Chlorine

HIGHLIGHTS

Processes and technology status – Commercial production of chlorine is via a continuous day-and-night electrolysis process that uses electricity as a raw material [1]. Most chlorine is manufactured by the electrolysis of sodium chloride solutions. The primary raw material for this process is rock salt (sodium chloride, NaCl), available worldwide usually in the form of underground deposits of high purity [2].

Cost – Chlor-alkali production relies on energy-intensive electrochemical technology, hence, the price of electricity represents roughly 40% of the operating cash cost. Membrane technology requires less electrical energy and lowers operating cash costs by an average of about 6 % [3].

Potential and barriers – The barriers ahead of chlorine production are the high cost and energy requirements of the process [4]. Therefore, coupling the Chlor-alkali process with other electrocatalytic processes holds great potential for future energy conservation and storage [4]. Relying intensively on electricity, the Chlor-alkali process can play an important role in the enhancement of grid resilience as well.

Chlorine – Chlorine (Cl_2), as a chemical precursor, is crucial for many important industrial processes including polymer synthesis, disinfection goods production, pharmaceutical manufactures, and wastewater treatment [4], [5]. The Chlor-alkali industry underpins about 55% of the European chemicals and pharmaceuticals industry which realized in 2009 a turnover of almost 660 billion euro [6].

The Chlor-alkali industry has three distinct products with separate applications and industrial utilizations [7]. Chlor-alkali produces the base chemicals chlorine (Cl_2) and sodium hydroxide (NaOH), as well as hydrogen (H_2) as a byproduct in the conventional process [8]. Figure 1 shows the applications for the chlorine produced in Europe [7]. About two-thirds of European chlorine production is used in engineering materials such as polymers, resins, and elastomers. The largest single end-use (35%) is PVC plastic for the construction, automotive, electronics, and electrical industries. The manufacturing processes of

Provided by Talieh Rajabloo
talieh.rajabloo@uhasselt.be

many chemicals, plastics, and medicines use chlorine. However, such as the plastics polyurethane and polycarbonate the end product is chlorine-free [6].

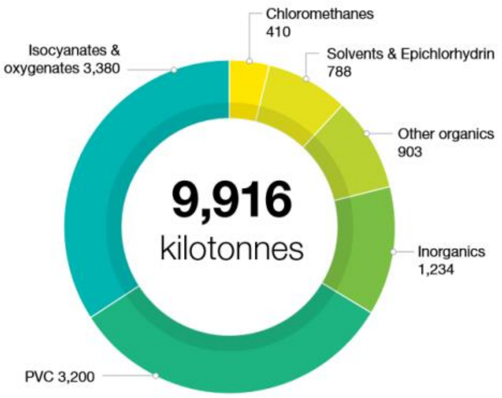

Figure 1. European chlorine in applications (EuroChlor, 2019), numbers are in Kilotones [7].

Process overview - Key feedstocks for chlorine production are Salt and Electricity [9]. Chlorine is produced through the Chlor-alkali process. In this process, electricity is applied to a solution of salt water or brine. The electricity separates sodium from chloride. Chlorine gas, hydrogen gas, and caustic soda (sodium hydroxide) solutions are the co-products of the Chlor-alkali process. Chlorine and caustic soda are building block compounds for thousands of useful products. Hydrogen can be recycled into the Chlor-alkali process as a zero-emission fuel [10], [11].

Chlorine is, also, produced in several other ways, for example, by electrolysis of molten salts and by non-electrolytic processes [12]. The electrolysis of sodium, magnesium, and calcium chloride (NaCl, $MgCl_2$, and $CaCl_2$) can generate elemental sodium, magnesium, or calcium metal besides the chlorine gas [12]. Figure 2 depicts the electrolysis of a molten mixture of NaCl and $CaCl_2$ resulting in the formation of elemental sodium and chlorine gas [13].

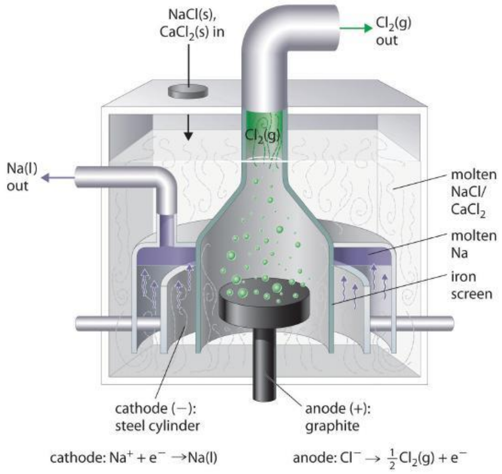

Figure 2. Downs Cell for the Electrolysis of Molten NaCl [13].

Most chlorine is manufactured electrochemically by the diaphragm, membrane, or mercury cell process. In each process, a salt solution (sodium, potassium, magnesium, or calcium chloride) is electrolyzed by the action of a direct electric current which converts chloride ions to elemental chlorine [12]. The two oldest methods, diaphragm and mercury cells have been used for over 100 years throughout the world and have been proven to be the most environmentally unfriendly

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talieh.rajabloo@uhasselt.be

through their use of asbestos and mercury, respectively [14].

Diaphragm Cell Technology – The most chlorine production in North America was from diaphragm cell technology in 2020. The products of this type of cell are chlorine gas, hydrogen gas, and cell liquor composed of sodium hydroxide and sodium chloride solution [12]. A nearly saturated sodium chloride solution (brine) enters the diaphragm cell anolyte compartment and flows through the diaphragm to the cathode section. Chloride ions are oxidized at the anode to produce chlorine gas. Hydrogen gas and hydroxide ions are produced at the cathode. Sodium ions migrate across the diaphragm from the anode compartment to the cathode side to produce cell liquor containing 10% to 12% sodium hydroxide. Some chloride ions also migrate across the diaphragm resulting in the cell liquor containing about 16% sodium chloride. The cell liquor is typically concentrated to 50% sodium hydroxide by an evaporation process. The salt recovered in the evaporation process is returned to the brine system for reuse [12]. Figure 3 shows an industrial implementation of a Chlor-alkali cell line with MDC-55 diaphragm cells [15].

Figure 3. Chlor-alkali cell line with MDC-55 diaphragm cells [15]

Mercury Cell Technology – Mercury Cell technology uses a stream of mercury flowing along the bottom of the electrolyzer as the cathode. The anodes are suspended parallel to the base of the cell, a few millimeters above the flowing mercury. Brine is fed into one end of the cell box and flows by gravity between the anodes and the cathode. Chlorine gas is evolved and released at the anode. The sodium ions are deposited along the surface of the flowing mercury cathode. The alkali metal dissolves in the mercury, forming a liquid amalgam. The amalgam flows by gravity from the electrolyzer to the carbon-filled decomposer, where deionized water is added. The water chemically strips the alkali metal from the mercury, producing hydrogen and 50% sodium hydroxide. The mercury is then pumped back to the cell inlet, where the electrolysis process is repeated [12].

In the Mercury cell technology, approximately 100 cells operate in series. Purified, saturated brine (25% (w/w) sodium chloride solution) at typically 333 K flows through the cell in the same direction as the mercury. This high salt concentration and the anode coating ensure the oxidation of chloride ions rather than that of water which would yield oxygen at the titanium anodes. The schematic of the mercury cell is shown in figure 4 [2].

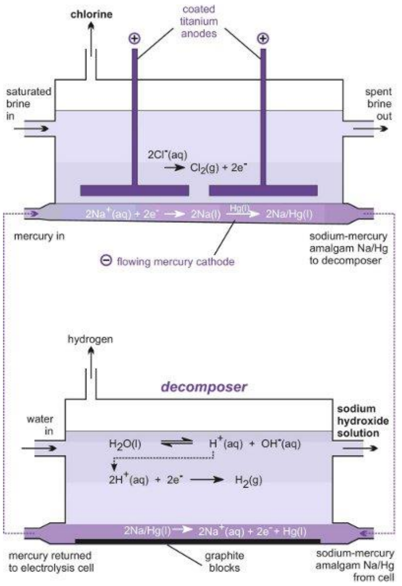

Figure 4. The mercury cell and decomposer [2]

Membrane Cell Technology – Overall, the technology shift from mercury cells to the membrane has led to lower electricity consumption in the sector. A membrane plant uses less electricity than a mercury plant but requires more steam to obtain the standard commercial concentration of caustic soda [6], [3]. Typically, a membrane plant may require about 15% fewer personnel than a mercury cell plant. Also, manpower for maintenance and costs of materials and spare parts for a membrane cell plant will normally be lower [3].

Membrane cell technology uses sheets of perfluorinated polymer ion exchange membranes to separate the anodes and cathodes within the electrolyzer. Ultra-pure brine is fed to the anode compartments, where chloride ions are oxidized to form chlorine gas. The membranes are cation-selective resulting in predominantly sodium ions and water migrating across the membranes to the cathode compartments. Water is reduced to form hydrogen gas and hydroxide ions at the cathodes. In the cathode compartment, hydroxide ions and sodium ions combine to form sodium hydroxide [12]. Membrane electrolyzers typically produce 30% to 35% sodium hydroxide, containing less than 100 ppm of sodium chloride. The sodium hydroxide can be concentrated further, typically to 50%, using evaporators [12]. Figure 5 shows an industrial Chlor-alkali cell line with membrane cells [15]. The power capacity of the 50 MW is the average capacity of industrial Chlor-alkali electrolysis processes employing membrane cells in Europe. This results in the chlorine

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talieh.rajabloo@uhasselt.be

production of 21.9 t/h for all the processes [8].

Figure 5. Chlor-alkali cell line with membrane cells [15]

The fast dynamics of Chlor-alkali electrolysis make chlorine production suitable for varying the operational level to the three different overcapacities, 2.5, 5, and 10 MW, that correspond to 5, 10, and 20% of the nominal capacity [8]. All three types of cells generate chlorine by electrolytic oxidation at the anode. Their differences lie in the cathode reactions, energy consumption, and environmental effects [5]. $2Cl^- \rightarrow Cl_2 + 2e^-$

Cost and energy – The cost of the Chlor-alkali industry is energy- and resource-dependent. Using electrical laws, the required amount of electrical energy for an electrolyzer is 5.95 PJ/Mt Cl_2. Due to resistance across the induced current between the anode and the cathode an additional voltage is needed. Usually, for the production of chlorine, a voltage of 2.5 to 3.5 V is required which leads to resistance-based heat production which increases electricity requirement by an additional 0.85-3.56 PJ/Mt_{Cl2}. The smaller the distance between the anode and the cathode, the smaller the resistance.

Roughly 40% of total production costs are spent on electricity and gas [7]. Natural gas fuels heat engines to produce the low-pressure steam for the processing of caustic soda. The steam is produced at a temperature of about 150 to 180°C and a pressure of 3 to 4 bar through on-site and off-site combined heat and power (CHP) plants [7]. A high concentration of 50% caustic soda is generated through multiple-effect evaporation of the dilute caustic soda (32%) that is drained from the electrolytic cells to meet market demands. This process requires a considerable amount of heat energy (1.5 PJ/Mt chlorine) and accounts for approximately 80% of the total steam demand for the Chlor-alkali manufacturing industry [7]. Besides, the industrial vacuum salt costs 57 €$_{2014}$/t, and the cost of required electricity is 237.6 [1] €$_{2017}$/kJ [7]. More detailed costs of the process for Dutch industries are available in Table 1 which are related to the years 2014 and 2017 [7].

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[1] Converted from 0.066 €$_{2017}$/kWh

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talieh.rajabloo@uhasselt.be

Table 1. Financial overview of the Dutch chlor-alkali manufacturing industry, Eurostat (2020)[7]

Chlor-alkali manufacturing financial overview			
Revenue	**% of total**	**Assumption unit market price**	**Source**
Chlorine	38%	chlorine: 200 EUR/ton	Brinkmann et al., 2014
Caustic soda	53%	caustic soda 370 EUR/ton	Brinkmann et al., 2014
hydrogen	9%	hydrogen 1600 EUR/ton	Brinkmann et al., 2014
Costs	**% of total**	**Assumption unit price**	**Source**
Electricity	37%	0.066 EUR/kWh	Schoots et al., 2017
Salt	16%	Industrial vacuum salt: 57 EUR/ton	Brinkmann et al., 2014
Gas	4%	6.8 MEUR/PJ	Schoots et al., 2017
Membranes	4%	20 EUR/ECU^2	Schmittinger et al., 2012

The chlorine gas captured upon electrolysis, just like the hydrogen gas, is also mixed with water vapor due to the high temperatures in the electrolyzers (approximately 20% steam, 80% chlorine gas), and the captured water is fed back to the de-chlorination unit[7]. About 10% of the electricity is used for lighting and operating pumps, compressors, and other necessary equipment. 90% of the electric current is used as raw material which cannot be substituted[6]. For the production of 850 kt/y chlorine, 1600 kt/y of industrial quality salt (98 %) and 2400 kt/y purified water, besides 0.18 kt/y of HCl (32%) are required.

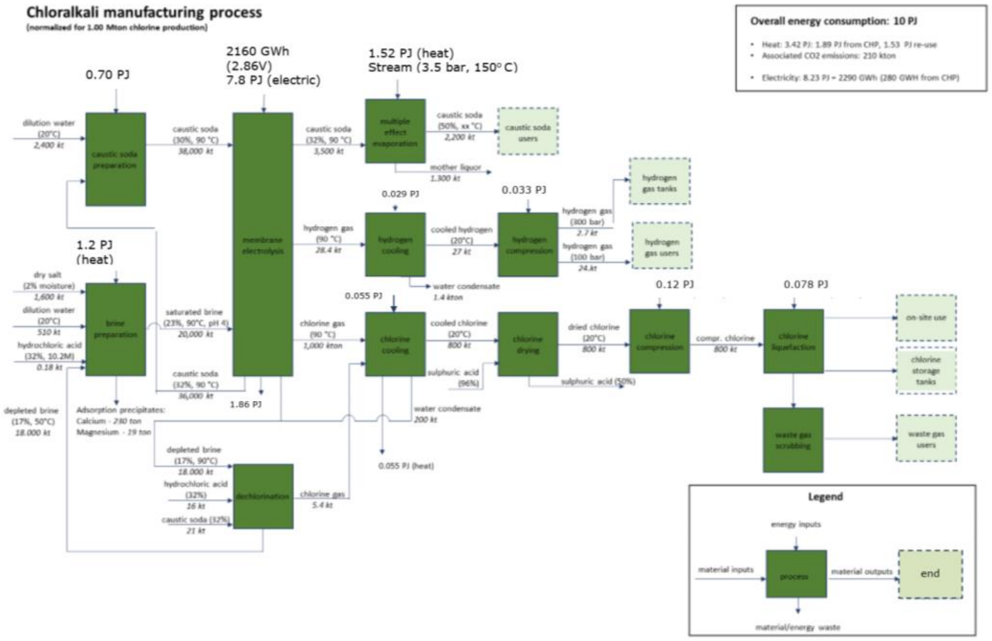

Figure 6. Chlor-alkali products and use; normalized for 1 Mt of chlorine[7].

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talieh.rajabloo@uhasselt.be

This process utilizes 2.8 PJ/y steam and 6.1 PJ/y electricity and generates 950 kt/y soda (50%), 24 kt/y hydrogen as a byproduct, and emits 180 kt/y CO_2 [7]. In the current technologies steam is generated from natural gas-fired CHP. Moreover, the emission-related to the electricity (6.1 PJ/y) is equal to 760 kt indirect CO_2 [7].

Production and consumption in Belgium – The production capacity of chlorine in Belgium in 2020 was 1074 kilotons [16]. INOVYN at Antwerp/Zandvliet Site has a world-class membrane electrolysis unit and delivers chlorine via pipeline to large industrial customers in the Antwerp chemical cluster. Most of the chlorine produced at the Site is used by BASF for the production of isocyanates. BASF returns the major part of the chlorine as hydrogen chloride gas back to INOVYN at Antwerp/Zandvliet Site, where it is transformed into ethylene dichloride [17].

Table 2. Chlorine production plants in Belgium, 1st January 2020 capacities [18]:

Company	Site	Produced Cl_2 (kt) [18]
INOVYN	Lillo	500
INOVYN	Jemeppe*	174
Vynova	Tessenderlo	400

* Plant Name: Inovyn - Jemeppe-sur-Sambre; Owner: Ineos, via Inovyn subsidiary, formerly Solvic. Ineos acquired Inovyn in July 2016. Location: Jemeppe-sur-Sambre, Belgium. Process: Membrane. Converted from mercury in 2001. Year Opened: 1897. Capacities (tons per year): 174,000 tons of chlorine (January 2017) and other chemicals [19].

Vynova site located in Tessenderlo, Belgium has two membrane electrolysis plants, one for caustic soda production and another for potassium hydroxide production [20]. In Tessenderlo, chlorine is converted to ethylene dichloride (EDC) and vinyl chloride monomer (VCM), two essential building blocks for the production of PVC [20]. Vynova has replaced the previous mercury cells at the Tessenderlo site with the membrane electrolysis technology. In membrane cells, the anode and cathode are separated by a membrane, which allows sodium or potassium ions to pass through, but prevents the flow of chlorine or hydrogen ions [21].

By the time membrane cell technology became accessible as an industrial solution, there was already a significant precedent established for the use of mercury cell chlorine plants. As such, converting from mercury cells to membrane cells was an expensive capital undertaking. Although there is a return on investment through energy savings, this return is not usually sufficient to justify the capital expense on its own. The rate of return for such projects is typically about 20 years. Nevertheless, these projects are necessary and reflect responsible practice: conversion from mercury cells to membrane cells is primarily driven by occupational health and environmental concerns [21]. Vynova has demonstrated industry leadership through an aggressive capital investment program for conversion to membrane cells across all its facilities [21].

Potential and barriers ahead of chlorine production – The barriers ahead of chlorine production are related to the high cost and energy requirements of the process. Starting from this point, the potentials for process improvements can emerge. For example, the dimensionally stable anode (DSA) technology can make the Chlor-alkali process more sustainable in the future, and, cost reduction of DSA opens new opportunities for sustainable and more affordable chlorine production [4]. Moreover, coupling the Chlor-alkali process with other electrocatalytic processes holds great potential for future energy conservation and storage [4]. In addition, chlorine production from seawater electrolysis is a promising strategy that can be achieved through the manufacturing of multi-component metal oxides [4]. Relying intensively on electricity, the Chlor-alkali process can play an important role in the enhancement of grid resilience as well. Flexible operation of the switchable Chlor-alkali process is a novel strategy for minimizing electricity costs in markets with volatile prices and to assist demand-side management. This strategy allows for adjusting the process to electricity price profiles e.g. by varying the production rate or the operation modes [22], [23], [24].

Emission from Chlor-alkali processes - The Chlor-alkali industry emits indirect CO_2 during the production of the electricity that the sector uses. The process itself does not emit direct CO_2. The average electricity consumption per ton of chlorine leads to 2.1 tons of $CO_{2\text{-eq}}$ [6].

Membrane cells with oxygen-depolarized cathodes (ODC), promises much greater energy savings and emissions reductions [5]. Table 3 compares the energy requirements of each cell type and the carbon dioxide emissions associated with the production energy [5].

Table 3: Average energy consumption and emissions for brine cells [5]

Cell type		Mercury	Diaphragm	Membrane	Membrane with ODC
Operating voltage/V		3.9-4.2	2.9-3.5	3.0-3.6	≈ 2
Energy consumption (MWh/t_{Cl2}) [2]	Electrolysis	3.4	2.7	2.6	1.8t
	Evaporation	0	0.6	0.2	0.2
CO_2 emissions/t		1.8	1.7	1.5	1.0

Yearly estimated CO_2 emissions related to Belgian chlorine production volume – As reported for the year 2020, the total chlorine production in Belgium was equal to 1074kt [18].

Chlor-alkali process plants in Belgium are all based on membrane cell technology from the year 2020. Considering emission from the Chlor-alkali process equal to 2.1 tons of $CO_{2\text{-eq}}$, the yearly indirect emitted CO_2 in Belgium is about 2.255 * 10^6 $t_{CO2\text{-eq}}/t_{Chlorine}$.

Decarbonization options for the Dutch Chlor-alkali industry - The Chlor-alkali manufacturing process can be conceptualized in a series of steps; steam generation, caustic soda preparation, brine preparation, electrolysis, caustic soda processing, hydrogen processing, and chlorine processing. Across the industry, the electrolysis process is the single largest consumer of electricity causing indirect emissions, whereas the steam generated in combined heat and power plants causes the direct emissions [7].

A key opportunity for decarbonization in the Chlor-alkali industry lies in the reduction of indirect emissions by the direct utilization of green electricity at electrolyzers. One solution is the large-scale implementation of zero-gap membrane electrolyzers to reduce electricity consumption. For the direct emissions, the placement of electric and biomass boilers or geothermal energy are the key decarbonization options [7].

Sustainable heat generation - Biomass boilers, electric boilers, and geothermal heat supply are the solutions toward sustainable heat generation [7].

Zero-gap membrane electrolyzers - Zero-gap membrane electrolyzers consist of an electrolytic cell where the anode and the

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[2] Estimated

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talieh.rajabloo@uhasselt.be

cathode are placed extremely close to the membrane wall that separates them; the distance between the electrodes is less than or equal to 1 mm. Minimization of the distance between the electrodes leads to a minimization of the voltage drop across the electrolyte and thus saves electric energy (typical 6-8%) [7].

Although the direct emission from the Chlor-alkali process is not lowered by the zero-gap technology, it has a serious influence on its electricity usage and thus helps to reduce indirect emissions [7]. Since the indirect emissions of the Chlor-alkali industry are considerably higher than the direct emissions, this alternate technology seems promising in reducing the net emissions [7].

Capacity increase to utilize peak-shaving production - Peak-shaving production means adapting Chlor-alkali production levels to the electricity prices. This is potentially not only financially interesting but can also help to make the production of chlorine more sustainable by direct utilization of green electricity [7].

Chlorine generation by electrolyzing of seawater -

Electrolyzed water has significant disinfection effects. Electrolyzed oxidizing water is an antimicrobial agent and electrolyzing of seawater results in high-chlorine seawater, which will have high potential applications in agriculture, aquaculture, or food processing. Electrolyzed oxidizing water is commonly produced by passing a dilute salt solution through an electrolytic cell, within which the anode and cathode are separated by a membrane. By subjecting the electrodes to direct current voltages, negatively charged ions such as chloride and hydroxide in the diluted salt solution move to the anode to give up electrons and become oxygen gas, chlorine gas, hypochlorite ion, hypochlorous acid, and hydrochloric acid, whereas positively charged ions such as hydrogen and sodium move to the cathode to take up electrons and become hydrogen gas and sodium hydroxide. Electrolyzed seawater, similar to electrolyzed oxidizing water, has strong disinfection effects, however, it is not as acidic as electrolyzed oxidizing water [25]. The rate of chlorine production via electrolysis of sweaters is reported to be around 1.7 g_{Cl2}/L with electric efficiency of 0.037 g_{Cl2}/kJ [25].

Plankton and bacteria are abundant in seawater, and certain coastal seawaters had been highly polluted. By contrast, deep ocean water (DOW) which makes up about 90% of the ocean volume is the cold (0-3

°C), salty seawater found deep below the surface of Earth's oceans [26].

Novel technologies for Chlorine production - To improve the efficiency of the Chlor-alkali process a major technology change is required. The oxygen reduction reaction (ORR) as an alternative cathode reaction can decrease energy consumption. A gas diffusion electrode that allows for the use of oxygen as the feed, and the oxygen depolarized cathode (ODC), leads to energy savings of up to 30% due to the lower thermodynamic decomposition voltages [27].

Table 4. Summary Table: Key Chlorine Data and Figures

Technical Performance	**Oxidation**
Feedstocks	- Salt (sodium, magnesium, and calcium chloride*) – [98% purity; 1882 kt/y]** - Purified water [2823 kt/y]**
Products	- Chlorine (Cl_2) [1000 kt/y] - Sodium hydroxide (NaOH) [1117.6 kt/y]** Hydrogen (H_2) [28.24 kt/y]
Required energy	- Electricity – [7.18 PJ/y]** - Steam [3.29 PJ/y]**
Emission	- CO_2 [211.8 kt/y]**
Chlorine production in Belgium	**2020** [16]
Chlorine (INOVYN and Vynova)	1074 kt
Costs and energy	
Energy (Electricity)	0.85-3.56 PJ/Mt$_{Cl2}$.
Salt cost	57 €$_{2014}$/t
Electricity cost	237.6 €$_{2017}$/kJ

* salt solution (NaCl, $MgCl_2$, and $CaCl_2$)

** for 1000 kt/y Chlorine (Cl_2) production

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talieh.rajabloo@uhasselt.be

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