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Risk analysis of the sodium hypochlorite production process: Focus on the chlorine line

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ABSTRACT

Mainly in the first part of COVID-19 pandemics, sodium hypochlorite was used as disinfectant, surprisingly also to spray over people. Several hazards may be associated to the production of this compound, such as chlorine gas toxicity and explosive hazards, due to the presence of hydrogen and chlorine, and corrosive hazards. Thus, loss prevention strategies must be *ad-hoc* developed to mitigate the risks. In the present work, the risk assessment of the first block of the process was performed, focusing the attention on chlorine risks. To this end, HAZOP analysis was first performed to identify the most critical top event, noticing the major issues in the quality of the final product and in the release of chlorine from pipes. Then, the fault tree analysis was built to calculate its failure rate. CFD simulations were used instead of empirical model to assess with a rigorous approach the chlorine dispersion, taking into account all the boundary conditions. In particular, by setting a hazardous chlorine concentration of 180 ppm corresponding to 50% fatalities for chlorine exposition for an exposure of 60 min, results without aspiration demonstrate the possibility for the cloud to impact workers at ground level also very far from the source point, while the chlorine cloud is moved upwards with a maximum length of 6.5 m when an aspiration is used, although the air ventilation speed is kept low.

1. Introduction

Being the risk dependent on the frequency of occurrence of any danger event and its consequences, it is necessary posing the attention on all the measures able to reduce the occurrence frequency in the industrial plants [1]. Qualitative tools (such as Hazard and Operability – HAZOP studies) [2–7] as well as quantitative methods (such as fault tree analysis and risk estimation models) [8–12] are widely used to evaluate suitable options for upgrade the design of the existing systems. Despite the different methodologies developed, the connection between the risk identification and the consequences calculations is often difficult, and the main reason for that is the mismatch of approaches [13]. The quantitative risk assessment for instance, allows the fine-tuning of a suitable chemical plant design, as well as of the management of the chemical processing facilities [14]. Accident simulation is nowadays an interesting method to perform the risk analysis, thus building a precautionary strategy. Gavelli and co-workers reported some computational fluid dynamics (CFD) programs nowadays available for industrial applications, especially for the modeling of the environmental sources and dispersion problems [15] . Accident simulation through CFD methods should be used as an effective way to carry out risk analysis and build protection procedures [16], ensuring accurate results even for complicated phenomena and geometries involved in the industrial plants [17]. Noteworthy, CFD results strongly depends on boundary conditions, source position etc. However, the coupling of CFD simulations together with risk assessment allow to generalize CFD results [18].

Among the numerous products of the chemical industry, sodium hypochlorite (NaOCl) represents one of the most popular, due to its effectiveness as a disinfectant, bleach, and sanitizer [19, 20]; therefore, several research reported the utilization of the cleaning methods of sodium hypochlorite [21] in combination with other substances, such as Fe^{2+} [22] and lactic acid [23]. Chlorine-containing disinfectants have been widely used, due to the reactivity of chlorine with organic substances to generate disinfection by-products [19, 24]. Thus, sodium hypochlorite presents antimicrobial activity with action on bacterial essential enzymatic sites promoting irreversible inactivation originated by hydroxyl ions and chlorination action [25, 26]. The electrolytic production of NaOCl from NaCl solutions began at the end of the 1800s and nowadays, the industrial production of hypochlorite is carried out by absorption of chlorine in a 21% solution of soda, both produced by

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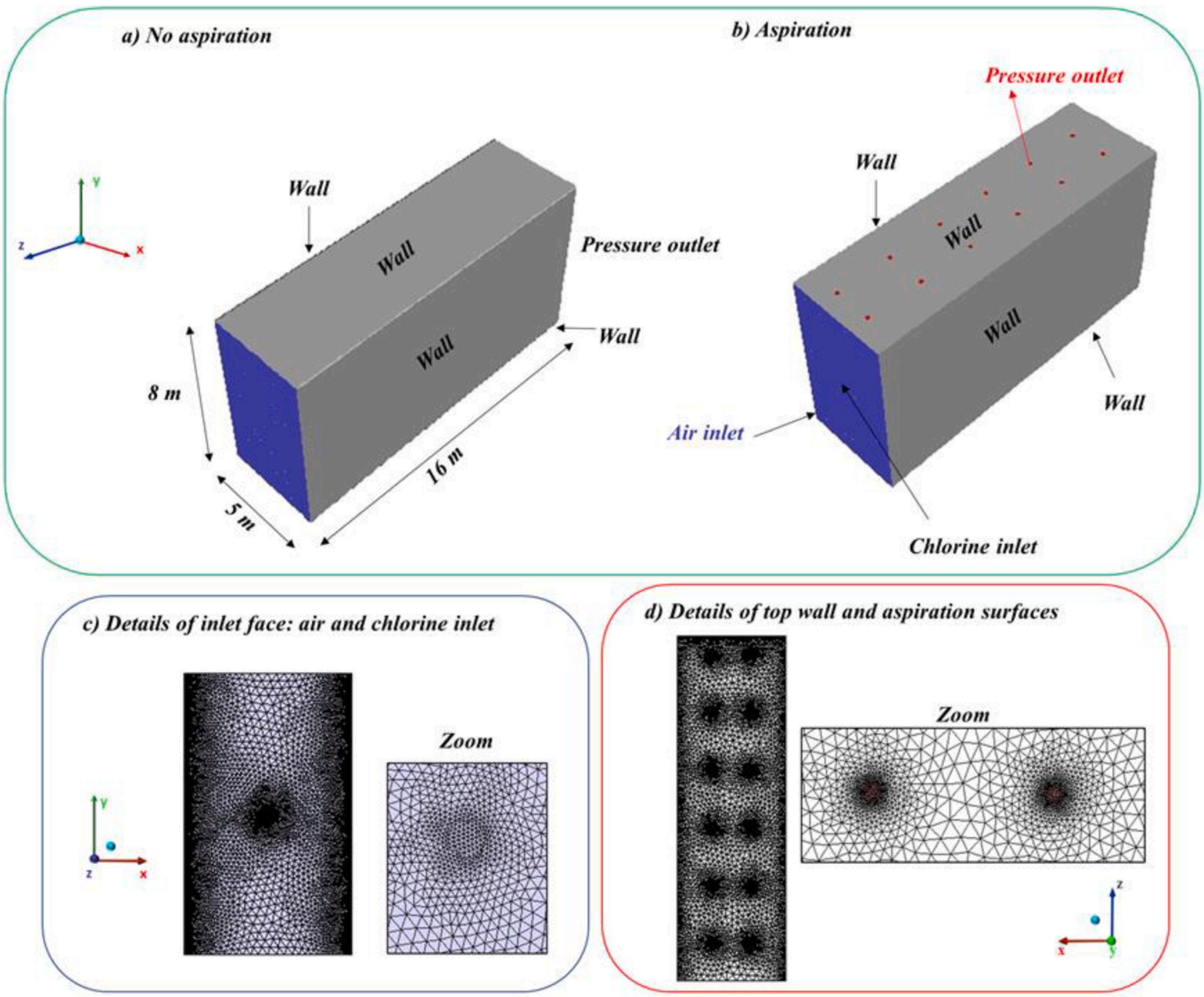

Fig. 1. Computation domain in case of no aspiration (a) and with aspiration (b) where boundary conditions are also reported. Details of the inlet face for air and chlorine used for both the conditions (c) and of the top wall and aspiration surfaces used in the second case (d).

electrolysis of a concentrated brine of NaCl [27–29]. Chemical plants for sodium hypochlorite production, based on the chlorine-soda process, are well known and poses several hazards such as chlorine gas toxicity and explosive hazards, due to the presence of hydrogen and chlorine; hence, safety measures should be taken in the design of the plant [30], thus requiring a systematic analysis to reduce the safety and environmental risks [8, 31–33]. In a previous work, in which a description of production process of sodium hypochlorite was reported, the risk assessment related to the part of the plant between the electrolytic cell and the chimney was performed, focusing the attention on hydrogen risks [34].

Conversely, in the present work, the attention is focused on the chlorine gas, which, once produced in the electrolytic cell, is sent to the sodium hypochlorite reactors through a vacuum piping. As concerns Cl_2, it represents an irritant and suffocating gas, with the threshold limit value (TLV) of the 3 mg/m^3 for the breathable air [3]. Inhaling small quantities of chlorine may cause shortness of breath, cough, and chest pain as well [35]. Several accidents related to chlorine loss occurred in the recent years, that had frequently caused the death or injury of many workers. In the early morning of May 6 (1991), for instance, in Nevada, a leak of chlorine gas created a hazardous cloud of poison gas over the city of Henderson, causing respiratory distress for over 30 people; by reconstructing what happened, it seems that the chlorine release was caused by the leak of brine from heat exchanger mixing with liquefied gas, thus creating a corrosive mixture which ate through pipes when product was transferred from storage tank [36]. Another important accident took place at Vieux-Thann (France) in a chlorine production plant; in particular, in the morning of January 13, 2004, the electrolysis installations were being started after a shut down. During the first start-up phase, the produced Cl_2 was sent to the bleach unit. However, after less than one hour, the hydrogen concentration in the residual gasses resulted too high, thus demonstrating a leak of chlorine. The prompt emergency shutdown and the presence of the confinement installations have allowed to direct the greater part of the leak to the neutralization tower, thus completely isolating the leak in 20 min: no employee or resident of the area was injured, and the quantity of chlorine gas released into the environment was limited to just a few kilos [37]. More recently, chlorine gas leak kills 12, injures 25 more after storage tank falls onto a ship at a Jordan port [38].

Despite the frequency of these accidents is not very high, the serious consequences make necessary the development of suitable strategies to perform the risk analysis, as well as the development of *ad-hoc* prevention/protection systems [39].

Based on the above – reported considerations, herein we proposed the risk assessment of a specific case study involving a sodium hypochlorite production plant, focusing the attention on chlorine risks. HAZOP analysis is first performed to identify the top events. For the most critical top event, the frequency analysis was performed by developing a fault tree. The consequence analysis was carried out by means of CFD simulations, that is considered as one of the promising cost-effective approaches in such analyses. Particularly, a continuous

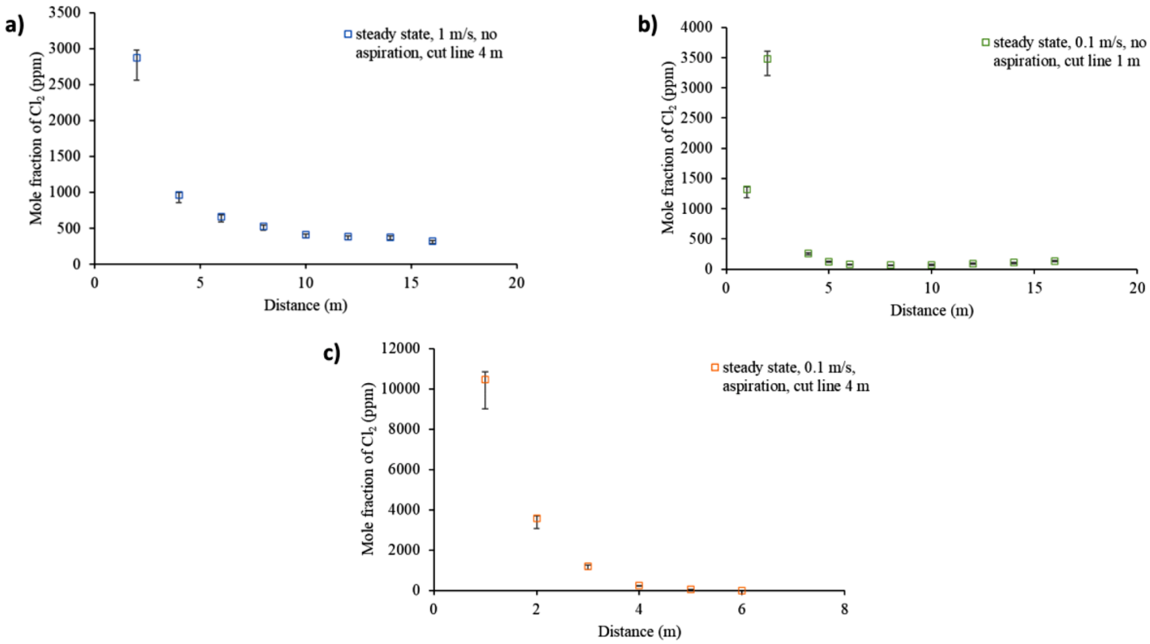

Fig. 2. Mesh convergence analysis for the three investigated scenarios considering Cl2 molar fraction on critical cut lines: a) chlorine release with wind speed at 1 m/s and no aspiration, convergence studied on a cut line 4 m high; b) chlorine release with wind speed at 0.1 m/s and no aspiration, convergence studied on a cut line 1 m high; c) chlorine release with wind speed at 0.11 m/s and aspiration, convergence studied on a cut line 4 m high.

release of chlorine from a leak generated on the pipeline inside an industrial shed was combuted and the size and shape of the effect zone relative to a 50% fatality level in different conditions of ventilation and aspiration was assessed.

2. Methods

This analysis started with the application of HAZOP method of the chlorine production section which led to the identification of the top events [40]. Once the most critical top event has been identified (chlorine release from transfer line), the individual risk was calculated and the effect zones at a 50% fatality level were built. Individual Risk at location x,y ($IR_{x,y,i}$) is given by the product between the frequency of the incident outcome case i (f_i) and the probability (p_{fi}) the incident outcome case i will result in a fatality. For a more conservative assessment, each incident outcome case was considered with an equal impact (probability of fatality = 1) throughout its geographical effect zone and $IR_{x,y,I}$ corresponds to f_i. Particularly, within the impact zone, the individual risk is equal to the frequency of that incident outcome case while outside is equal to 0. In this specific case, incident outcomes refer to the exposure to chlorine. To quantify the boundary concentration level of the effect zone, the probit correlation for chlorine deaths (K_1 = -8.29, K_2 = 0.92) was used and the value was set at 5.00 [41]. For an exposure of 60 min, the chlorine concentration to have 50% of fatalities is equal to 180 ppm. To assess the frequency of the top event, fault tree probabilistic method was carried out and the failure rates of the basic events were collected from literature data. To quantify the isorisk curves size, a three-dimensional CFD model of the chlorine release and dispersion in a typical industrial shed where the electrolyser and the sodium hypochlorite synthesis reactor could be housed was developed.

The continuum phase is modelled through the Reynolds-averaged Navier-Stokes equations (Eulerian approach). The continuity and momentum balance equations are

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \boldsymbol{u}) = 0 \tag{1}$$

$$\frac{\partial (\rho \boldsymbol{u})}{\partial t} + \nabla \cdot (\rho \boldsymbol{u}\boldsymbol{u}) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \rho \boldsymbol{g} \tag{2}$$

where ρ (kg m^{-3}) is the fluid density, $\boldsymbol{u}$ (m s^{-1}) is the fluid velocity vector, p (Pa) is the static pressure, $\boldsymbol{\tau}$ (Pa) is the stress tensor, $\boldsymbol{g}$ (m s^{-2}) is the gravity vector. As the natural ventilation problem in this study belonged to a flow with a high Reynolds number and high shear rate, these were solved using the standard k-ε model with standard wall function and considering compressibility effects [42].

As regards the species transport, Ansys predicts the local mass fraction of the specie through the solution of a convection-diffusion equation for the specie. The conservation equation takes the following general form:

$$\frac{\partial (\rho Y_{Cl_2})}{\partial t} + \nabla \cdot (\rho \boldsymbol{u} Y_{Cl_2}) = -\nabla \cdot \boldsymbol{J}_{Cl_2} \tag{3}$$

$$\boldsymbol{J}_{Cl_2} = -\left(\rho D_{Cl_2,m} + \frac{\mu_T}{Sc_T}\right) \nabla Y_{Cl_2} - \frac{D_{Cl_2,T} \nabla \mathrm{T}}{T} \tag{4}$$

where Y_{Cl_2} (-) is the local mass fraction of chlorine, $\boldsymbol{J}_{Cl_2}$ (kg m^{-2} s^{-1}) is the diffusion flux of chlorine which arises due to gradients of concentration and temperature, $D_{Cl_2,m}$ (m^2 s^{-1}) is the mass diffusion coefficient for chlorine, μ_T (Pa s) is the turbulent viscosity, Sc_T (-) is the turbulent Schmidt number, $D_{Cl_2,T}$ (m^2 s^{-1}) is the thermal (Soret) diffusion coefficient, T (K) is the absolute temperature. A pressure-based solver was adopted, and the SIMPLE algorithm was employed [43]. The time step adopted was 10^{-3} s and residuals were set at 10^{-6}. The time step was verified during the post processing through the Courant number, always between 10 and 20.

The computational domain and mesh were built and refined by means of the Design Modeler and Meshing packages of Ansys (Release 2020 R2). The industrial shed, the considered computational domain, is 16 m long, 5 m wide and 8 m high. Two configurations were considered: the former without aspiration system while the latter has got 12 aspiration holes (pressure outlet -0.3 barg, diameter 20 cm; rectangular mesh 1.5 × 3.0 m) [44]. In all the scenarios, chlorine is released from a discrete chlorine source located at a height of 4 m of the industrial shed face with a mass flow rate equal to 20 kg/h (chlorine produced from the electrolytic [44]) and a diameter of 2 cm. The domain as well as the boundary conditions are showed and reported in Fig. 1. The inlet

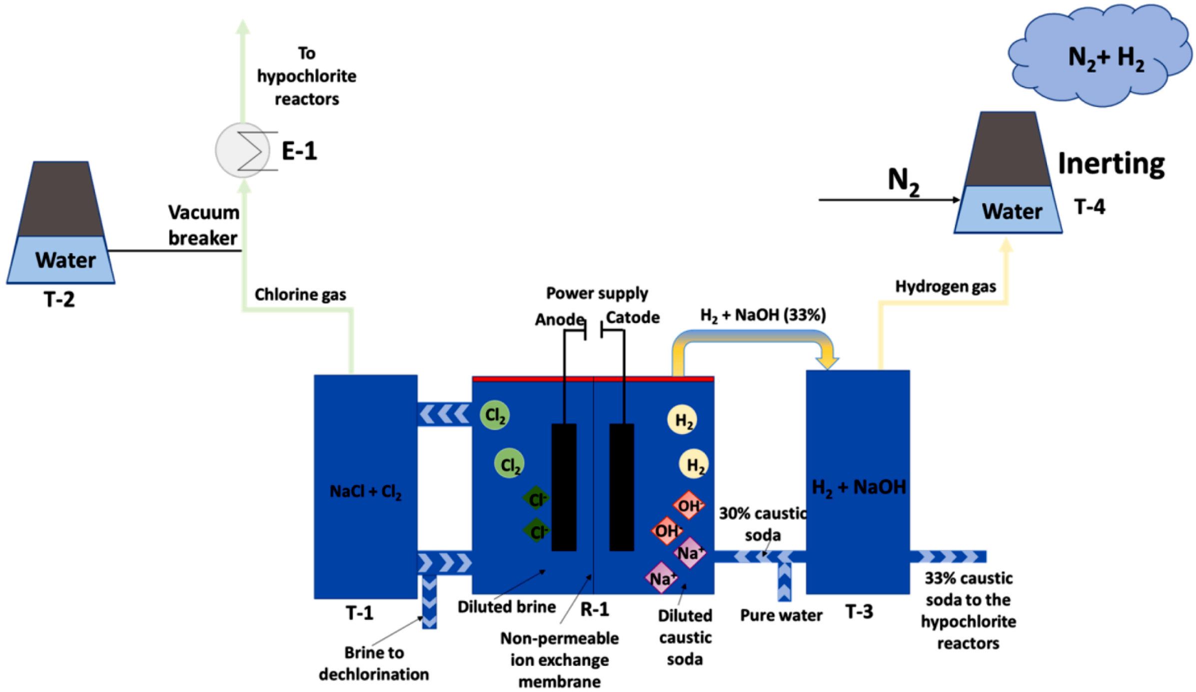

Fig. 3. Flow diagram focused on the units between the electrolytic cell and the sodium hypochlorite production reactors.

Table 1
HAZOP analysis referred to the scheme reported in Fig. 3.

Parameters	Guide words	Causes	Effects	Protections/notes
Flux (Cl_2 from the electrolytic cell R-1)	No/less	Wrong manual valve closure	High pressure within the cell; Not completed batch	Recovery of solution in the reactors. Limit switches alarm in closing
Flux (Cl_2 from the electrolytic cell R-1)	More	Flow rate is not reduced while finishing	Not correct hypochlorite composition; quality problems	-
Flux of the service water	Less	Breakage of tube in the condenser E-1	Inlet of water in the process stream; outlet in T-2; dilution of the product; increasing of pH values	Periodic pH measurements
Flux of Cl_2	Less	Wrong flanged mating seal	Inlet of air to the loop	Cl_2 system works in vacuum, that leads to air inlet
Temperature of Cl_2	More	Failure of temperature transmit in closure	Higher humidity of Cl_2 to the reactors. Lower concentration of hypochlorite produced. Quality problem.	Chemical analysis of hypochlorite produced
Temperature of Cl_2	Less	Failure of temperature transmit at the start	Not significant	-
Pressure of Cl_2	More	Failure of pressure transmit in closure	High pressure (>1) within the cell and in the vessel (T-1); Cl_2 release via hydraulic seal (T-2) due to the overpressure	A pressure alarm and a pressure switch on the H_2 line stop the plant
Pressure of Cl_2	Less	Failure of pressure transmit at the start	Low pressure; Entry of air	Hydraulic seal with an adequate height of liquid
Vessel T-1 level	More	Failure of the level transmission through a level transmit in closure.	Tank filling, high backpressure, and pressurization of Cl_2 line; membrane failure, reduction of Cl_2 production	A level switch stops the cell and brine feed. Blockage of the output line from T-1
Vessel T-1 level	Less	Failure of the level transmission through a level transmit at the start.	Defusing of the pump; low inlet flow at the cell; possible membrane damage	Blockage of the cell
Hydraulic seal (T-2) level	Less	Missed restoration of the hydraulic seal level	The required vacuum is not guaranteed at initial phase, before the activation of the cell	Check list to guarantee the hydraulic seal control
Cl_2 composition	Other than	Oxygen in the chlorine circuit due to a lower efficiency of the membranes	No significant; efficiency problem	Introduction of an analysis system of oxygen concentration
Cl_2 composition	Other than	Water in the chlorine circuit due to missed intercepts of case drain	Dilution of the product, difficult pressure adjustment; quality problem	Valves signals

velocity of air was set at 0.1 m/s and 1 m/s on the Air inlet surface (see Fig. 1) to simulate two different conditions of natural ventilation while on pressure outlet surface a pressure of 0 barg was set. The bottom and the lateral walls of the shed were set as the wall boundary condition.

For both the configurations a tetragonal unstructured mesh was optimized and used with 424,219 for no aspiration scenarios (minimum elements volume 3.9e-09 m^3 and maximum 1.4e-01 m^3) and 488,727 for the scenario with aspiration (minimum elements volume 9.3e-11 m^3 and maximum 2.3e-01 m^3) elements respectively, refined close to the chlorine inlet and the 12 aspiration holes (we set 200 elements along the surface edge of each surface). The optimal meshes were obtained by performing simulations at different cell number, converging to the lowest cell number with the highest accuracy. In particular, two additional meshes were used for the three investigated cases, containing 10^6 and $3\bullet10^5$ elements respectively. From the comparison of the chlorine fraction on different cut lines, it can be seen that in the most critical

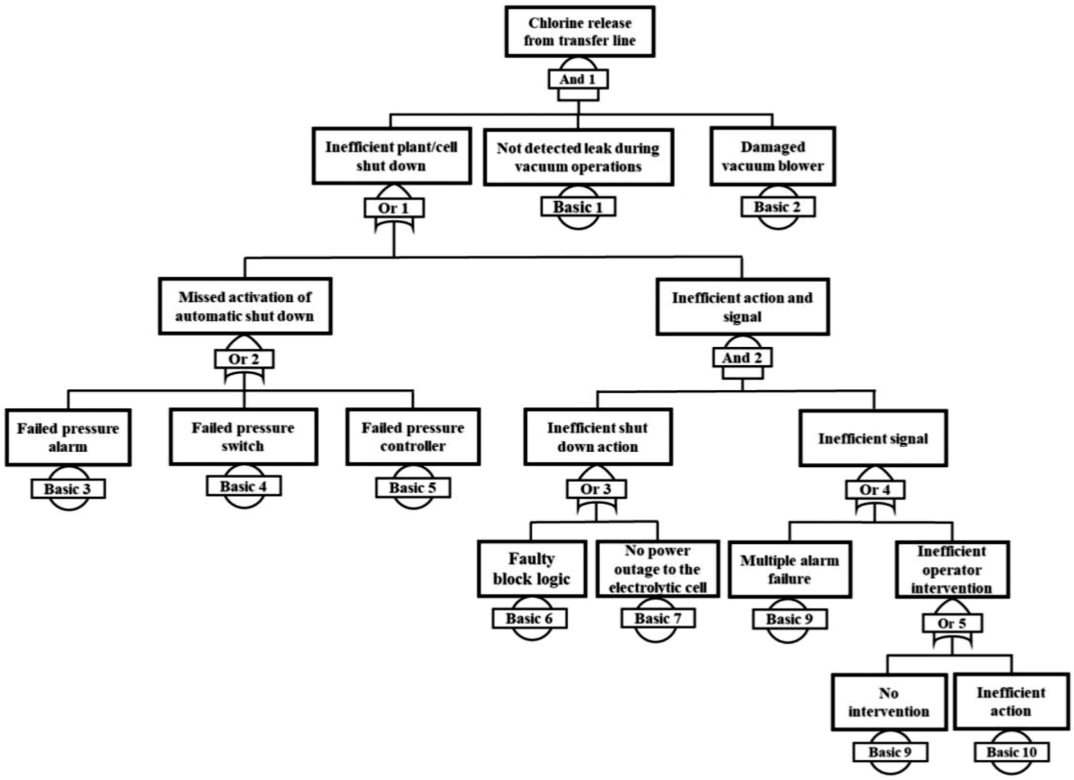

Fig. 4. Chlorine release from transfer line fault tree.

areas of the domains (i.e., those where the concentration gradient in the steady state is greatest) the deviation between the mesh used and the finer mesh is about 5% while with the coarser mesh a deviation ranging from 13 to 20% (Fig. 2) In order to use a low number of cells, the intermediate mesh was used.

3. Results

The HAZOP method was carried out on the part of the plant in which chlorine is present. Particularly, we focused on the process section where chlorine is produced (from the electrolytic cell R-1 to sodium hypochlorite reactors via vessel T-1 and heat exchanger E-1 in Fig. 3).

In Table 1 the results of the HAZOP analysis are given. Five different elements (Flux, temperature, pressure, level, composition) were analyzed, at different conditions of deviation from normal values:

1) The main sources of the variation of fluxes (for the chlorine line as well as for the service water) may be associated to the wrong valve closure (for the chlorine line) or the breakage of tube in the condenser (for the service water), thus leading to the dilution of the desired product. To solve the problems, periodic inspection of the valves, use of switch alarms and regular controls of the product concentration are recommended.
2) Higher or lower temperatures of chlorine gas may be caused by the failure of the temperature transmit. A higher humidity of Cl_2 (when temperature $T >$ normal value) leads to quality problem of the desired product. For this reason, an adequate monitoring of the product is necessary.
3) As concerns the pressure, values higher than the nominal ones may affect the chlorine release via the hydraulic seal. The failure of the pressure transmit during the closure is the main reason for this effect, while the inlet of air may be caused by the failure of the pressure transmit at the start-up. Some protections are thus necessary, such as an adequate height of liquid in the hydraulic seal as well as the use of pressure alarms.

Table 2
Failure rates.

Event	Failure type	$\lambda \bullet 10^{-6}$ (events/year)	P	Reference
1	Not detected leak during vacuum operations	11.4		[45]
2	Damaged vacuum blower	9.1		[46]
3	Failed pressure alarm	14		[47]
4	Failed pressure switch	8.3		[47]
5	Failed pressure controller	16.9		[48]
6	Faulty logic block	1.45		[49]
7	No power outage to the electrolytic cell	2.0		[50]
8	Multiple alarm failure (generic)	0.2		[47]
9	No intervention		0.1	[51]
10	Inefficient action		0.01	[52]

4) Higher or lower levels may be ascribed to some failure of the level transmit, in closure or at the start, respectively. The most dangerous effect is surely the rupture of the membrane cell, to which corresponds the reduction of the chlorine product as well as the possible formation of hydrogen-chlorine mixtures.
5) Finally, the presence of other substances in the chlorine circuit may cause quality problems, for which it is recommended the use of an analysis system for the undesired species.

Therefore, from the HAZOP analysis it was found that the major issues are related to the quality of the final product. Moreover, there are safety issues due to the inlet air due to vacuum system failures or to the release of chlorine from pipes. In this work, the consequence analysis together with the frequency one of this last event was carried out.

Fig. 4 shows the developed fault tree to calculate the frequency of the top event "chlorine release from transfer line". By using the typical top-down approach, we identified the intermediate events and the

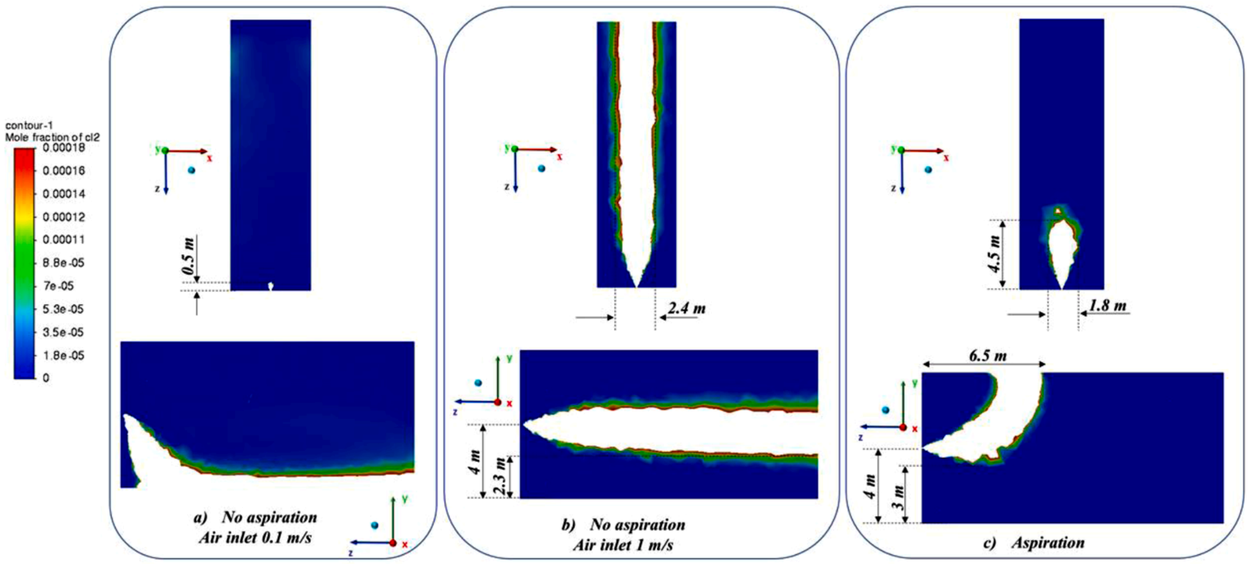

Fig. 5. CFD results after 1 h with no aspiration and air inlet velocity of 0.1 m/s (a) and 1 m/s (b) and with aspiration and air inlet velocity of 0.1 m/s (c). Effect zones geometries and sizes are also reported, relative to a probit value of 5.

component level failures (basic event) that cause the system level failure (top event) to occur. Particularly, a chlorine release may happen due to the simultaneous fault of the vacuum blower installed on the chlorine line, the missed leak detection and the inefficient shut down of the plant. This last event was identified as an intermediate event and was developed further to reach the basic events reported in Fig. 4.

A list of basic events as well as their failure rates is given in Table 2, where λ is the failure rate (event/millions of hours) collected from literature and technical reports as reported in the last column. The average frequency evaluated from these data for 3 years of plant operation of 25,000 h (8333 h/year) is equal to 10^{-3} events/year.

Then, by using the probit function set at 5, we calculated the chlorine concentration value to reach the 50% fatality level after 60 min corresponding to 180 ppm.

Instead of using the typical empirical models, we used CFD calculations to visualize the size and shape of the effect zones with different conditions of ventilation and aspiration (Fig. 5). In Fig. 5, the zones where chlorine concentration is higher than 180 ppm are reported in white. It is worth noting that once the probability of fatality p_{fi} is set at 1 within the effect zone, the frequency calculated by the fault tree analysis is equal to the risk index IR within the effect zone (while it is zero elsewhere), whose size and shape is equal to those of the white areas shown in Fig. 5. As shown in Fig. 5, in the case of low ventilation speed, the chlorine disperses from the release point (located at 4 m high) downwards due to the negative buoyancy effect. In this case, it is easy for the cloud to impact workers at ground level. In the case of more sustained ventilation the momentum prevails over the buoyancy and the cloud is always anchored at the release point but develops in length, descending to a maximum of 2.3 m above the ground. In the case of aspiration, although the air ventilation speed is kept low (better comfort), the chlorine cloud is moved upwards with a maximum length of 6.5 m, and this represents the best case. Considering the value of the risk index that falls within a range of values for which a further assessment is required, it is important to design and use appropriate prevention measures (e.g., ventilation and aspiration systems) to reduce the extent of the effect zone until the risk is reduced to acceptable limits.

4. Conclusions

In this work, we focused on the risks related to the production and use of chlorine in the synthesis process of sodium hypochlorite, mainly used as a disinfectant. The HAZOP analysis highlighted two types of problems: some relating to the quality of the product, others relating to safety, such as the case of chlorine release due to the breakage of the tube connecting the electrolyzer to the hypochlorite synthesis reactor. A suitable fault tree was developed to calculate the failure rate of this top event and a CFD model was developed to size the effect zone of the case of different ventilation and aspiration configurations. Results showed the great importance of the connection between the different methodologies for the risk assessment and confirmed the great potentiality of CFD simulations in case of toxic gases release in indoor environments. In particular, the importance of an *ad-hoc* ventilation-aspiration system was highlighted, useful to reduce the size of the effect zone (where the individual risk is high and equal to 10^{-3}/year) and to limit the impact that the chlorine cloud may have on workers present on the ground.

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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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