Fig. 4 — Storage capacities and state-of-charge (in percent) for different technology combinations as a result of design and operation optimization for the self-sufficient energy supply of a residential building with PV as the only energy source for one representative year. The columns depict different technology options for the energy storage solutions: LIB: lithium-ion battery; RFB: redox-flow battery; PEM: proton-exchange membrane fuel cell and electrolysis; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier. The color indicates the state-of-charge of the storage in percent. The abscissa represents the day of the year, and the ordinate the hour of the day. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

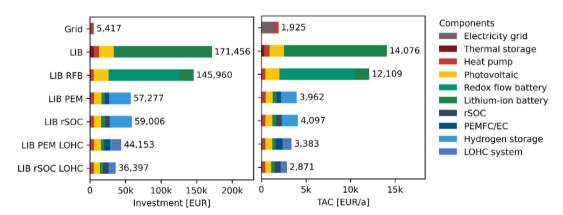


Fig. 5 — Total investment costs and total annualized cost for different technology configurations as a result of design and operation optimization for one representative year. Explicit values for each component can be found in the Appendix in Tables A-1 and A-2. LIB: lithium-ion battery; RFB: redox-flow battery; PEM: proton-exchange membrane fuel cell and electrolysis cell; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier.

Energy supply scenarios

Based on the self-sufficient system shown in Fig. 2, we selected different technology configurations that differed in their choice of storage. The cases comprised a lithium-ion battery electric system only (LIB) or a lithium-ion battery supported by a redox flow battery system (LIB RFB), hydrogen

pressure vessels (LIB PEM, LIB rSOC), or hydrogen pressure vessels and LOHC storage capacities (LIB PEM LOHC, LIB rSOC LOHC). Heat storage (80 °C) is possible in all configurations. Sizing and operation for a self-sufficient supply have been optimized for the representative year with 1-h time-steps towards an objective function representing the equivalent total

annualized cost. As a reference case, exclusive electricity supply from the grid was considered (appliances and heating exclusively utilizing a heat pump). The price of electricity was 0.30 €/kWh, which corresponds to the average price of electricity for German households in 2019 [73]. We do not assume an increase for the year 2030 in the scenarios.

Results and discussion

Fig. 5 show the resulting optimal values for the conversion and storage capacities, as well as the resulting equivalent annual and investment costs. The results indicate that selfsufficient operation within a PV capacity limit of 26.8 kW_p requires large seasonal storage capacities to cover periods with low or no PV generation. With only battery storage available (LIB), a relatively large heat pump with a rated power of 6.8 kW_{th}, electrical storage of 458.9 kWh_{el}, and thermal storage of 60.0 kWh_{th} (1.0 m³) is needed to ensure supply during winter periods with low PV generation. With the support of a redox flow battery, thermal storage can be neglected in favor of more electrical storage (570.7 kWhel), resulting in approximately the same energy storage capacity as in the LIB case and no reduction in PV generation capacity. Adding the possibility of hydrogen storage to the system reduces the need for PV generation on the less efficient northwestern parts of rooftops in each case and the PV usage factor per installed capacity is higher. It can be observed that reduced generation capacity is turned into a hydrogen storage size up to ten times larger than the battery capacities in the LIB case. The heat integration between the rSOC and LOHC systems (LIB rSOC LOHC) reduces hydrogen storage needs by 25% compared to the combination of PEM and LOHCs (LIB PEM LOHC). The former requires a 2.9 kW_{el} electrolysis capability and 1.4 kW_{el} fuel cell-rated power, whereas the latter manages to supply the building with an rSOC-rated power of 0.9 kWel (for both operating modes). The LOHC dehydrogenation in the LIB PEM LOHC system must be electrically-heated, as no excess heat from the PEM fuel cell can be used. This electricity demand is reduced in the LIB rSOC LOHC system by utilizing excess heat from the rSOC. High-pressure hydrogen vessels can then be omitted, and significantly decreasing thermal storage size.

The storage operation pattern for the different technology combinations displayed in Fig. 4 shows how storage durations and frequencies differ for the same technology in other system settings. The size of the battery in the LIB case is determined by a single storage cycle from autumn to winter, resulting in a storage capacity of 458.9 kWh $_{\rm el}$. The advantage of a fast-reacting battery storage device would be inefficiently used in this case. Systems with batteries and fuel cells exhibit a high capacity for hydrogen storage for long-term applications and lower capacity battery storage for short-term periods (17.4 kWh $_{\rm el}$ for LIB PEM, 17.1 kWh $_{\rm el}$ for

LIB rSOC). In the case of a PEM fuel cell and electrolysis with LOHC storage, a separation of storage patterns for daily load-shifting with the battery and weather fluctuations with the hydrogen storage and seasonal patterns with the LOHC storage can be observed. The heat integration (LIB rSOC LOHC) results in less LOHC storage capacity and fewer components (no pressure vessels) than the combination of the PEM and LOHC. Furthermore, it can be observed that a small battery is highly beneficial to all systems for intra-day load shifting. By serving the load peaks, it allows for the lower rated power of the conversion components (i.e., electrolysis and fuel cells).

The cost comparison in Fig. 5 illustrates that the heat-integrated rSOC LOHC system reduces investment costs by 80% compared to the LIB case. The high investment for the LIB system is due to the high capacity costs of battery storage. As essential material costs set lower limits on battery prices, learning curve models that take into account substantial growth in the market for electric vehicles set a lower boundary at 114 €/kWh_{el} (124 \$/kWh_{el})¹¹ for current battery technology in 2030 [74]. If this lower boundary is used in the LIB case instead of the assumed 301 €/kWh_{el}, the total system investment costs are reduced to €119,230, which is still three times the cost of the LIB rSOC LOHC system. The high battery storage capacity needed for fully energy autarkic supply remains costly, even with more optimistic price reductions assumed for battery storage.

Hydrogen storage with pressure vessels reduces system investment costs by 71% compared to the PV LIB case, but achieving a large storage capacity with pressure vessels still proves expensive. When pressure vessels are selected, a PEM system is the preferred choice due to its more variable operation and lower operating temperatures.

Combined with LOHC storage, the LIB rSOC LOHC system exhibits the largest reduction (80%) compared to the PV LIB case. Learning curve models for residential fuel cells suggest a price range of between 1800 and 3670 €/kW_{el} (2000 and 4000 \$/kW_{el}) for 2030, including uncertainties in growth and experience rates for a cumulative installed rated power of 5 GW [75]. The prognosis for residential fuel cell-based micro-CHP systems fueled by natural gas are in the same range [76]. The assumed investment costs for the optimal solution for the rSOC in the LIB rSOC LOHC case is €7190 for 0.91 kW_{el} (7901 €/kW_{el}) rated power. As the rSOC combines fuel cell and electrolysis functions in one component, this price can be considered attainable for a 2030 system.

Compared to the base case with the grid supply in Germany, where the majority of annual expenses (€1485 or 77%) were spent purchasing electricity from the grid, the yearly premium that a house owner would have to pay for a fully self-sufficient system is 52% of the price of the full electricity supply from the grid (for appliances and heating exclusively with a heat pump). The prices for electricity purchases are subject to uncertainty in the market, and therefore could potentially become higher in the future.

¹¹ Exchange rate: \$1 = €0.9.

Previous studies at the municipal level have shown the average levelized costs of electricity (LCOE) for electrical self-sufficiency to be 0.41 \$/kWh_{el}\$ [13] and 0.41 \$\infty\$/kWh_{el}\$ in the German case [77]. We calculated the LCOE for the single-family house as the ratio of annualized cost for the electricity supply (excluding the costs for heat-related components such as heat pumps, heat storage, and electric heaters) over the building's consumed electric power. In the case with grid supply, the LCOE corresponds to the price for electricity from the grid, which is 0.30 \$\infty\$/kWh_{el}\$. The cost-optimal hydrogen case (LIB rSOC LOHC) results in an LCOE of 0.42 \$\infty/kWh_{el}.

Conclusions

In this paper, we showed that hybrid hydrogen home storage systems, in combination with highly energy-efficient buildings, can enable fully energy-autarkic residential buildings to be realized. As a case study, we analyzed a single-family residential supply system with roof-mounted PV as the only source of energy and compared different storage technology configurations for an energy self-sufficient supply. Under our assumptions, energy self-sufficiency can be achieved with hydrogen storage for an annual premium of 52% compared to an electricity supply from the grid by 2030. Although battery storage is optimal for short-term uses, substantially lower storage capacity costs for seasonal storage are desirable. The use of a hydrogen conversion and storage system yields total annualized cost reductions of 72-80% for the self-sufficient supply of electricity and heat throughout the year compared to lithium-ion battery systems. For high-pressure hydrogen storage, the choice of technology between a PEM or rSOC system results in similar TACs (reductions of 72 or 71%). The application of LOHCs as a storage medium exhibits a clear cost reduction for both conversion technologies compared to high-pressure storage (76-80%). By thermally-integrating the LOHC reactor with a hightemperature rSOC, which is not possible with a lowtemperature PEM, total conversion and storage capacities can be further reduced, yielding the most economical solution.

Given the established, fast-growing market of residential PV—battery systems, it can be assumed that customers are not merely concerned with economic factors when opting for energy self-sufficiency. In particular, private customers see home storage systems as consumer goods, rather than pure capital goods. Independence from suppliers and future price developments, contribution to the energy system transformation, and an overall interest in the technology all play a part in the decision to obtain a self-sufficient energy supply [4]. Nevertheless, we demonstrated that battery storage alone is not sufficient for the energy-autarkic supply of residential buildings.

It must be noted that modeling assumptions regarding system technology, input data, and constraints highly influenced the results of this study. Site conditions strongly impinge the dimensioning of the energy supply system. It should also be noted that dimensioning of the components is

based on perfect foresight regarding optimally-dimensioned components for one typical weather year, whereas in a real application, safety factors would need to be considered to cover edge cases. However, a storage extension to cover such situations can be achieved at a low cost in the case of LOHCs. Therefore, in accordance with previous studies [30], our results confirm that hydrogen storage systems will be indispensable to increased energy autarky in future energy systems located at high geographical latitudes.

Compared to a single-family house, self-sufficiency can be more simply achieved by means of a group of different demand profiles through diversification in multi-family houses or districts [78]. It has been shown in previous studies that the economically-optimal level of self-sufficiency modulates with system scale [79]. Solutions for joint self-consumption in multi-apartment buildings, districts, municipalities, and countries must therefore be further investigated at the technological and regulatory levels in order to promote the smart interlinking of generation and demand systems.

Data availability

The authors note that the data underlying this analysis, as well as the source code for the reproduction of results, is published under:

Knosala, Kevin; Kotzur, Leander; Röben, Fritz T.C.; Stenzel, Peter; Blum, Ludger; Robinius, Martin; Stolten, Detlef (2021), "Hybrid Hydrogen Home Storage for Decentralized Energy Autonomy", Mendeley Data, https://doi.org/10.17632/zhwkrc6k93.1.

Author contributions

P.S. and L.K. developed the research idea. F.R. and K.K. collected the techno-economic data and created the model. P.S., L.K., and K.K. analyzed the data and interpreted the results. M.R. and L.B. participated in the discussion and data analysis. D.S. and M.R. supervised the project. K.K. wrote the manuscript with support from P.S., L.K., F.R., and M.R. All of the authors contributed to discussions and the review and editing of the manuscript.

Declaration of competing interest

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

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Appendix

1. Time-series data.

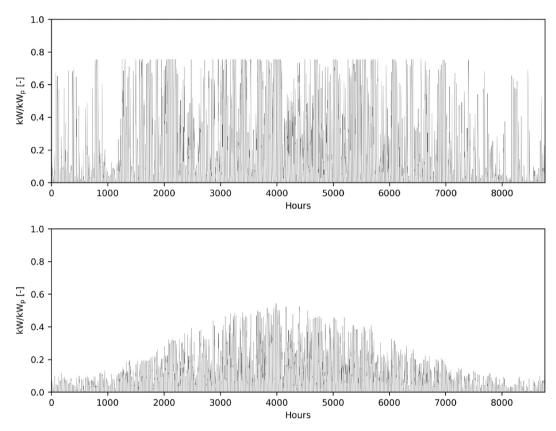


Figure A-1 PV capacity factors for southeastern (top) and northwestern roof orientations (down).

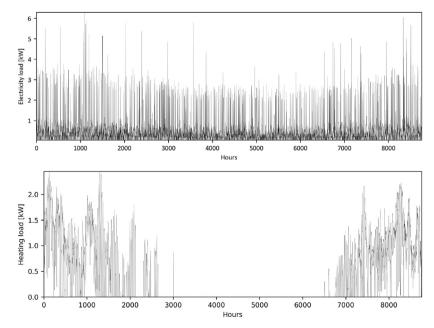


Figure A-2 Electricity (top) and heating load profiles (down) for the single-family building.

Optimal investment and total annualized cost per component.

Table A-1 Investment costs for the optimal supply system per scenario and component. LIB: lithium-ion battery; RFB: redox-flow battery; PEM: proton-exchange membrane fuel cell and electrolysis cell; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier.

		Investment cost [€]									
	Grid	LIB	LIB RFB	LIB PEM	LIB rSOC	LIB PEM LOHC	LIB rSOC LOHC				
Heat pump	5221.37	7643.81	5336.15	5103.23	4894.81	5102.32	5056.20				
Thermal storage	196.37	5318.28	0	854.87	724.76	459.39	320.53				
Electricity grid	0	_	_	_	_	_	_				
Photovoltaic	_	20,597.60	20,597.60	10,298.80	10,298.80	10,298.80	8853.31				
Redox flow battery	_	-	99,990.56	_	_	_	_				
Lithium-ion battery	_	137,897.00	20,035.90	5227.77	5143.28	4412.41	4725.26				
rSOC	_	_	_	_	6765.04	_	7190.22				
PEM	_	_	_	2696.99	_	8282.11	_				
Hydrogen compressor	_	_	_	636.09	661.63	950.04	79.29				
Hydrogen storage	_	_	_	30,221.48	31,179.85	2675.44	0				
LOHC de-/hydrogenation	_	_	_	_	_	4931.08	4689.31				
LOHC storage	-	_	_	_	_	7992.20	5562.92				

Table A-2 Total annualized cost for the optimal supply system per scenario and component. LIB: lithium-ion battery; RFB: redox-flow battery; PEM: proton-exchange membrane fuel cell and electrolysis cell; rSOC: reversible solid oxide cell; LOHC: liquid organic hydrogen carrier.

1 5 7 5		TAC [€/year]										
	Grid	LIB	LIB RFB	LIB PEM	LIB rSOC	LIB PEM LOHC	LIB rSOC LOHC					
Heat pump	429.28	628.44	438.72	419.57	402.43	419.49	415.70					
Thermal storage	11.32	305.67	0	49.20	41.70	26.48	18.48					
Electricity grid	1485.32	_	_	-	-	_	-					
Photovoltaic	_	1591.02	1591.02	796.02	796.02	796.34	684.41					
Redox flow battery	_	_	8400.31	-	-	_	-					
Lithium-ion battery	_	11,551.73	1678.90	438.76	431.64	370.34	396.65					
rSOC	_	_	_	-	634.34	_	674.20					
PEM	_	_	_	522.46	-	776.60	-					
Hydrogen compressor	_	_	_	94.80	92.53	141.59	11.82					
Hydrogen storage	_	_	_	1736.03	1791.13	154.13						
LOHC de-/hydrogenation	_	_	_	_	_	380.75	362.08					
LOHC storage	_	_	_	_	-	459.39	319.80					

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