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A review of Heat Integration Strategies in the Cu-Cl Thermochemical Cycle for Hydrogen Production

M. W. Abdulrahman[1], Y. A. Fenta[1], Y. Fonkamo[1]
[1]Rochester Institute of Technology-Dubai
Dubai, UAE
mwacad@rit.edu; yaf2642@rit.edu
ydf8711@g.rit.edu

Abstract **-** The copper–chlorine (Cu–Cl) thermochemical cycle is a promising candidate for large-scale hydrogen production due to its moderate operating temperatures and compatibility with nuclear and solar heat sources. However, its multi-step nature introduces significant thermal management challenges. This paper reviews internal heat integration strategies aimed at improving the energy efficiency of the Cu–Cl cycle. Emphasis is placed on the recovery and reuse of sensible heat from intermediate streams—such as CuCl, HCl, and product gases—within hydrolysis, thermolysis, and electrolysis stages. Integration approaches including direct heat exchange, heat exchanger networks, and thermal energy storage are evaluated based on their thermodynamic feasibility and system-level impact. The findings indicate that effective heat integration can reduce external thermal input requirements by up to 15%, contributing to improved sustainability and economic viability of hydrogen production via the Cu–Cl cycle.

Keywords**:** Cu-Cl cycle; hydrogen production; heat integration; thermochemical.

1. Introduction

The copper-chlorine (Cu-Cl) thermochemical cycle represents one of the most promising pathways for sustainable hydrogen production, particularly due to its relatively moderate temperature requirements (ranging from approximately 25°C to 530°C) and potential compatibility with nuclear and solar heat sources [1], [2], [3], [4], [5], [6], [7]. Its multi-step nature-typically comprising hydrolysis, thermolysis, electrolysis, and product recovery stages-makes it thermodynamically attractive but also inherently complex in terms of thermal management. Each reaction step involves unique temperature levels, reaction enthalpies, and phase behaviors, leading to significant potential for thermal energy loss if not carefully integrated [8], [9], [10], [11].

To reach commercially viable efficiency levels and reduce the levelized cost of hydrogen (LCOH), internal heat recovery and reuse must be maximized. Without proper heat integration, overall cycle efficiency can drop below 30%, while theoretical models suggest values exceeding 40% are attainable through effective heat management [1], [12]. This makes internal heat integration—the recovery and redistribution of thermal energy among the process streams—crucial for both economic and environmental viability.

The primary objective of this literature review is to examine and synthesize existing research on internal heat integration strategies applied to the Cu–Cl cycle, focusing on their thermodynamic performance, technical implementation, and system-level impacts. Rather than evaluating external energy sources (e.g., solar or nuclear heat), the review concentrates on strategies that reuse thermal energy between process steps within the cycle itself, thereby reducing dependency on external utilities and minimizing exergy destruction.

This review seeks to analyze the thermal and phase profiles of each reaction step in the Cu–Cl cycle to understand where heat is released or required, and how it can be redistributed efficiently. For example, steps such as hydrolysis and thermolysis are strongly endothermic, requiring significant thermal input to drive the decomposition and transformation of copper-based intermediates. In contrast, cooling and condensation stages offer opportunities to recover low- grade heat from product streams such as HCl and Cl_2 gases. These gases are typically released at temperatures between 100 and 150 °C, and their thermal energy can be captured using cascade exchangers or indirect loops to preheat incoming feed streams or support water vaporization [1], [12]. For example, studies involving spray reactor systems show that the post-hydrolysis HCl stream can be

quenched and its sensible heat utilized before electrolysis [13]. These recovery points, though individually modest in temperature, collectively contribute to significant exergy preservation when integrated effectively [14], [15].

The scope of this review is deliberately restricted to internal thermal integration, which refers to the recovery and redistribution of thermal energy within the Cu–Cl thermochemical cycle itself, where heat generated by one process step is redirected to meet the energy demands of another step without relying on external input sources. This approach emphasizes the strategic reuse of sensible heat, which is the thermal energy associated with a change in temperature of a substance without a phase change. For example, when molten CuCl exits the thermolysis reactor at ~530 °C and is later cooled before electrolysis, it retains a significant amount of sensible heat that can be transferred to colder streams such as the hydrolysis feed or water preheaters [16], [17], [18]. Sensible heat is recoverable through standard thermal equipment such as counterflow heat exchangers, and its magnitude is proportional to the product of the material's mass, specific heat capacity, and temperature difference [16], [19].

Internal integration strategies are central to improving overall system performance and include methods such as direct heat exchange (DHE), heat exchanger networks (HENs), thermal energy storage (TES), and cascade heat reuse. These techniques allow energy released from high-temperature stages-like thermolysis or product cooling-to be reused in subsequent lower-temperature stages such as hydrolysis or electrolysis, thereby reducing external utility demand and improving both energy and exergy efficiencies [14] [15], [19], [20].

By contrast, external heating technologies refer to thermal energy sources introduced from outside the system boundary, including solar concentrators, nuclear heat exchangers, or combustion-based heaters. While such technologies are essential to initialize or stabilize the Cu–Cl cycle—particularly for thermolysis which demands high-grade heat—they are only considered within this review when they directly facilitate internal recovery processes, such as serving as buffers for thermal energy storage losses or sustaining reaction temperatures during energy deficits [21] [19].

By narrowing the scope to internal heat integration, this review bridges the gap between theoretical thermodynamic optimization and practical implementation challenges, with a focus on how internal heat reuse can enhance the Cu–Cl cycle's efficiency, stability, and scalability in real-world hydrogen production systems.

2. Heat Flow in the Cu-Cl Thermochemical Cycle

The hybrid four-step copper–chlorine (Cu–Cl) thermochemical cycle is a closed-loop process that decomposes water into hydrogen and oxygen through a series of thermal and electrochemical reactions involving copper and chlorine intermediates. Effective internal heat integration depends critically on understanding the thermodynamic characteristics of each step, including temperature ranges, phase transitions, energy requirements (endothermic vs. exothermic), and potential for heat recovery. This section provides a detailed breakdown of the heat flow through the full 4-step Cu–Cl cycle, highlighting areas for integration and exergy optimization.

2.1. Reaction Steps and Heat Characteristics

2.1.1. Step 1: Hydrolysis

Chemical Reaction:

$$2CuCl_{2(aq)} + 2H_2O_{(g)} \rightarrow Cu_2OCl_{2(s)} + 2HCl_{(g)}$$

Operating Conditions:

The hydrolysis reaction is typically conducted within the temperature range of 375–410 °C, but multiple sources identify a more precise optimum temperature near ~395–400 °C. Ferrandon et al. (2010), through spray reactor experimentation, found that HCl yield was maximized around 390–400 °C, with conversion efficiency decreasing below 375 °C and side-product formation (CuCl) increasing above 410 °C [13] [14], [15], [22], [23], [24], [25]. Tang et al. (2024) further supported this range by identifying ~395 °C as the point of maximum Cu_2OCl_2 formation under a $CuCl_2$:H_2O molar ratio of 1:20 [26].

The reaction is generally carried out at or near atmospheric pressure, using superheated steam and excess H_2O to drive the equilibrium forward. $CuCl_2$ is usually introduced in an aqueous or atomized form, and steam is injected at high temperature to initiate the endothermic transformation. The outlet gas stream (HCl) exits at temperatures close to 380–400 °C, while solid Cu_2OCl_2 is retained in the reactor for transfer to the thermolysis step.

Thermal Energy Requirements:

The total heat demand includes the sensible heat for $CuCl_2$. While $CuCl_2$ is regenerated and recycled within the Cu–Cl cycle, its entry temperature into the hydrolysis reactor is system-dependent. In experimental setups [13], it often enters near room temperature (~25 °C), but in integrated systems, it may be preheated to 100–200 °C via internal heat recovery [14] [14], [15], [24], [27]. The value of the sensible heat required is calculated from:

$$Q = n\, C_p\, \Delta\, T$$

where:
Q = heat required (in joules or kJ)
n = number of moles of $CuCl_2$
C_p = specific heat capacity of $CuCl_2$
ΔT = temperature change

For conservative analysis, assuming a starting temperature of 25 °C, the total heat of $CuCl_2$ is 62.5 kJ. In advanced configurations, this value could drop to ~30 kJ if preheated to 200°C. The sensible heat for H_2O (25°C – 100°C) is 226 kJ and the latent heat of vaporization at 100°C is 1626 kJ. The heat of superheating (100°C – 400°C) is 438 kJ. In this case, the total heat of water will be 2290 kJ. The reaction enthalpy of hydrolysis is 316 kJ [26]. In this case, the grand total heat requirement for hydrolysis reaction is 2668.5 KJ per 2 mol of $CuCl_2$ [28].

Integration Potential:

The downstream thermolysis step operates at 500–530 °C and produces hot molten CuCl and O_2 gas [29], [30], [31], [32]. These carry significant sensible heat, which can be internally recovered. The Molten CuCl recovery is ~134–150 kJ/mol H_2. These covers ~10–12% of the hydrolysis step's thermal input. Integration possible via direct exchange (DHE) or indirect molten salt loops [14], [16], [20].

2.1.2. Step 2: Thermolysis

Chemical Reaction:

$$2Cu_2OCl_{2(s)} \rightarrow O_{2(g)} + 4CuCl_{(l)}$$

Operating Conditions:

The thermolysis step is conducted at high temperatures ranging from 490 to 535 °C, with optimal decomposition of Cu_2OCl_2 occurring at approximately ~505 °C. Wajda & Gabriel (2018), in a pilot-scale reactor study, identified ~505 °C as the temperature at which maximum conversion to molten CuCl and O_2 gas was achieved with stable phase transitions and no significant side reactions [3], [33], [34], [35]. Similarly, Thomas et al. (2021) reported that decomposition efficiency peaked between 500–510 °C, particularly under longer residence times in reactor cores [36]. Operating below 490 °C results in incomplete decomposition of Cu_2OCl_2, reducing the hydrogen production rate in downstream steps. Temperatures above 530–540 °C may lead to excessive energy input, potential CuCl volatilization, and thermal degradation of reactor internals. The reaction is typically carried out at atmospheric pressure in a high- temperature reactor designed to handle solid-to-liquid phase transitions and maintain stable residence time for complete reaction. The products, molten CuCl and hot O_2 gas, exit the reactor at near reaction temperature (~500–530 °C) and carry significant sensible heat, which offers valuable opportunities for internal heat recovery [37], [38], [39], [40], [41], [42].

Thermal Energy Requirements:

Reaction Enthalpy: Thermolysis is a strongly endothermic step with a reaction enthalpy of ~286 kJ/mol Cu_2OCl_2 [33], [43]. For the decomposition of 2 mol Cu_2OCl_2 (the output of one hydrolysis batch), the thermal input becomes 572 kJ.

Sensible Heat in Products: After decomposition, the products molten CuCl and O_2 gas exit the reactor at high temperatures (~500–530 °C), retaining significant sensible heat that can be internally recovered before cooling.

- CuCl Sensible Heat: The specific heat capacity of molten CuCl is approximately 54.4 J/mol·K across 450–600 °C. With 4 mol CuCl produced and assuming the ambient room temperature to be 25 °C, the sensible heat of the CuCl is 109.9 kJ [19].
- O_2 Sensible Heat: Given a molar heat capacity of 29.4 J/mol K, the sensible heat for the same temperature difference per 2 mol of Cu_2OCl_2 is 14.8 kJ.

Total recoverable sensible heat from products amounts to be 124.7 kJ.

Integration Opportunities:

The sensible heat is highly valuable for internal reuse. It can be used to preheat $CuCl_2$ and water before hydrolysis, can partially or fully vaporize water needed for HCl production, and can reduce heating duty on TES or direct heat exchangers. More advanced recovery mechanisms such as TEG-based indirect systems and heat exchanger networks (HENs) have also been proposed to extend this benefit [16], [19], [44].

2.1.3. Step 3: CuCl Electrolysis

Chemical Reaction:

$$2CuCl_{(aq)} \rightarrow 2Cu_{(s)} + Cl_{2(g)}$$

Operating Conditions:

The CuCl electrolysis step is conducted at low temperatures, typically between 20 and 80 °C, with most designs operating optimally around 60–70 °C [45]. While early pilot studies explored operation over a broad range, more recent modeling and techno- economic evaluations suggest that the optimal performance point lies around 60 °C, where ionic conductivity, copper deposition rates, and current efficiency are balanced with minimal parasitic heat loss. Operating at lower temperatures (e.g., <40 °C) can significantly reduce electrode kinetics and increase solution resistance, while higher temperatures (>75 °C) may increase material degradation risks and affect electrolyte stability. The process occurs in an aqueous electrochemical cell, where CuCl is oxidized at the anode to produce Cl_2 gas, and Cu^+ ions are reduced at the cathode to deposit solid copper. The electrolysis unit is operated under near-atmospheric pressure in a divided cell configuration to prevent recombination of Cu^+ and Cl_2. To maintain performance, the temperature must be tightly controlled, often using water cooling or internal circulation loops. Integration with upstream cooling from molten CuCl (post-thermolysis) offers an opportunity to preheat other cycle streams and recover low-grade heat [15], [44], [45].

Thermal Energy Requirements:

This step requires external DC electrical power to split CuCl. The operating voltage is typically 1.0–1.3 V. The specific energy demand is 200–250 kJ/mol H_2 [14]. The process operates at low temperature and generates low-grade waste heat. Prior to entering the electrolysis cell, CuCl must be cooled from its molten state (~530 °C) to the electrolysis temperature (~60 °C). This cooling releases recoverable sensible heat of 51.1 kJ.

Integration Potential:

The sensible heat released during CuCl cooling presents strong potential for integration. It can be used for steam preheating for hydrolysis or electrolysis, feedwater heating, and TES charging in indirect loops [14], [15], [46].

2.1.4. Step 4: HCl Electrolysis

Chemical Reaction:

$$2HCl_{(g)} \rightarrow H_{2(g)} + Cl_{2(g)}$$

Operating Conditions:

The HCl electrolysis step is performed at relatively low temperatures, typically around 80–110 °C, with ~100 °C identified as the optimal value for membrane conductivity, gas-phase ion transport, and system efficiency. Operating below 80 °C can lead to increased membrane resistance and reduced gas production rates, while temperatures above 110 °C may risk membrane dehydration and corrosion of cell components. The process is conducted in a solid polymer or membrane-based electrolyzer, where gaseous HCl is dissociated into hydrogen (H_2) at the cathode and chlorine (Cl_2) at the anode. The system requires strict moisture and temperature control to maintain membrane performance and prevent product crossover [44], [45], [47].

Thermal Energy Requirements:

HCl electrolysis is an endothermic electrochemical process requiring external electrical power. Literature values for voltage and energy input include: operating voltage which is typically 1.7–2.2 V, and specific energy demand: 290–330 kJ/mol H_2. This energy is significantly higher than the CuCl electrolysis step due to the strong H-Cl bond energy and the need to maintain membrane conductivity [14].

In the Sensible heat management, the hot HCl gas, produced during hydrolysis at ~375–400 °C, must be cooled to ~100 °C before entering the electrolyzer. This temperature drop represents a significant low- to medium-grade heat recovery opportunity (17.46 kJ)

Integration Potential:

Cooling of HCl gas offers an opportunity to recover ~17–18 kJ per reaction cycle. The product gases (H_2 and Cl_2), though exiting at ~100 °C, carry additional low-grade sensible heat that can be captured for use in feedwater preheating or electrochemical temperature maintenance. These recovery strategies support system-wide thermal integration, particularly when coupled with heat exchanger networks or indirect loops [15], [46], [48].

2.2. Key Heat Recovery Points

Efficient thermal integration in the Cu–Cl cycle depends heavily on identifying high- grade, medium-grade, and low-grade waste heat sources that can be internally reused. Each of the four steps presents specific opportunities for sensible and latent heat recovery, which can reduce the need for external thermal input and increase system efficiency.

2.2.1. Heat Recovery from Molten CuCl (Post-Thermolysis → Pre-Electrolysis)

Following thermolysis at ~530 °C, molten CuCl must be cooled to ~60 °C before entering the aqueous CuCl electrolysis step. This ~470 K temperature drop presents a substantial opportunity to recover medium-grade sensible heat within the Cu–Cl cycle. Using the molar heat capacity of molten CuCl, 54.4 J/mol K [16], [49], the sensible heat released by 2 mol of CuCl cooled from 530 °C to 60 °C is 51.1 kJ.

For 4 mol of CuCl (the amount produced per 2 mol of Cu_2OCl_2 during thermolysis), the recoverable heat becomes 102.3 KJ. This ~102 kJ of medium-grade heat is valuable and should not be wasted. If left unrecovered, this energy would dissipate as thermal losses to cooling systems, reducing the overall efficiency of the cycle.

Recovery Mechanisms:

According to Suppiah et al. (2004) [50], molten CuCl cooling is typically facilitated by thermal oil circuits or indirect recuperative exchangers, which isolate corrosive streams while extracting useful heat [16]. The recovered heat can be reused to[50]:

- Preheat $CuCl_2$ and water entering the hydrolysis reactor (~375–400 °C), reducing external energy demand.
- Support steam generation at 120–150 °C, feeding HCl electrolysis or hybrid water–vapor systems.
- Charge thermal energy storage (TES) modules that act as buffers during load fluctuations.
- Drive low-grade auxiliary systems, such as membrane drying or desorption units in gas purification [19].

System Level Impact:

While the raw heat recovery potential from molten CuCl is ~102 kJ per batch, integration strategies (such as pinch analysis or heat exchanger networks) can allow for its effective reuse within the cycle, minimizing exergy loss. For example, Kadam & Yadav (2023) [14] demonstrate that internal preheating and cascade reuse of such heat streams could improve overall thermal efficiency by up to 6–8%, depending on the exchanger effectiveness and matching temperature gradients.

2.2.2. Cooling of Hot HCl Gas (Post-Hydrolysis → Pre-HCl Electrolysis)

During the hydrolysis step, HCl gas is produced at high temperatures (~375–400 °C) and must be cooled to ~100 °C before it enters the HCl electrolyzer. This 300 K temperature drop provides a valuable opportunity to recover low- to medium-grade sensible heat, especially given the clean, single-phase nature of the gas stream.

Using the molar heat capacity of HCl gas (~29.1 J/mol·K) (Farsi et al., 2020) and assuming 2 mol of HCl produced per reaction cycle, the amount of heat is 17.46 kJ. Although modest in magnitude compared to molten CuCl recovery (~102 kJ), this thermal stream is continuously available and can be easily harnessed using compact gas-gas exchangers.

Materials and heat Exchanger Designs:

The HCl gas stream is highly corrosive, especially at elevated temperatures and moist conditions. Ehyaei et al. (2024) recommend specialized exchanger materials. They Emphasized using Hastelloy, Teflon-lined piping, or PTFE- coated plate exchangers [48].

Integration Opportunities:

Recovered heat from hot HCl can be reused in several parts of the Cu–Cl cycle [14], [51]:

- Preheating feedwater or $CuCl_2$ solution before hydrolysis, saving on external thermal duty.
- Low-pressure steam generation to support water splitting, TES buffer charging, or auxiliary thermal loops.
- Drying or desorption processes, particularly where electricity is limited and heat is needed at <150 °C.
- Charging small TES reservoirs, especially in intermittent solar-nuclear systems.

System-wide Relevance:

Though each cycle yields ~17 kJ of recoverable heat from HCl gas, the cumulative effect across continuous operation is non-negligible. When paired with molten CuCl cooling (102 kJ), the combined post-thermolysis/post-hydrolysis thermal recovery can reach ~120 kJ per cycle-enough to significantly lower preheating loads for electrolysis and steam. Sadeghi et al. (2023) report that recovering this low-grade heat through pinch-integrated HENs can reduce external thermal input by 4-6%, particularly when integrated with indirect TES or smart heat sink routing [16], [19], [20], [44].

2.2.3. Heat Recovery from Product Gases (H_2 and O_2)

In the Cu–Cl thermochemical cycle, hydrogen (H_2) is generated exclusively in the HCl electrolysis step, while oxygen (O_2) is produced in the high-temperature thermolysis reactor. Both exit their respective units at elevated temperatures and require cooling, offering an opportunity to recover low- to medium-grade sensible heat that would otherwise be lost to ambient dissipation. This section quantifies the recoverable heat from these gases and outlines their integration pathways.

Hydrogen – Product of HCl Electrolysis:

The HCl electrolysis step operates typically at ~100 °C, producing 1 mol of H_2 per reaction cycle. Before purification, compression, or storage, the hydrogen gas must be cooled to ambient temperature (~25 °C). This cooling process releases recoverable low- grade sensible heat. Taking the molar heat capacity of hydrogen gas to be ~28.4 J/mol.K, the heat is 2.13 kJ.

Although small in absolute value, this heat is clean, non-corrosive, and can be efficiently captured using gas–liquid heat exchangers or plate-type microexchangers, especially in hybridized or closed-loop systems.

Oxygen – Product of Thermolysis:

During thermolysis, 1 mol of O_2 is generated per reaction batch at approximately 530 °C, and is typically cooled to ~25 °C before venting or reuse. This represents a 505 K drop, providing medium-grade sensible heat suitable for internal recovery. Taking the molar heat capacity of oxygen gas to be 29.4 J/mol.K, the amount of heat is 14.8 kJ. This heat can be redirected to support feedwater preheating, TES charging, or auxiliary heating loops for non-critical thermal loads. In large-scale systems, it may also help maintain temperature uniformity in intermediate storage tanks or reactor wall jackets [12], [15], [42], [43].

Integration Relevance and System Implication:

While lower than the ~102 kJ recoverable from molten CuCl, this ~17 kJ cumulative recoverable heat per cycle of gas-phase sensible heat is:

- Thermodynamically clean (no phase change or corrosion issues).
- Available continuously in high-throughput operations.
- Easy to integrate into HENs or TES systems for secondary energy recovery.

3. Exergy Considerations

In heat integration planning, it is not sufficient to track energy quantities alone; the quality of that energy-its exergy-determines its true usefulness. Exergy quantifies the maximum theoretical work that can be extracted from a heat stream as it comes into equilibrium with its surroundings. High-temperature streams carry more exergy, and using them improperly leads to irreversible losses due to entropy generation [10]. High-grade thermal energy from the thermolysis step (~500°C) carries substantial exergy. If this energy is used to heat a stream with a large ΔT (e.g., from 25°C to 80°C), most of the exergy is destroyed in the process, even if the total energy is transferred. The exergy E of heat Q at a temperature T is given by:

$$E = Q\left(1 - \frac{T_0}{T}\right) \quad (1)$$

where T_0 is the ambient temperature. For an ambient temperature of 25 °C (~298 K), the exergy factor for a heat at 500 °C (~773 K) is approximately 0.61. At 100 °C (373 K) however, the exergy factor drops to approximately 0.20. This highlights that only a fraction of the thermal energy at lower temperatures remains exergically useful, mandating the thorough analysis of exergy throughout the cycle.

3. Conclusion

This review highlights the critical role of internal heat integration in enhancing the thermodynamic and operational efficiency of the Cu–Cl cycle. Strategic recovery and redistribution of sensible heat, particularly from high-temperature thermolysis outputs, can significantly reduce external energy demands and improve overall system performance. Techniques such as DHEs and HENs offer practical routes for implementing integration, especially when coupled with effective temperature matching and material compatibility. As hydrogen production systems move toward industrial deployment, future research should focus on pilot-scale implementation, corrosion-resistant exchanger materials, and integration with renewable heat sources to further advance the Cu–Cl cycle's commercial potential.

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