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Summary

Tackling safety issues related to emerging techniques is crucial in making them safer and acceptable. Among these strategic techniques, the Carbon Sequestration Chain is for sure one of the most promising solutions in governing the Global Warming. It includes three distinct steps, namely the capture, the transportation and the storage process that allow for the sequestration in deep reservoirs of emitted CO_2 for mid-to-long term disposal.

This innovative technique requires very large and extensive infrastructures including capture plants, pipeline networks and injection systems in suitable sites. From a safety perspective, the transportation step via dedicated pipelines is critical given the large amounts of handled CO_2 and its asphyxiating and denser-than-air properties that make any loss of containment (LOC) very peculiar. Criticalities escalate once these pipelines push on very populated areas (i.e. Europe) where their deployment may interfere with residential and industrial sites as well as with strategic transport routes and facilities.

On this light, accurate and detailed hazard studies (Quantitative and Societal Risk Analysis) are crucial in supporting and demonstrating the acceptability, the safety and the reliability of such an innovative Carbon Sequestration solution.

This research work covered key aspects of Safety Science and Technology applied to CO_2 pipeline networks, as a crucial and critical step in the framework of the Carbon Sequestration chain.

The activity was mainly driven by the need to fill some relevant knowledge gaps affecting and partially invalidating the outcomes of the safety studies applied to CO_2 pipelines. In fact, expected significant inaccuracies result in a weakened overall risk assessment procedure. Wider gaps are related to the unavailability of reliable and comprehensive source models for the CO_2 released from the pipeline as a result of failures, punctures and ruptures. In addition, experimental data for tuning purposes are substantially lacked especially in what pertaining to peculiar properties of a CO_2 release.

In view of the above, a synergic experimental and modelling activity was performed in order to make a modelling tool available for hazard analysis purposes applied to the CO_2 transportation step.

The research work has been based on an exhaustive literature survey that highlighted both a deficiency in modelling CO_2 transportation hazards and discrepancies in how to manage and model CO_2 releases from damaged pipelines. In addition, no comprehensive release models emerged to be available and extensive supporting experimental data were lacked.

The research work firstly analysed the detailed behaviour of the CO_2 during a rapid depressurization both from a thermodynamic and a fluid dynamics perspective. This substance, in fact, displays peculiar properties mainly resulting in multiphase releases of supercritical, liquid, gaseous and solid CO_2 (dry-ice), therefore complicating any investigation and modelling activity. Solid-liquid-vapor CO_2 systems have been investigated as well as the conditions supporting the

occurrence of multiphase sonic flows across the pipeline rupture as a relevant parameter in the qualitative and quantitative evolution of the release.

Alongside, an experimental campaign was started in order to collect self-made data to be used in the model tuning step. In this context, a release apparatus has been sized and arranged, enabling a time recording of main CO_2 release parameters (pressure, temperature, heat transfer). Charging pressures up to 75 bar and initial temperatures in the range 0-40 °C were investigated. Major results were observed concerning the overall discharge dynamics and the occurrence of competitive mechanisms in its determination (phase change mechanisms in the system bulk, discharge dynamics). Depending on the initial conditions, neither an isenthalpic nor an isentropic expansion is met even if these typical transformations are usually invoked in risk assessment procedures. At laboratory scale, evidences showed that gaseous charges may evolve in multiphase releases even under moderate charging pressures (40-60 bar). Incidentally, information about other relevant discharge parameters were derived and/or deducted, namely the discharge coefficient and the conditions leading to the occurrence of solid-vapor mixtures (always neglected in quantitative risk assessment studies).

The research work then focused also on the development of a CO_2 release model, lacked in usual CO_2 hazard studies. The aim of this module was to recommend an exhaustive and a properly detailed approach useful for Risk Assessment procedures. The model is robustly based on a set of conservation balance equations, thermodynamics issues, heat transfer and phase-change constitutive relations and reliable equations of state.

The challenge of making a comprehensive model available has been achieved thus supporting hazard studies and estimates in the framework of the Carbon Sequestration chain.

The test against experimental data revealed a good qualitative and quantitative agreement with most of experimental trials that have been adequately matched and interpreted. This modelling procedure has also allowed to analyse mechanisms not experimentally affordable.

The extension of the model to real scale operations (scale-up) has been focused on its capability to give a description of expected outcomes of releases across different ruptures and under variable operative conditions. Modelling outcomes have shown that the rupture severity affects both the minimum expected temperature inside the pipeline and the depressurization path to a multiphase release. These parameters are relevant in the determination of the release evolution and duration thus orienting emergency planning.

A more detailed analysis has provided the specific combinations of operative conditions and rupture pattern leading to multiphase releases and potential simplified modelling conditions. In this sense, results have showed that solid CO_2 content up to 45 % by mass can be expected during a release of CO_2 thus invalidating actual adopted assumptions in standard Carbon Sequestration hazard studies.

In addition, specific conditions lead to allowable modelling simplifications. Release evolutions in very large pipelines (length to diameter ratio larger than $6 \cdot 10^4$) admit the bulk isothermal condition and the temperature dependency can be ignored accordingly. A negligible wall effect, i.e. a neglectable heat transfer phenomenon across the domain boundaries, can be instead invoked with an operative pressure lower than 45 bar and in pipelines shorter than 1500 m.

Among major listed outcomes, that are discussed in detail in the Doctoral thesis, the proposed model has made it possible to address main knowledge gaps of hazard studies applied to CO_2 transportation infrastructures. A successful experimental tests-modelling-validation approach has supported reliable remarks to fix common inaccuracies and modelling pattern adopted in prevailing CO_2 hazard assessments.

The continuous research for new methodologies and tools to be applied to emerging techniques has been the main driver of this three-years research work. The belief that the Safety Science and Loss Prevention studies applied to innovative technologies can drive and improve their implementation and dissemination, strengthened this research experience.

Further developments are ongoing to improve this research topic and one of the most significant achievements has culminated with the study and the proposal of a European Carbon Sequestration supply chain. In this study, a coupled economic- and risk- optimization criterion has been used to best drive the network routing as an innovative integrated and holistic approach. Such a methodology provides a valuable way to assess the incidence of risk mitigation and safety measures on the overall sustainability and economics of the Carbon Sequestration chain.

Problem addressed

Safety issues applied to new and emerging technologies are a key factor in determining their acceptability and safer operative conditions. Common risk assessment techniques can be used to assess hazards and risks related to emerging systems in addition to innovative tools like the Computational Fluid Dynamic and process simulators.

The Carbon Sequestration Chain represents an innovative medium to long term solution to govern the Global Warming and takes place into three distinct steps: the capture from large stationary source, the transportation through a suitable and dedicated pipeline network and the final injection step.

Hazards related to the CO_2 handling operations emerge given its asphyxiant nature and the leakage potential from the infrastructure.

These emerging technologies suffer a lack of reliable risk assessment methodologies that usually neglect relevant phenomena during the failure and release evolution. In this sense, unreliable results apply to the sizing, planning and operation of such these infrastructures. In addition, their recent application suffers also a lack of available data for model tuning purposes.

In this framework, this research work aimed at filling existing gaps with the following relevant topics:

- development of a comprehensive source model for risk assessment purposes and applicable to complex releases from a CO_2 handling infrastructure;
- collection of relevant experimental CO_2 release data to be used in the model tuning procedure;
- definition of safety distances around the CO_2 infrastructure for planning and operational purposes;
- support the development of an optimized European CO_2 supply chain based on both economic and hazard criteria.

State of the art

The CCS (Carbon Capture and Storage) technique is an option that is being developed to control CO_2 emissions from large stationary sources.

This solution is comprised of the capture, the transport of the CO_2 and the final storage operations. The intermediate step may involve different media like trucks, trains, ships or pipelines ([1]). Among these, the transport by pipeline is considered one of the best solutions especially when dealing with large quantities of CO_2 over distances up to 800 – 1000 km.

The experience gained with the CO_2 transportation on large scales is mainly derived by infrastructures located in the USA but it is directly applicable to other regions. Most CO_2 pipelines, in fact, are situated in areas characterized by very low-density population, this influencing the precautionary measures taken.

The deployment of CO_2 pipelines in other regions, like in Europe, will be linked to large networks located in very densely populated areas. IPPC on CCS has underlined the lack of safety experience in what concerning the operation of CO_2 pipelines in densely populated regions as a gap in the knowledge ([2]).

Safety is one of the key aspects that should be assessed in the planning and operation phase of the CO_2 transport. Recently several QRA (Quantitative Risk Assessment) studies for CO_2 pipelines have been proposed ([3,4,5]).

Their analysis allows for the identification of several gaps and uncertainties concerning especially the source modeling, the dispersion behavior as well as the establishment of CO_2 threshold values and possible effects at different distances from the infrastructure ([6,27]).

These uncertainties and gaps lead to controversies around the CO_2 transport hazards. For example, some authors claim the low (tolerable) risk in the CO_2 handling operations ([7,8,27]) while others suggest that risks associated are well understood ([9,24]). In addition, it is generally suggested that there is no enough experience in designing and operating CO_2 pipelines in highly populated areas and therefore CCS and EOR systems pose a risk at least comparable than pipelines carrying hydrocarbons ([1]).

A systematic evaluation of impacts of uncertainties in input parameters on the results of QRA has been performed in some studies ([1,2]) leading to the conclusion that following aspects need further knowledge to fill existing gaps:

- failure scenarios and their probability;
- CO_2 releases from pressurized domains;
- dispersion modeling.

Following sections are dedicated to the investigation of main knowledge gaps affecting these QRA aspects applied to CO_2 with a special focus on the CO_2 releases modeling.

QRA procedures generally refer to two types of failure scenarios: puncture and full-bore rupture.

In what concerning pipelines, failures are mainly due to corrosion, infrastructure defects, ground movement, operational errors and third-party interferences. Recently, NaTech causes like those invoking earthquakes and floods have been included in safety reports.

Different failure distributions between the puncture and the full-bore rupture are usually employed and consensus is still missed ([1]).

Therefore, overall cumulative rates assumed are ranging from 0.7 to 6.1 per 10000 km per year. However, these data are often based on natural gas pipelines and may not be valid for CO_2 pipelines due to the peculiar properties of this substance. In addition, as discussed by some authors ([1,26]), historical failure rates for CO_2 infrastructures cannot be compared with those of natural gas being the gained experience limited ([28]).

Differences with pipelines carrying natural gas may arise because of the acidic behavior of the CO2 in presence of impurities, like water, inducing higher failure rates ([10,29]). Other impurities are represented by SO_x, NO_x, O_2 and H_2S.

The effect of varying failure rates on the QRA results is represented by varied and unreliable risk contours that are strictly linked to the safety distances assessment. Some authors ([1,3,25]) underline that these uncertainties lead to safety distances derived from a pressurized CO_2 release varying from 37 to 117 m.

The modeling of the CO_2 behavior subjected to a rapid depressurization is a critical step in QRA of CO_2 pipelines and it is the main topic of this research work. This step consists in the definition of the source term that it will contribute to the subsequent atmospheric dispersion through the investigation of the source aspects (geometry, direction), quantity, velocity and release duration.

In what concerning the CO_2 behavior, thermodynamic aspects are crucial in the determination of reliable results.

It is known that any release is specific to the substance released as its properties vary in time with the phenomenon evolution. Among these, the most important are the pressure, the volume and the temperature that define the instantaneous CO_2 state due to the depressurization. A first main issues of concern are the lack of comprehensive thermodynamic models able to describe all aggregation states pertaining to the CO_2 that may arise (supercritical, gaseous, liquid or solid).

Studies and experimental evidences give reason to state that main mechanisms involved in a rapid CO_2 depressurization are those listed below that are depending on the initial aggregation state:

- occurrence of a pressurized release that may be persistently gaseous or in form of a multi-phase mixture (vapor-liquid or vapor-solid);
- flashing phenomena that interest the liquid portion of the expanding jet;
- rainout and snow-out phenomena;
- atmospheric dispersion of a heavy-gas cloud and eventual sublimating dry-ice bank formation acting as a delayed dispersion hazard source ([11]).

In the light of the listed features, the modeling of the outflow following a CO_2 pipeline failure is challenging given the large number of linked and complex phenomena ([23,28]) that are governing the whole discharge process. Some of them, however, are characterized by uncertainties and gaps also derived by the lack of many supporting experimental data ([1,12]).

The dense release following a full-bore rupture is calculated relying on transient two-phase models to take into account the discharge of vapor-liquid mixtures across the rupture plane. These models were originally applied for the LPG releases ([13]) but are usually generalized for other substances giving rise to uncertainties especially in the CO_2 case that is characterized by the peculiar thermodynamic behavior. In cases where the CO_2 is handled in its gaseous state, i.e. in EOR operations, a transient model for gas pipelines is instead used.

All these models lack a comprehensive approach able to manage all phenomena linked to the rapid depressurization including the prediction of the solid phase occurrence giving then rise to the snowout.

Different model approaches are available and some of them are indicated below:

simplified models made of algebraic equations (AEs) and ordinary differential equations (ODEs) resulting in scenarios differently conservative;

general models based on conservation equations (ODEs and PDEs) and thermodynamic correlations (AEs) giving rise to mathematical structures usually handled through numerical approaches ([14,15]);

detailed studies based on the Computational Fluid Dynamic (CFD) approaches characterized by a high degree of detail coupled with a relevant computational – burden demand usually not adoptable in emergency situations.

Uncertainties affecting these approaches are firstly linked to the thermodynamic expansion nature followed by the CO_2 during the rapid depressurization. Available experimental data do not actually give a unique interpretation of the occurring expansion that ranges between the (ideal) isentropic and the (ideal) isenthalpic pathway. Any accuracy affecting the supposed depressurization pathway will alter the thermal dynamic as well as the mixture nature resulting from the pressure gradients.

In addition, considerations must also be given to the effects of frictions and heat transfer which are both expected during the blowdown phenomena and are phase – dependent mechanisms.

To date, large part of the proposed outflow models relies on the HEM (Homogeneous Equilibrium Model) approach ([1,12,16,17]), thus imposing mechanical and thermal equilibrium during the decompression process. The gap is consequently linked to the absence of non-equilibrium phase transitions, phase-slip and delayed boiling mechanisms.

According to some authors ([1,17]), while ignoring phase-slip may be justified by experimental evidences, the same approach based on non-equilibrium vaporization is difficult to be a priori justified. The final part of the present work investigates the effects in neglecting these aspects on the release modeling.

All available models allow for relevant outcomes to be fed to into the subsequent dispersion model. These outcomes are the release duration, the instantaneous discharged mass flow rate and the out coming mixture quality. The effect of varying assumptions may lead to the lack of these essential parameters especially in terms of number and nature of expected phases, these strictly linked to the thermal and fluid dynamic system behavior.

The pressurized release of CO_2 gives usually rise to a dispersing heavy gas that is directly derived from the expanding jet and/or the sublimating dry-ice bank. Because of the CO_2 density field, this cloud moves close to the ground and its behavior is therefore influenced by local conditions and orography.

Features of a denser-than-air dispersion process are reported below ([1]):

- the behavior is deeply affected by local wind and temperature field;
- the resulting cloud behaves dependently on the local terrain conditions;

- heat and mass transfer with surroundings and with the ground are expected to occur;
- the substance encounters gravity-induced spreading and density gradients reduce vertical mixing leading to stratification mechanisms.

These features apply to the CO_2 dispersion since common CCS and EOR operative conditions are likely to give rise to a heavy-gas dispersion following a rapid depressurization.

Limitations linked to heavy-gas models employed are due to the fact that they have not been specifically developed and addressed to the CO_2 dispersions. In addition, no modeling solutions involving the solid phase soil deposition ([18]) are available except for complicated CFD simulations whose runtimes are totally unsuitable for emergency responses.

In addition, important factors are affecting the CO_2 dispersion behaviour.

The rupture of buried pipelines is linked to the ejection of the soil lying above causing the formation of a crater. Its shape and size are expected to induce modifications on the release features as well as the soil deposition of solid CO_2. The resulting jet will have an impinging behavior (75 % velocity reduction, ([8])) and the formation of a sublimating dry-ice bank is in fact usually linked to this occurrence ([11]). It is known that the soil type and the pipeline depth are crucial parameters in the determination of the crater size even if no models are available to estimate it.

In addition, the supposed expansion pathway will influence the subsequent dispersion step. This is mainly due to the appearance of the dense phase (initially liquid, finally solid) that may partially fall on the ground giving rise to the aforementioned dry-ice bank ([11]). In this case, the atmospheric dispersion experiences the additional delayed source represented by the sublimated CO_2 from the bank surface. Some authors highlighted some uncertainties related to flash calculations ([19]) that are inducing errors in the vapor quality estimation and therefore in the occurrence of rainout and snow-out processes.

The dispersion mechanism is also initially influenced by the direction and the momentum of the released CO_2. For example, different scenarios in terms of spatial CO_2 concentrations are expected depending on the occurrence or not of crater-release collisions ([8]).

In addition, meteorological conditions have a crucial role on the hazards related to the CO_2 dispersion. Actual debate is focused on the combination of release direction and meteorological stability class linked to the worst scenarios that may take place ([20]).

Finally, many gaps exist in what dealing with the effects on the human health. The expected impact estimation suffers some knowledge gaps. In fact, acute and chronic exposure to more vulnerable people still should be assessed. Current studies are based on full healthy subjects ([21]) and proposed threshold concentrations are derived from old investigations. In addition, only few Probit functions are available for QRA purposes indicating that no international standard for the CO_2 threshold exposure limit exists ([8]).

Moving from the conclusions of some works ([1,4]), the most important issues concerning the knowledge gaps in QRA's for CO_2 pipelines are listed below:

- it is not certain if natural gas failure rates are applicable to the CO_2 case. Supposed divergences may amount up to an order of magnitude;

- preliminary results show that the type of release is a crucial parameter affecting the hazard related to a rapid depressurization of a CO_2 pipeline and there is no consensus on the type of release that should be used in QRA's studies;
- methodological standards are required;
- the lack of experimental data prevents the development of reliable CO_2-specific models;
- no comprehensive release models are available and the predictions in what dealing with the mixture quality discharged from the pipeline deeply affect the subsequent dispersion process.

Key Innovations

– the proposal of a new comprehensive CO_2 release model to be included in Safety Studies, lacked in usual Quantitative Risk Assessment procedures applied to Carbon Sequestration operations;
– the exploitation of a combined experimental and modelling approach to support the development of innovative and useful technique and results for Safety studies purposes;
– the collection of CO_2 release experimental data needed to better focus on main peculiar mechanisms leading to multiphase discharges from Carbon Sequestration infrastructures;
– the adoption of a CFD (Computational Fluid Dynamics) approach in the Safety Assessment field to support the development of detailed tools and conclusions in the determination of safety distances;
– the proposal and preliminary design of a new and innovative Carbon Sequestration Chain whose routing is optimized both from an economic and risk perspective.

Applications, implementations, results

The performed research activity finds its main application in the field of Applied Safety Science. In detail, illustrated results can be applied to the development, planning and operation of the transportation step in the Carbon Sequestration Chain.

Firstly, the quantification of safety distances from Carbon Sequestration pipelines can surely drive and support their routing in a safer and sustainable way in order to identify, minimize and treat main related risks due to accidental containment releases.

The methodological approach employed can be used as an example of an inclusive way to combine the experimental and modelling practice in fields where a lack of data and/or methods exists.

In addition, the proposed model can be straightforwardly used in safety assessment operations applied to CO_2 handling operations thanks to its well-balanced feature between computational burden and results detail. Its structure allows for the extension to different pure substances and mixtures composed by CO_2 subjected to a release. Main release parameters can be obtained as basis for further investigations.

Detailed results show that:

- assumptions and simplifications usually employed in QRA applied to CO_2 transportation fail under common operative conditions. This is especially true for hypothesis leading to the neglection of solid phase appearance during rapid releases and once the isothermal approach (for long pipelines) is employed;
- the model extension and application to real infrastructures shows that some existing pipelines may be subjected to three-phase releases thus including the solid CO_2. Potential scenarios lead to the deposition and formation of sublimating banks that should be included in any QRA procedure to ensure successful safety and rescue operations;
- non-equilibrium thermodynamic phenomena may arise, especially under conditions of large orifices and in highly-pressurized pipelines. In this context, very altered dynamics are expected;
- safety distances around CO_2 pipelines are in the order of hundreds of feet and can be worsened by the appearance of a solid phase thus generating a slowly sublimating dry-ice bank that acts as delayed CO_2 source;
- CO_2 transportation infrastructures are not without risks and the CO_2 asphyxiating property requires very careful planning and sizing activities. A coupled economic and risk minimization plan is thus required. Results show that the best routing (minimizing overall costs and societal risk) is strictly depending on the modelling accuracy and may be different from typical Oil&Gas pipeline corridors. Further studies are required including more detailed consequences modelling approaches and the effect of the simultaneous gas pipeline presence.

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