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Utilisation of reactive ionic liquids for energy storage and regulation of the power grid

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Chlorine is an essential feedstock for polymers and pharmaceuticals, with annual production exceeding 100 Mt. Nearly all Cl₂ is obtained by chlor-alkali electrolysis, which consumes about 2.58 MWh of electricity per tonne. Renewable energies such as wind and solar would lower CO₂ emissions, but electrolyzers would then have to adopt to fluctuating, renewable energies while downstream processes still require a constant chlorine supply. Large inventories of pressurised, liquefied Cl₂ are impractical owing to toxicity and stringent safety setbacks. Triethylmethylammonium trichloride, [NEt₃Me][Cl(Cl₂)], could become a key technology enabling safe chlorine storage. This room-temperature ionic liquid, made from inexpensive [NEt₃Me]Cl and chlorine, can store 0.79 t_{Cl₂} t⁻¹ while maintaining a low chlorine vapour pressure. Chlorine can be released by mild heating, vacuum, or water addition; alternatively, the loaded liquid can serve directly as a chlorinating agent. With an energy density near 1.1 MWh t⁻¹, the trichloride could couple electrolysis with real-time power availability and allow chlorine to be produced in sun-rich regions and shipped safely elsewhere.

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1. In this manuscript, new technologies to store and handle chlorine are discussed. Those technologies may support the production of chlorine with renewable energies, helping balancing of the electrical grid and reducing carbon dioxide emissions.
2. Chlorine production is extremely energy intensive, *e.g.*, consuming approximately 1.7% of the electricity in Germany. Currently, chlorine production is highly baseload dependent, so relying on fossil energies, due to the lack of a safe chlorine storage.
3. With the help of new chlorine storage systems, the production of chlorine can be adapted to the availability of renewable, yet fluctuating energies. This manuscript also outlines the enormous energy demand of the chlorine industry and the lack of innovation in this important field.

Introduction

The ongoing climate crisis demands an urgent shift toward cleaner energy sources to reduce greenhouse gas emissions. Consequently, solar and wind power have witnessed remarkable growth because they are essentially carbon-free during operation.^{1,2} The resulting increase in the intermittent generation capacity of wind and solar poses challenges to the current energy system, which is based on large, centralised and constant generation capacity.³ Addressing these chal-

lenges necessitates enhancing the accuracy of forecasts, expanding and modifying grids, and in particular increasing system flexibility.⁴ Moreover, energy-intensive industries, such as the chemical sector, are also facing escalating electricity price volatility due to the increasing share of intermittent renewables. These industries typically operate on a year-round, continuous basis, with only intermittent interruptions for maintenance purposes, and have a substantial and constant demand for electricity. Combined with high price volatility, this constant demand has a growing negative impact on their economic performance.⁵ This increased demand for system flexibility in the power supply system, coupled with the mismatch between variable renewable output and constant industrial demand⁶ underscores the need for robust energy-storage systems capable of buffering supply shortfalls and preventing costly downtime as much as storing energy in periods of high wind and solar generation to avoid the need of expensive gas-powered generation

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in other periods. In response, the scientific community and industry stakeholders are investigating a broad range of solutions, each offering unique advantages, design complexities, and cost profiles.^{7,8}

Electrochemical energy storage (EES) is one of the most established categories, primarily encompassing battery technologies such as lead-acid, lithium-ion, and sodium-sulfur. These convert electrical input into chemical energy and release it on demand, making them suitable for short- to mid-term storage (minutes to hours). Commercial lithium-ion batteries provide near-instantaneous balancing capabilities but are until now limited to a specific energy of 90–300 kWh_{el} t^{−1} (higher specific energies could be achieved in the future).^{9–11} Recent progress in solid-state and metal-air batteries has shown promise for enhancing both energy density and operational safety.^{7,10–12} However, issues such as raw material supply (e.g., lithium, cobalt) and end-of-life disposal remain areas of ongoing research and policy development.¹³

Electrical energy storage mechanisms store energy directly in electric or magnetic fields, as in capacitors, supercapacitors, or superconducting magnetic energy storage (SMES). Capacitors and supercapacitors support high-power applications and instantaneous load balancing, while SMES systems provide similarly fast response at excellent round-trip efficiencies—albeit at considerable cryogenic costs (Fig. 1).^{8,14}

Thermal energy storage (TES) relies on heating or cooling a working medium—such as water, molten salts, or phase-change materials—to store energy for later release. Sensible-heat systems (e.g., hot-water tanks) help shift loads for building heating or industrial processes, whereas phase-change materials can store more heat per unit mass or volume, thus improving energy density and maintaining a constant tempera-

ture level.^{7,12} Thermochemical storage is also emerging as a means of achieving extended duration capacity, though it entails higher engineering complexity.¹⁵

Mechanical energy storage stores potential or kinetic energy. Pumped-hydro energy storage (PHES), already in widespread use, elevates water to a higher reservoir when surplus electricity is available, then recovers it through turbines. Compressed air energy storage (CAES) uses underground caverns or vessels to store pressurized air, while flywheel systems rely on massive rotating disks to hold kinetic energy.^{16,17} Although these technologies are proven even on large scales, they are constrained by relatively high capital costs.⁸

Lastly, chemical energy storage encompasses “power-to-X” carriers, such as hydrogen, ammonia, and synthetic natural gas, which are generated from surplus electricity and then either stored until needed. Upon demand, they can be re-converted to electricity *via* fuel cells or turbines, providing long-term, seasonal balancing and leveraging existing infrastructure. In addition, hydrogen and ammonia can also be used directly as they are crucial base chemicals finding countless applications in the chemical industry.^{18–20}

A parallel area of research to power-to-X energy storage is demand-side flexibility (demand response) for energy-intensive industrial processes. This enables consumers to reduce their electricity costs by shifting their load profile to times when electricity prices are lower, and/or to generate additional income through the targeted marketing of load flexibility.²¹ The combination of demand response with product storage (power-to-X) in energy-intensive industries is of particular relevance due to the constant and central availability of their high loads throughout the year, as well as the existing infrastructure.²²

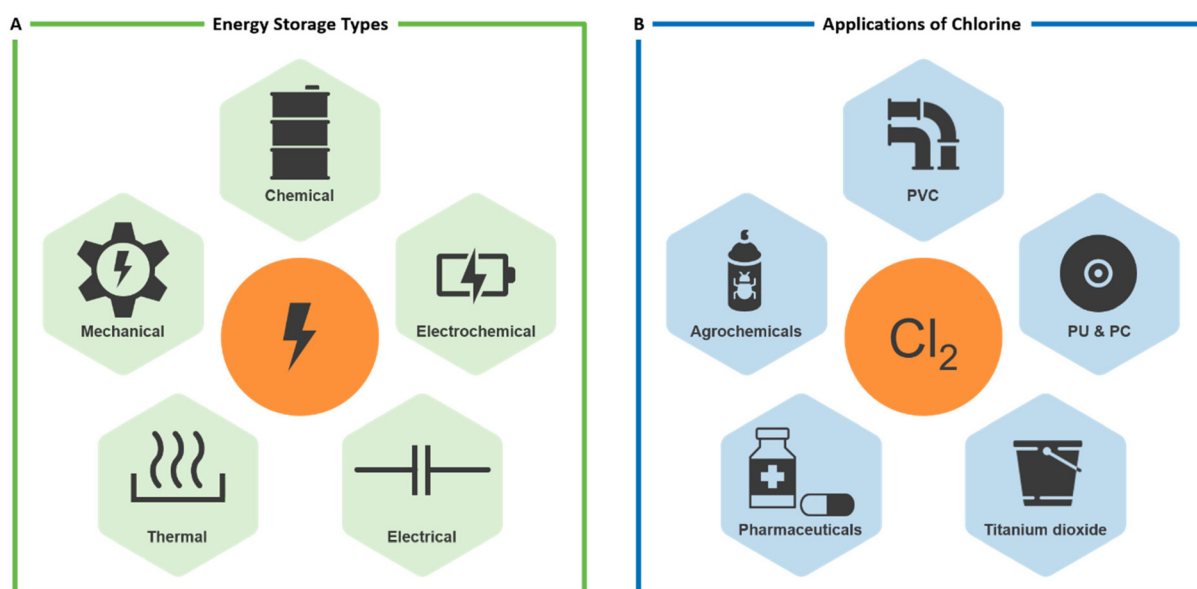


Fig. 1 Important types of energy storages (A) and selected applications of chlorine (B).



In this context, we focus on a base chemical for which the production requires large quantities of electrical energy: elemental chlorine (Cl_2).

Chlorine as an energy-intensive base chemical

Chlorine ranks among the most versatile base chemicals, being indispensable for vast segments of the chemical industry.^{23–25} It is required for the production of over 50% of all industrial chemicals and polymers,²⁵ for 90% of all pharmaceuticals, and 86% of all agrochemicals.²⁶ For example, chlorine is reacted with ethylene to ethylene dichloride (EDC) which is then cracked providing vinyl chloride, the precursor for polyvinyl chloride (PVC), a versatile polymer that was produced on a global scale of 38 million tons in 2015 (German production in 2022: 1.209 Mt).^{27,28} Related to that, polyurethanes (PU, German production in 2022: 0.977 Mt)²⁸ and polycarbonates (PC, German production in 2022: 0.369 Mt)²⁸ are commonly derived from phosgene (COCl_2), a chemical produced from chlorine and carbon monoxide on a global scale of approximately 12 mio t per year.²⁹ In addition, many other materials, *e.g.*, silicones, phosphorus trichloride (important for agrochemicals, pharmaceuticals, and flame retardants), and some metals (*e.g.*, titanium for TiO_2) require chlorine for their production.^{23–25,30}

Today, the global chlorine production exceeds 100 million tons annually and is even growing.³¹ In Europe, 95.2% of chlorine is produced *via* chloralkali electrolysis, which provides caustic soda (NaOH) and hydrogen as valuable by-products. The chloralkali electrolysis can be further subdivided into the modern membrane process accounting for 86.8% of the European chlorine production and the older but still partially used diaphragm process accounting for 8.4% of the European chlorine production. The remaining 4.8% is produced by other methods, such as the reaction of HCl and NaOCl to yield chlorine.³² Although the membrane process is highly optimized, each ton of chlorine requires on an average 2.58 MWh_{el} of electrical energy,^{33,34} making the electricity costs responsible for 67–77% of the production costs.³⁵ As a result, Germany, Europe's largest producer, currently devotes approximately 1.7% of its national electricity to chloralkali plants.³⁶

This high energy demand has notable implications for both operational costs and carbon footprints. On the other hand, powering the chloralkali electrolysis exclusively with renewable energy could significantly reduce carbon dioxide emissions. However, renewable energy, particularly from solar and wind energy sources, is inherently volatile depending on weather circumstances affecting the stability of the electricity grid. At the moment, chloralkali producers already contribute to the electricity grid stability by slightly adjusting the production to the availability of electricity.^{37,38}

However, as stated by Euro Chlor “when operating completely on wind and solar electricity, a completely new way of thinking is required, and multiple options need to be considered. These include storing electricity [...], increasing the flexibility of own production as well as that of downstream

users, and increasing the storage volumes of own and downstream produced products”.³⁷

To achieve flexibility by storing electricity is a versatile – and very popular – solution (see above). Nevertheless, this approach should be complemented by more specific solutions. Alternatively, the chlorine production by chloralkali electrolysis could be operated flexible harvesting renewable energy when they are available. The preferred process in chloralkali electrolysis, the membrane technology can be operated very flexibly within its technical limits. Consequently, it is characterised by high demand response potential and thus adaptable to fluctuating renewable energies.³⁹ In this context, the direct storage of chlorine could significantly streamline the adoption of chloralkali plants to renewable energies. When sun and wind are abundant, the chloralkali electrolysis could be ramped up producing surplus chlorine which would then be stored. On the other hand, when renewable energies are less available, the chlorine-consuming processes could be fed by previously stored chlorine.

However, the storage of chlorine, particularly on large scale, is inherently dangerous due to the high toxicity of chlorine.^{40,41} For example, in 2022 in the harbour of Aqaba, Jordan, 13 people died and more than 250 were injured when a vessel of pressure-liquefied chlorine ruptured.⁴² In fact, on average, one chlorine-related incident occurs for every million tonnes of chlorine produced in Europe each year.³² Thus, the transport of chlorine is strictly regulated in some countries, such as Switzerland.⁴³

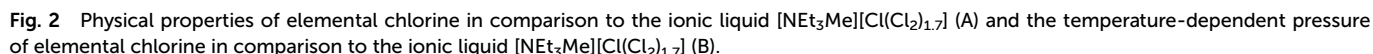
When chlorine is stored locally, stringent safety requirements have to be fulfilled. In Germany, for example, specific buffer zones around chlorine-related facilities are prescribed to ensure public safety. The size of the buffer zones depend on factors such as facility type, storage quantities, and local safety measures. In case of chlorine, a distance to areas in need of protection of 1.35 km is recommended.⁴⁴ Due to those restrictions, the storage of large quantities of chlorine is typically avoided. This limits also the demand response potentials considerably of the chloralkali electrolysis as only a few chlorine value chains such as PVC can cope with a fluctuating chlorine feed.³⁹ Therefore, to enable large-scale chlorine storage, any new approach must first address the inherent hazards of elemental chlorine.

Trichloride-based ionic liquids as indirect energy storages

Despite the broad applications of chlorine, its utilization is significantly limited due to high dangers associated with chlorine. It is a highly toxic, corrosive, and reactive, greenish-yellow gas with a boiling point of $-34\text{ }^\circ\text{C}$ (Fig. 2A). When stored and transported, it is typically pressure-liquefied forming liquid chlorine which has a vapor pressure of 6.7 bar at $20\text{ }^\circ\text{C}$.²³

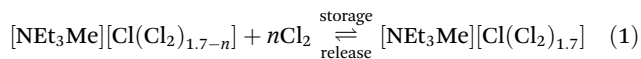
More recently, Riedel and coworkers developed trichloride-based ionic liquids (ILs) that enable the safe storage of chlorine while maintaining a low chlorine pressure even at elevated temperatures (Fig. 2B).^{30,36,45} Therefore, in the event of a storage leak, no chlorine gas but rather a liquid would escape





When $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_n]$ is used as a chlorine storage medium, the chlorine storage can best be described as a chemical equilibrium reaction (eqn (1)), which reaches at 25 °C and 1 bar the fully loaded system $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$.⁴⁵

For the release of chlorine, this equilibrium has to be shifted back to the starting materials. By heating the fully loaded system $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ to 80 °C, 0.5 equiv. of chlorine can be released, which leads to the partially loaded storage $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.2}]$. Notably, a complete chlorine release can be realized by applying vacuum or by the addition of water (see above).⁴⁵



To release 1.0 t of chlorine from $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ by heating to 80 °C a total amount of 7.677 t of $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ is required. In this scenario, the trichloride-based chlorine storage can indirectly store $0.34 \text{ MWh}_{\text{el}} \text{ t}_{[\text{Cl}_3]}^{-1}$.

Heating the storage

The energy necessary to release 1.0 t of chlorine from 7.677 t of $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ consists of two components: firstly the energy required to heat the system, and secondly, the reaction enthalpy for chlorine release.

The energy required to heat the $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ from 25 °C to 80 °C can be estimated using the specific heat capacities (c). The specific heat capacity of the storage medium changes during the chlorine release, but the specific heat capacities of the fully loaded and unloaded storage are known ($c([\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]) = 1.92 \text{ J g}^{-1} \text{ K}^{-1}$, $c([\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.2}]) = 1.60 \text{ J g}^{-1} \text{ K}^{-1}$). For the following estimation, a linear change of the specific heat capacity with the degree of chlorine release is assumed, resulting in an average specific heat capacity of $c^{\text{avg}} = 1.76 \text{ J g}^{-1} \text{ K}^{-1}$ for the system.⁴⁵

Additionally, the mass of the storage decreases from 7.677 t to 6.677 t, giving an average mass of the storage of 7.177 t.

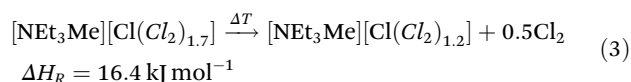
When the average heat capacity as well as the average mass of the system are considered, heating the system from 25 °C to 80 °C requires the following amount of energy (eqn (2)):

$$E = c^{\text{avg}} \times m^{\text{avg}} \times \Delta T = 6.95 \times 10^8 \text{ J} = 162 \text{ kWh} \quad (2)$$

Note that the ambient temperature can also lead to an increase of the chlorine pressure. While a fully loaded storage $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ exceeds a vapor pressure of 1 bar above 35 °C, at a reduced loading (e.g., 1.25 equivalents Cl_2) the pressure remains below 1 bar even at 50 °C. Thus, to avoid cooling the storage to stay below 1 bar, we propose limiting the chlorine loading if necessary.

Reaction enthalpy

The reaction enthalpy for the release of 0.5 equiv. of chlorine from the fully loaded storage was determined to be 16.4 kJ mol^{-1} (eqn (3)).⁴⁵



To release 1.0 t of chlorine, 7.677 t $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ (28 201 mol) are needed which gives a total reaction enthalpy of $4.62 \times 10^8 \text{ J} = 128 \text{ kWh}_{\text{th}}$. Therefore, the total amount of energy required to release 1.0 t of chlorine from $[\text{NEt}_3\text{Me}][\text{Cl}(\text{Cl}_2)_{1.7}]$ is approximately $(162 + 128) \text{ kWh} = 290 \text{ kWh}_{\text{th}}$. For comparison, this is the energy required for the production of 112 kg elemental chlorine ($2.58 \text{ kWh}_{\text{el}} \text{ kg}^{-1}$),^{32,33} or 5.8 kg of hydrogen ($50 \text{ kWh}_{\text{el}} \text{ kg}^{-1}$)⁴⁹ – however, it has to be outlined that temperatures of 80 °C might be available from exogenous processes within an industrial architecture, or could be

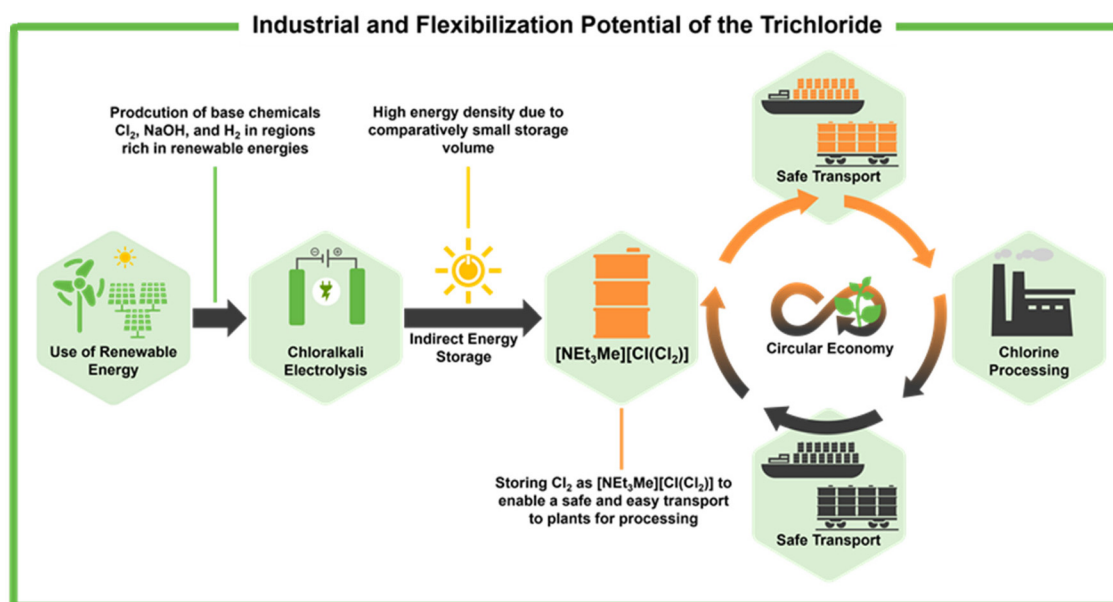


Fig. 3 Envisioned use of the trichloride-based chlorine storage as an energy shuttle enabling the safe transport of chlorine over large distances.



achieved at high efficiency levels with heat pumps and heat storage further improving the overall storage efficiency.

Direct usage of the loaded chlorine storage

Notably, it is known that trichlorides can be utilized as chlorination agents.^{30,50–52} The most prominent example is the solid reagent $[\text{NET}_3\text{Me}][\text{Cl}_3]$ that is a useful and easy-to-handle chlorination agent, for example, for the anti-addition of chlorine to alkenes and alkynes, the α -chlorination of ketones, or the chlorination of electron-rich arenes.⁵³

Therefore, instead of releasing and subsequently utilising the chlorine from the chlorine storage, the loaded chlorine storage $[\text{NET}_3\text{Me}][\text{Cl}_3]$ can directly be used for chlorination reactions, *e.g.*, the synthesis of the base chemical phosgene (COCl_2), showing an even enhanced reactivity compared to elemental chlorine.⁵⁰ In this way, the energy required for the release of chlorine could be saved and the storage medium could be used at far higher density. However, it is unlikely to replace elemental chlorine in all established chlorination processes.

Nevertheless, the biggest current disadvantage of storing chlorine on industrial scale, the danger associated with large quantities of pressure-liquefied chlorine, could essentially be overcome by using trichlorides as storage systems. In this way, the trichloride storage could contribute to the flexibilization of the chlorine production and consequently support the adoption to renewable energies.

Trichlorides as global energy shuttle

Trichloride-based Ionic Liquids could also be considered as a medium for international transport of chlorine. Utilising the trichloride-based Ionic Liquid, this would enable chlorine to be produced along with hydrogen and caustic soda *via* chlor-alkali electrolysis in regions rich in renewable energy, be stored as trichloride $[\text{NET}_3\text{Me}][\text{Cl}(\text{Cl}_2)]$, transported over long distances, and provided to chemical processes (Fig. 3). It could thus be considered as a partial substitute for hydrogen imports from comparable locations.⁵⁴ Further analysis is required to assess the economic viability of the required resilient transport infrastructure. A comparison would require an assessment compared to the flexible domestic production using available electricity in periods of large wind and solar production and the attractive storage properties of trichloride. Also, safety assessments in comparison to international ammoniac or hydrogen transport are required.

Conclusions

Chlorine is a crucial base chemical for the chemical industry, essential for the synthesis of numerous bulk polymers, intermediates, and pharmaceuticals. Its global production, which exceeds 100 Mt annually, relies almost entirely on chloralkali electrolysis, a process that requires 2.58 MWh_{el} of electricity per tonne of Cl_2 . Supplying this power from wind, solar, and other renewable energy sources could markedly reduce industrial CO_2 emissions. However, achieving this goal demands

that chloralkali electrolyzers operate flexibly in response to fluctuating renewable output.

While the chloralkali electrolysis can modulate load, most downstream plants need a constant chlorine stream. However, as pressure-liquefied Cl_2 is highly toxic and corrosive the storage of large quantities of chlorine is impracticable. Consequently, grid integration of chlor-alkali plants is presently limited by the absence of a safe, high-capacity chlorine reservoir. Recent work showed that triethylmethylammonium trichloride, $[\text{NET}_3\text{Me}][\text{Cl}(\text{Cl}_2)]$, could become a promising solution. This safe chlorine storage medium is an easy-to-handle room-temperature ionic liquid formed by reacting chlorine with the inexpensive salt $[\text{NET}_3\text{Me}]\text{Cl}$.

Chlorine can be released for the storage by heating, vacuum, or water addition; alternatively, the loaded liquid can serve directly as a chlorinating reagent. Its energy density approaches $1.1 \text{ MWh}_{\text{el}} \text{ t}^{-1}$, and thus outperforms even lithium-ion batteries on large scale. These ammonium trichlorides could decouple electrolysis from real-time demand, thus leveraging renewable surpluses to bridging periods of limited renewable output. To realize this vision further investigations and particularly chemical engineering endeavours like temperature- and concentration dependent viscosities, behaviour at very high temperatures, or storage tank designs are required, which are ongoing in our laboratories.

Conflicts of interest

The authors declare the following competing financial interests: R. W. and S. R. are inventors on two patents related to this work (no. WO 2019215037 A1, filed 2018 and no. WO2021069757 A1, filed 2021). P. V., R. W., and S. R. are inventors on two patents related to this work (no. WO2022128951 A1, filed 2022 and no. WO2022128950 A1, filed 2022). M.K., P. V., and S.R. are inventors on one patent related to this work (no. EP23206621, filed 2023). The authors declare that they have no other competing interests.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this perspective. All data shown has been cited within the perspective

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