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Study of the Vaporization Rate of Liquid Nitrogen by Small- and Medium-Scale Experiments

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Potentially severe consequences associated to accidental liquefied natural gas (LNG) spills have motivated a number of different studies, including experimental work, towards their prediction. Most of these studies focus on vapour dispersion, however there is limited information regarding source term of vapour formation, which includes liquid spill, pool spreading and liquid vaporization rate.

The present work investigates the contribution of different heat transfer mechanisms to the vaporization rate of cryogenic liquid through the series of small and medium-scale, well-controlled and instrumented experiments performed with liquid nitrogen (LN_2). The vaporization rate of LN_2 was measured and correlated to the individual contributions of conductive, convective and radiative heat transfer.

The experiments on convection showed that this heat transfer mode can play a significant role in the vaporization rate of the cryogenic liquid. In these set of experiments, its contribution to total heat flux to an LN_2 pool spilled on a concrete surface was measured to be as high as 30 % after 10 min for moderate wind speed of 2-3 $m.s^{-1}$. The experimental data also showed that the sidewalls of the liquid containment may play a key role in the resulting amount of convective heat transfer and indicates that walls or fences could be used as a good mitigation method.

1. Introduction

For cryogenic liquid releases, like LNG, the vaporization rate of the formed liquid pool is controlled by the heat transfer from the ground, from radiation and convection. Experimental data on the vaporization rate of LNG resulting from these different heat transfer modes are very limited. The existing models for source term calculation have multiple limitations and thus there is a significant amount of work to be done in both the experimental and modelling fields (Webber et al., 2010).

Early experimental analysis on LNG spill on land indicated that, at least at the early stages of the spill the vaporization rate is mainly governed by conductive heat transfer from the solid surface to the liquid (Burgess and Zabetakis, 1962). Subsequently, models assuming this heat transfer mode is always the dominant one were developed (Briscoe and Shaw, 1980). Such models assume perfect thermal contact between the pool and the ground and one-dimensional heat conduction. They also require the knowledge of the ground heat transfer characteristics, which can dependent on several factors such as moisture content of the ground.

Another approach when dealing with heat transfer from the ground would consist on taking into account the different boiling regimes resulting from the formation of bubbles on vapour on the liquid ground surface. Indeed, a large temperature difference between the liquid and the ground would exist at early stages of the spill which will induce vigorous boiling and generate a vapour film at the liquid-ground interface and then limit the heat transfer. Three boiling regimes are recognized: film, transition and nucleate. Complex heat transfer phenomena are involved in the boiling process. The prediction of heat transfer for each boiling regime for different types of ground and liquid mixtures still requires a substantial experimental and theoretical effort. Correlations can be found in the literature to calculate the heat transfer in such cases (Opschoor, 1975; Kalinin et al., 1976; Klimenko, 1981; Conrado and Vesovic, 2000).

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Other heat transfer mechanisms like convection and radiation are however often considered to be less crucial to model and are reported to count on average for less than 5 % of the total heat transfer to the pool (Cavanaugh et al., 1994; Webber et al., 2010; Woodward and Pitblado, 2012). Véchot et al. (2012) showed that convection would play a significant role in the pool boiling rate particularly at later stages (after 10 min) of the pool boiling.
It still remains necessary to measure and model the contribution of each of these heat transfer modes to the total vaporization rate of a cryogenic liquid in order to have a full picture of the governing mechanisms and subsequently develop and validate scientifically sound source term models able to predict the vaporization rate of the liquid at all stages of the spill.
The present study is a part of a research project that focuses on the development and validation of source term model related to LNG spill on land. Experimental work was performed at Texas A&M University at Qatar to investigate the contribution of each of these heat transfer mechanism to the vaporization rate of cryogenic liquid nitrogen (LN_2). This involved several laboratory, wind tunnel and field experiments. The work presented in this paper only covers conduction and convection at laboratory and in a wind tunnel. For the experiments LN_2 was used as a safer analogue of LNG. The investigations will be extended to LNG in the near future.

2. Experimental setup

Laboratory scale experiments include the tests in 7 litres Dewar flask in which the LN_2 vaporization rate was measured (by a laboratory balance) with individually controllable contributions from convective (generated by an electric fan) and thermal radiative (generated by light bulb) heat transfer in the presence of a baseline conductive heat transfer rate. The details of this work are given in Véchot et al. (2012).
Wind tunnel experiments were performed at the Fire Station 2 of the Ras Laffan Industrial City, Qatar, with the collaboration of Qatar Petroleum. They were designed and performed with the idea of scaling up the laboratory experiments with the Dewar flask and to extend the study on convective and radiation heat mechanisms to include conduction on different substrates. The wind tunnel experiments include the spills of LN_2 into two square shape boxes made of concrete or polystyrene (Figure 1).
The concrete box (0.5 m x 0.5 m x 0.4 m) was made of cement and fine sand in proportion of 1:4. The box was also equipped with 0.4 m tall lips made of a thin stainless steel sheet placed on the top of the concrete surface. The external surfaces of the sheet were insulated with a 15 cm wide layer of polystyrene. Two heat flux plates and 12 thermocouples were embedded into the concrete at different depths and locations and two heat flux sensors were installed in the walls to monitor a heat flux and temperature inside a pad and walls, respectively (these data are not shown in this paper). This box was used to measure the heat transfer by conduction mainly while limiting the effect of convection.
The polystyrene box (with containment size of 0.48 x 0.48 x 0.1 m and 0.15 m thick base and walls) was designed to study the effect of convection and radiation. The box is entirely made of polystyrene which tends to limits the heat losses by conduction. The box was equipped with embedded heat flux plates and thermocouples in its base. The heat losses via the box walls were measured. The box was also designed to allow the addition of walls of different height around the liquid nitrogen pool surface such allowing experiment with both bounded and unbounded pools.
During an experiment the box (concrete or polystyrene) was placed on a balance (maximum load = 300 kg and resolution = 10 g). The mass of the box was recorded every 1 second and used to measure the LN_2 vaporisation rate. The box itself was placed in a wind tunnel of 2.04 m wide, 0.855 m tall and 12 m long which was especially designed to isolate the box from natural wind and to ensure a controllable and stable airflow. A 1.2 m diameter variable speed electric fan was placed at the entry of the tunnel and the centre of the concrete pad was located 5.64 m from the outlet of the fan. The wind data were measured by two ultrasonic anemometers (81000, R.M. Young USA). An artificial radiation panel composed of nine 200 W bulbs evenly distributed above a containment box, which ensures uniform radiation level over the surface of the box. Sufficient distance between the bulbs and the box ensures that the radiation level was independent of vertical position of liquid level, since the changes of liquid level were very small during the experiment (less than 40 mm). Uniformity of radiation in a box was confirmed by measurements in blank test (without LN_2) using a solar radiometer (LP02, Hukseflux USA) and the average value was taken for subsequent analyses. The ambient temperature was taken in several locations of the surroundings. The air humidity was measured as well. All the latest mentioned sensors were manufactured by OMEGA and the data were recorded with acquisition system supplied by IOtech.
During the wind tunnel experiments, LN_2 was discharged into the boxes at a certain flow rate which was not measured. However the moment when the liquid touched the surface was monitored by

thermocouples. The vaporization rate of liquid nitrogen was analysed after the spill was stopped (thus the measurements for the beginning of the spill are missed since the flow rate of spill was not known).
Finally, field experiment was also performed in a concrete box with a square surface of 0.9 x 0.9 m, and with maximum containment of about 173 L of liquid. The results will be published in a future paper.

3. LN_2 vaporisation due to conduction

The conductive heat transfer from the concrete ground to the cryogenic liquid pool has been investigated by two set of small scale wind tunnel experiments. The experiments aimed to study the effect of heat conduction through the concrete to the liquid pool and investigation of its magnitude, which can be subsequently used as comparison to convective and radiative heat transfers.
For these experiments the liquid containment was covered with polystyrene lid to eliminate convective and radiative heat transfer to the pool. The measured liquid vaporization rate is directly proportional to the total heat flux to the pool, which includes the conduction through the ground (concrete base) as well as conduction through the walls and lid. The latest were monitored with heat flux sensors placed inside and on top (the latest also measured convective heat from surrounding air) of walls and lid. The conductive heat flux from the concrete to the liquid pool was obtained by subtracting the heat flux through the walls and the lid from the total heat flux.
The result, conduction through the concrete base is shown in Figure 2a (dotted line). The linear behaviour of heat flux (q) to the inverted square root of time ($time^{-0.5}$) has been observed (points in Figure 2b). This behaviour can be modelled with ideal conduction theoretical model.
Assuming 1-dimensional conduction from the semi-infinitive ground with perfect contact of cryogenic liquid with the ground surface and thus constant temperature at the boiling point of liquid, the boiling rate is proportional to the inverted square root of time (Carslaw and Jaeger, 1986) as follows:

$$q=\left(\frac{k\rho c_p}{\pi}\right)^{1/2}\left(T_{amb}-T_{liq}\right)\cdot t^{-1/2}=A\cdot t^{-1/2} \tag{1}$$

where A (in kJ $s^{-0.5}m^{-2}$) can be determined experimentally. This value can be also calculated theoretically if the properties of the concrete are known and independently measured. Carslaw and Jaeger (1986) published the values for concrete with aggregate (1:2:4) as shown in Table 1. With these properties and using the liquid nitrogen boiling point (77 K) and the ambient temperature of 306 K (as measured on experimental day), A would be equal to 182 kJ $s^{-0.5}m^{-2}$.
The experimental data shown in Figure 2 were well fitted with Equation (1) and the value of A was estimated at 135.2 kJ $s^{-0.5}m^{-2}$.
The experimental value of A (Table 1) is of the same order of magnitude but smaller than the one from the literature. This may be due to the fact that the thermal properties of the concrete used in the experiment (not independently measured) were different from the ones from Carslaw and Jaeger (1986) due to the difference of composition of the concrete. However, it may be the effect of additional resistance at the concrete surface due to vapour film or large bubbles formation. Regardless, the differences between the experimental result and theoretical value were about 25 %. This stresses the importance of independently measure the thermal properties of concrete before using the approach above and to draw final conclusions. It is worth to note that Reid and Wang (1978) obtained similar linear behaviour when measuring LNG boiling rates over different types of substrates like sand insulated concretes, sand with less than 4 % of water, soil with less than 8 % of water and polyurethane, at least at early stages of the experiments. Corrugated aluminium showed a different behaviour and where not properly represented by the model above.

4. LN_2 vaporisation due to convection

Véchot et al (2012) reported experimental work at laboratory scale on the quantification of the vaporization rate of liquid nitrogen under forced convection. They showed that values of heat transfer by forced convection are comparable to predicted values of heat transfer by conduction particularly at the later stages of the spill and should not be ignored. They also highlighted the role of the side walls of the liquid pool containment device (dikes or impoundments) in limiting the air motion above the liquid pool and therefore the heat transfer by convection.
Experiments in the wind tunnel were performed to measure the magnitude of the heat transfer by convection and compare it to the values of heat transfer from concrete as described above. Vaporization rate were measured under nearly no wind conditions (< 1 m/s and turbulent intensity > 40 %, due to the

natural air motion in the wind tunnel) and under forced convection (using a fan) resulting in moderate wind speed of 1.4-3 m.s^{-1} (with turbulent intensity at about 15 %).

Experiments were also performed to measure the effect of walls around the liquid pool surface on the vaporisation rate (either because of a decrease of the initial pool level or because of the addition of polystyrene walls on the top of the box). The height of the walls around the pool was quantified as the ratio δ/D, which is corresponds to the distance between the top of the wall to the liquid surface (δ) over characteristic length of the pool (D, dimension of the side of the pool).

Figure 3 shows that the measured of convective heat flux from these experiments. As expected, the higher the wind speed the higher the amount of heat transferred by convection. The convective heat flux as high as 3 kW m^{-2} were measured with unbounded pool (low δ/D) for moderate wind speed close to 3 m s^{-1} (Figure 3a). This value is equal to about 50 % of conductive heat flux at 10 min of liquid nitrogen spill on concrete (which is about 6 kW m^{-2}, Figure 2a), so 30 % of the total heat flux to the pool. It's worth noting that the range of wind speed investigated cannot be considered as particularly high. These experimental results confirm that the contribution of convection to the total vaporization rate of cryogenic liquid spilled on the ground should not be ignored and may account for more than 5 % of the total heat transfer to the pool as reported in the literature (see above).

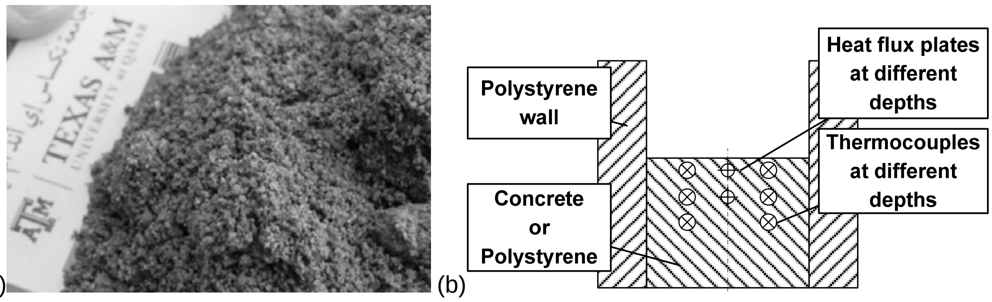
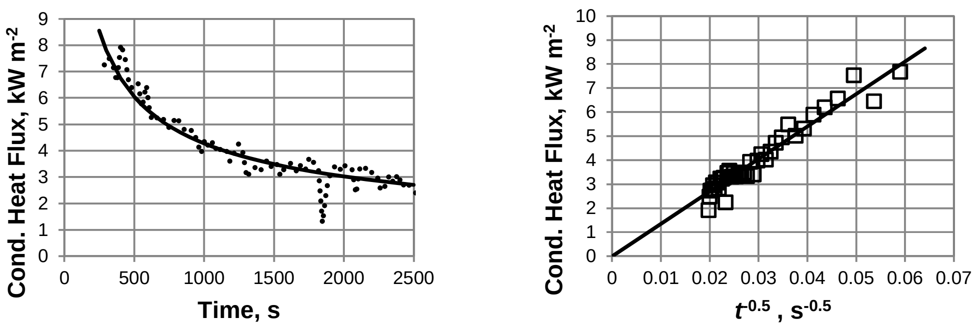
(a)


Figure 1. The picture of the concrete powder (a) and the general scheme of the experimental pad (b)



(a) (b)

Figure 2. Experimental data of conduction and model fitting (solid line): (a) timeline; (b) linear behaviour of heat flux to inverted square root of time

Table 1: Comparison of parameter A from equation (1) to theoretical calculation

	k, W/(m K)	ρ, kg m^{-3}	C_p, kJ/(kg K)	A, (kJ s$^{-0.5}$ m^{-2})
This work	n/d	n/d	n/d	135.2
Theoretical calculation Carslaw and Jaeger (1986)	0.92	2243	0.963	182

n/d - not determined

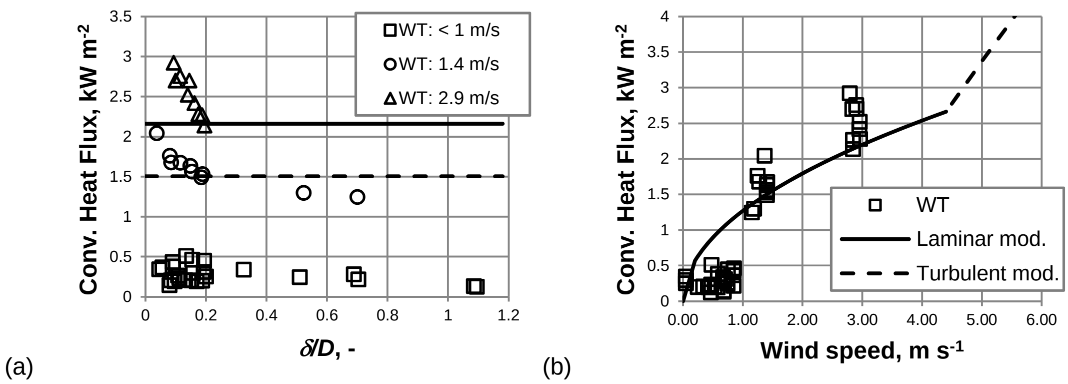


Figure 3: Validation of modelling (solid and dotted lines) with experimentally determined convective heat flux to the pool: (a) The bund walls height effect; (b) wind speed effect

Figure 3 also shows that higher sidewalls around the liquid surface tend to decrease the heat transfer by convection. The convective heat transfer is noticeably sensitive to δ/D for values of δ/D between 0 and 0.20 for wind speeds higher than 1 m/s. This may be related to the turbulences as the high walls hold a layer of vapour and limit air motion directly above the liquid surface. This greatly limits the heat transfer by convection and indicates that walls or fences could be a good mitigation method to limit the vaporization rate of a pool of cryogenic liquid. For wind speed lower than 1 m/s the results show measured values of convective heat flux between 0.2 and 0.5 kW m^{-2}, with a low sensitivity to δ/D. Over the range of investigated wind speeds, an increase of δ/D above 0.4 does not seem to significantly induce further limitation of the heat transfer by convection (Figure 3a).
The experimental results were compared to convective heat transfer rates predicted by forced convection correlations for an isothermal flat plate of characteristic length *D* as follows (Woodward and Pitblado, 2012):

$$q_{convection} = \frac{Nu \cdot k}{D}\left(T_{amb} - T_{liq}\right) \tag{2}$$

$$Nu_{\text{turbulent}} = 0.664\,\text{Re}^{1/2}\,\text{Pr}^{1/3}\,;\; Nu_{\text{laminar}} = \left(0.037\,\text{Re}^{4/5} - 562\right)\text{Pr}^{1/3} \tag{3}$$

As shown on Figure 3 the application of the simple, above correlations for wind speed of 1.4 and 2.9 m/s will tend to underestimate the convective heat flux for low values of δ/D (< 0.2). It will be worth testing such correlations at larger scale for fully developed turbulent flow above the liquid pool.
A deeper analysis of the wind profile in the tunnel above the pool is necessary to fully understand and predict the effect of convective heat transfer on the overall vaporization rate. Computational Fluid Dynamic modelling of the air motion above the liquid surface for bounded or unbounded pool would certainly help in this respect. The approach should also be complemented with additional larger scale experiments and modelling in order to recommend optimal wall's height around impoundment areas containing spilled cryogenic liquids. This will be done in the future within this research program.
The experiments also investigated the effect of radiation on the vaporisation rate. The results were similar to the ones obtained at laboratory scale (Véchot et al, 2012) and showed that only 30-65 % of incident radiation from the bulb (which was about 0.5 kW m^{-2}) reached the liquid pool.

5. Conclusions

The work presented in this paper investigates the contribution of conductive and convective heat transfer mechanisms to the vaporization rate of a cryogenic liquid through the series of small and medium-scale experiments performed with LN_2. The experiments on conduction showed that the heat transfer from a concrete substrate to the LN_2 pool was reasonably well represented by a simple heat transfer conduction model assuming perfect thermal contact between the liquid and the ground surface.
The experiments on convection showed that this heat transfer mode can play a significant role in the vaporization rate of the cryogenic liquid. In these set of experiments, its contribution to total heat flux to an LN_2 pool spilled on a concrete surface was measured to be as high as 30 % after 10 min for moderate wind speed of 2-3 m s^{-1}. The experimental data also showed that the sidewalls of the liquid containment may play a key role in the resulting amount of convective heat transfer. This is attributed to the fact that the

high walls hold a layer of vapour and limit the air speed above the liquid surface. This indicates that walls or fences could be a good mitigation method to limit the vaporization rate of a cryogenic liquid pool.
Future work will include the study by CFD modelling of the air motion above the liquid surface for bounded or unbounded pool and the resulting vaporization. Finally, a series of medium scale field experiment of LNG spill on concrete ground under various weather conditions will be done at the state of the art LNG training facility currently developed by Qatar Petroleum at the Ras Laffan Emergency and Safety College (Doha, Qatar) to support the research on LNG source term modelling.

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Nomenclature

A	parameter in the equation (1), kJ $s^{-0.5}m^{-2}$
D	characteristic length of the pool, m
q	heat flux, kW m^{-2}
t	time, s
k	thermal conductivity, W m^{-1} K^{-1}
ρ	density of the substrate, kg m^{-3}
C_p	heat capacity of the substrate, J kg^{-1} K^{-1}
Nu	Nusselt number
Pr	Prandtl number
Re	Reynolds number
T_{amb} / T_{liq}	ambient / liquid nitrogen temperature, K
δ	distance between the top of the wall to the liquid surface, m

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