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Numerical Simulation of Leakage and Diffusion Process of LNG Storage Tanks

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Abstract

Background

The previous researches mainly focused on the potential hazards associated with LNG leaks and the level of the influence of external environmental factors on the dispersion effect of LNG spills. Few considerations were given to phase change. Therefore, in order to investigate the evolution process of LNG liquid pool and gas cloud diffusion, the effect of phase change on dispersion during LNG release is studied to analyze the behavior characteristics of LNG liquid pool expansion and gas cloud diffusion, and the effect of the leaking aperture on the gas cloud diffusion process is also studied.

Methods

The Eulerian model and Realizable $k-\varepsilon$ model were used to numerically simulate the liquid phase leakage and diffusion process of LNG storage tanks. The homogeneous Eulerian multiphase model was adopted to model the phase change process after LNG leaks to the ground. The Eulerian model defines that different phases are treated as interpenetrating continuum, and each phase has its own conservation equation. The average diameter of LNG droplet and NG bubble were set to 0.01m. The standard $k-\varepsilon$ model and realizable $k-\varepsilon$ model are commonly used to describe turbulent motion. However, the realizable $k-\varepsilon$ model can not only effectively solve the problem of curved wall flow, but also simulate free flow containing jets and mixed flows. In addition, the realizable $k-\varepsilon$ model had higher accuracy in concentration distribution by simulating Thorney's heavy gas diffusion field test. Therefore, the realizable $k-\varepsilon$ model was selected for gas diffusion turbulence.

Results

The diffusion of the explosive cloud was divided into heavy gas accumulation, entrainment heat transfer and light gas drift. The vapor cloud gradually separated into two parts from the whole "fan leaf shape". One part was a heavy gas cloud, the other part was a light gas cloud which spread with the wind in the downwind direction. The change of leakage aperture had a greater impact on the whole spill and dispersion process of the storage tank. The increasing leakage aperture would lead to 10.3 times increase in liquid pool area, 78.5% increase in downwind dispersion of methane concentration at 0.5LFL, 22.6% increase in crosswind dispersion of methane concentration at 0.5LFL and 249% increase in flammable vapor cloud volume. Within the variation range of the leakage aperture, the trend of the gas cloud diffusion remains consistent, but the time for the liquid pool to keep stable and the gas cloud to enter the next diffusion stage was delayed. The low-pressure cavity area within 200m of the leeward surface of the storage tank will accumulate heavy gas for a long time, forming a local high concentration area.

Conclusion

Within the variation range of leakage aperture, there will always be a local high concentration area within 200m downstream of the storage tank. In the field near the storage tank, the clouds settle and accumulate towards the ground in the state of gas-liquid two-phase flow, and the density of the cloud is gradually lower than the air in the far field, manifesting as light gas diffusion. The methane concentration in this area is high and lasts for a long time, so it should be the focus area of alarm prediction.

Keywords: LNG leakage and diffusion; Combustible cloud; Phase change; Plume flow; Leakage aperture

1. Introduction

Liquefied Natural Gas (LNG) is mostly methane with small amounts of ethane, propane,

butane and nitrogen. It is expected to be the second largest energy source in energy composition in 2030^[1]. However, liquefied natural gas has exposed many safety problems in terms of LNG leakage and vapor explosion. Scholars in China and overseas have conducted many studies on the prediction of possible hazards associated with LNG vapor dispersion. Koopman et al.^[2] carried out the Burro series of tests in 1980 to observe the diffusion of LNG vapor clouds under different conditions after LNG leaked to the water surface. It was found that the diffusion behavior of the vapor cloud was affected by the way of LNG spill. In 1983, the Coyote series of test^[3] was conducted to study the ignition and flash evaporation processes of LNG. The rapid phase transition, vapor cloud diffusion and pool fire were all observed in this test. Brown et al.^[4] carried out Falcon series of experiments to study the leakage and diffusion of LNG under obstacle conditions, accurately evaluating the effectiveness of the fence to mitigate the harm of LNG gas cloud diffusion. In addition, Several mathematical models have been developed to simulate heavy gas diffusion based on experimental data, such as DEGADIS, SLAB^[5], FEM3^[6,7], etc.. Field tests can reproduce the actual situation of LNG leakage and diffusion, However, the cycle is too long and the repeatability is poor. Thus, CFD simulation is used as a promising alternative to calculate the diffusion distance of LNG. Giannissi et al.^[8] simulated the LNG diffusion in an open and obstructed environment based on Falcon series experiments. It was verified that the leak source model greatly affected the diffusion of LNG, and the best case to simulate the leakage source was to model the source as two phase. Vílchez et al.^[9] used DEGADIS model to predict the explosion distances of vapor cloud after LNG leakage and defined the diffusion safety factor (DSF) to estimate these distances. Li et al.^[10] evaluated the effect of safety clearance on the diffusion of cylindrical floating LNG through FLACS software.

The result demonstrated that the safety gap increased the size of the gas cloud far from the cylindrical FLNG release position, and decreased the size of the gas cloud near the release position. Zhang et al.^[11] studied the process of LNG leakage and diffusion in different wind directions. The result showed that the LNG spread farthest along the horizontal downwind direction. Marsegan et al.^[12] carried out numerical simulation of LNG diffusion under active and passive barriers, founding that the active barrier effectively reduced the diffusion range of LNG by accelerating the entrainment between air and gas. Nguyen et al.^[13] conducted a liquid pool evaporation experiment with different leak rates on the water surface. They proposed a model to express the function relationship between evaporation rate, leakage rate and time based on the experimental results and one-dimensional heat conduction model. Gopalaswami et al.^[14] developed a transient three-dimensional multiphase model in CFX based on the comprehensive test data and numerical simulation data, and found that wind affected the evaporation and diffusion of LNG by carrying additional heat and unsaturation. Ikealumba et al.^[15] studied the effects of atmospheric and ocean stability on LNG diffusion. They found that the instability caused by the waves would aggravate the leakage hazard of LNG ships. Luo et al.^[16] proposed an integrated multiphase CFD model to simulate the complete process of LNG leakage on the water surface. The study found that water storage would shorten the horizontal diffusion distance of the gas cloud. Dasgotra et al.^[17] simulated the diffusion of heavy gas in natural gas storage facilities. They found that the average diameter of gas cloud ranged from 0 to 500 m under relatively stable weather conditions. Giannissi et al.^[18] investigated the effect of environmental humidity on the diffusion of LNG, and it was concluded that in the case of high environmental humidity, the explosion distance of gas cloud would be reduced.

The above researches mainly focused on the potential hazards associated with LNG leaks and the level of the influence of external environmental factors on the dispersion effect of LNG spills. Few considerations were given to phase change. Therefore, the effect of phase change on dispersion during LNG release is studied to analyze the behavior characteristics of LNG liquid pool expansion and gas cloud diffusion, and the effect of the leaking aperture on the gas cloud diffusion process is also studied.

2. Materials and Methods

2.1 Numerical model

The homogeneous Eulerian multiphase model was adopted to model the phase change process after LNG leaks to the ground. The Eulerian model defines that different phases are treated as interpenetrating continuum, and each phase has its own conservation equation. The average diameter of LNG droplet and NG bubble were set to 0.01m.

The process of LNG leakage takes place in open air space, so the flow and diffusion process of gas is greatly affected by atmospheric motion. The standard k - ε model and realizable k - ε model are commonly used to describe turbulent motion. However, the realizable k - ε model can not only effectively solve the problem of curved wall flow, but also simulate free flow containing jets and mixed flows, which has obvious advantages compared with the standard k - ε model. In addition, the realizable k - ε model had higher accuracy in concentration distribution than the standard k - ε model by simulating Thorney's heavy gas diffusion (Freon-12) field test^[19]. Therefore, the realizable k - ε model was selected for gas diffusion turbulence.

The turbulent kinetic energy k equation is as follows.

$$\frac{\partial(\rho k)}{\partial t} + \frac{\partial(\rho k u_i)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[\left(\mu + \frac{\mu_t}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + G_k + G_b - \rho \varepsilon - Y_M + S_k \quad (7)$$

The turbulent dissipation rate ε equation is as follows.

$$\frac{\partial(\rho\varepsilon)}{\partial t} + \frac{\partial(\rho\varepsilon u_i)}{\partial x_i} = \frac{\partial}{\partial x_j} \left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon} \right) \frac{\partial \varepsilon}{\partial x_j} \right] + \rho C_1 E \varepsilon - \rho C_2 \frac{\varepsilon^2}{k + \sqrt{\nu \varepsilon}} + C_{1\varepsilon} \frac{\varepsilon}{k} C_{3\varepsilon} G_b + S_\varepsilon \quad (8)$$

2.2 Parameter setting

A $16 \times 10^4 \text{ m}^3$ large cylindrical LNG storage tank was chosen for the simulation. The outer diameter of the tank is 82 m and the height is 50 m. The structural dimension of the tank is shown in Fig.1. The normal operating pressure of the storage tank was 25kPa, and the maximum liquid level in the tank was 34.6m. The origin of the computational domain was located at the center of the bottom of the tank. The coordinate of the leakage hole center point was (41, 10, 0), which was located on the leeward side of the tank. The leakage hole sizes were respectively 0.1×0.1 m, 0.13×0.13 m, 0.15×0.15 m, 0.18×0.18 m and 0.2×0.2 m. Considering the calculation accuracy, the computational domain was determined to be 1000m × 250m × 500 m in the x , y and z directions, and the tank with a blocking rate of 2.78% was placed at a distance of 200 m downwind. The whole computational domain was discretized by structured grid, and the specific grid division was shown in Fig. 2. In order to adapt to the change of flow field and ensure the accuracy of solution, the grid around the leakage hole was encrypted by block method. The independence of grid and time step had been verified. Overall, the total number of cells in the calculation domain was finally determined to be 1,865,345, and the simulation time step was set to 0.1s.

Fig.1 The geometry and boundary settings of the large-scale LNG storage tank. a: geometric schematic of the tank; b: the boundary settings of the tank

Fig. 2 Meshing of computational watershed

The gas flow was modeled by solving the mass, energy and momentum equations of each phase as well as the heat and mass transfer equations on the interface. The drag force, lift force and virtual mass force of each phase were considered in the model, which had a great influence on the movement of particles between phases^[20]. It required complex equations to be solved for processing LNG into a mixture of different components , which usually increased the calculation difficulty and thus caused a deviation in the calculation. Therefore, LNG was regarded as pure liquid methane with a volume fraction of 100%. At the initial moment, the boiling temperature and the air temperature were respectively 111.6K and 300K. After the low-temperature liquefied natural gas exchanged heat with the air, the temperature of methane raised. Due to the variation of the density, viscosity, specific heat capacity and heat transfer coefficient of methane with temperature, the above parameters were converted into a function with time by referring to the corresponding empirical formula^[21-24].

In order to represent the node coordinates more accurately and ensure the convergence of calculation, double precision solver and implicit method were used in the calculation. Couple algorithm was adopted for pressure and speed coupling. And the momentum and energy equations were discretized by the second order upwind scheme. Fig. 3 shows the meshing of LNG leakage diffusion experiment. The calculation domain was established with the size of $900\text{m} \times 500\text{m} \times 50\text{m}$ on the X-axis, Y-axis, and Z-axis respectively. The x - z plane was placed on the ground and the y -direction was the vertical height. Furthermore, the wind direction remained unchanged throughout the calculation domain. The boundary conditions on the left and right sides of the calculation domain were the velocity-inlet and the pressure-outlet, respectively. Hexahedral mesh units were used for mesh generation, while the area around the pond was divided into fine meshes. A total of 803,287 cells were used for subsequent simulations.

Fig.3 The meshing of LNG leakage diffusion experiment

3. Results and Discussions

3.1 Model validation

In this paper, data from the Burro 8 spill test^[25] which conducted in 1980 was

used as the basis of the validation analysis. In the test, LNG was released onto the water surface of a round pond, with 25 gas concentration monitors placed at different heights in the downwind. Besides, the water pond had an average diameter of 58 m, with an average water level about 1.5 m below the surrounding ground level. The basic data obtained from this series of tests were good, which were often used for model verification. Based on Burro series tests, the reliability of the multiphase model was evaluated by comparing the numerical results with the experimental results based on the diffusion range and concentration change of methane. Fig.4 and Fig.5 show the contour distribution of methane volume fraction after LNG spill 80 s on the $x = 57\text{m}$ and $y = 1\text{m}$ planes, respectively. In Fig. 4(a), (b) and Fig. 5(a), (b), the distribution areas of methane with different volume fractions on the horizontal and vertical planes were basically consistent with the experimental data. And Fig.4(c) and Fig.5(c) show the comparison of the coverage areas of dispersion clouds with different volume concentrations. There is a very good quantitative agreement between the simulation results and the experimental data. Besides, Table1 shows the comparison between the calculated and experimental values of maximum volume fraction of methane at different distances in downwind direction. It showed that the calculated maximum volume fraction of methane was lower than that of the experiment, however, in the area away from the leakage source, the calculated maximum volume fraction of methane was higher than that of the experiment. The reason was that the coupled heat transfer between the ground and the LNG vapor cloud was assumed to be constant in the simulation. Actually, the heat produced by ground heat transfer and solar radiation was variable. The error analysis method of heavy gas diffusion model proposed by Emark et al. ^[26] was used to analyze the deviation between the

value of simulation and test. The method includes relative deviation (FB), geometric mean deviation (MG), geometric mean deviation (VG), relative mean square error (MRSE), relative mean square error (FAC2) and normalized mean square error (NMSE), which can be used to judge the validity of the numerical model. The deviation between numerical and experimental values is shown in Table 2. It could be seen that all deviation were within the range allowed by the evaluation parameters. Therefore, the multiphase model is suitable for the study of LNG leakage and diffusion.

Fig.4 Comparison of experimental and simulated values of methane volume concentration at a vertical height of 1 m

Fig.5 Comparison of experimental and simulated values of methane volume concentration at 57m in downwind direction

Table 1 Experimental and simulated values of maximum volume fraction of methane at different distances in downwind direction

Downwind distance/m	Maximum methane volume fraction at 1m height/%	
	Test measured value	Fluent simulation value
140	16.49	15.4
400	4.25	5.32

800	1.93	2.25
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Table 2 The error comparison of simulation results

Deviation statistics	FB	MG	VG	MRSE	FAC2	NMSE
Ideal value	0	1	1	0	1	0
Evaluation standard	(-0.4,0.4)	(0.67,1.50)	<3.3	<2.3	>0.5	<4
Burro 8	-0.18	0.88	1.03	0.04	0.87	0.23

3.2 Basic characteristics of LNG storage tank leakage and diffusion

3.2.1 Simulation of LNG storage tank wind field

LNG storage tank will obstruct the flow of wind speed and affect the diffusion of LNG. In this study, the average wind speed at the height of 10 m was 4 m/s, and the wind speed of inflow profile was implemented in a user-defined function (UDF) which was embedded in the numerical model as the boundary condition. Fig. 6 shows the wind speed distribution of different planes in the calculation domain. As shown in Fig.6(a), the wind speed at the boundary of the entire wind field was evenly distributed in the vertical plane of 30m. And the wind speed varied with height to form gradient wind, which was the same as the wind field distribution law of the real atmospheric environment. However, the atmospheric flow near the storage tank was affected, resulting in changes in wind speed and direction. When the wind flowed from the top and both sides of the storage tank, it caused a high wind speed zone with the speed of 7m/s on top of the storage tank (shown in the black box, Fig.6) and a low wind speed zone with the speed of less than 1m/s on both sides of the storage tank(shown in the red box, Fig.6). In Fig.6(b), in the area away from the storage tank, the wind speed was maintained at 4m/s, however, in areas near the storage tank, the wind speed was reduced due to obstruction. A detention zone was formed on the windward side of the tank due to the obstruction of the tank, and the wind speed decreased sharply. When the wind bypassed both sides of the tank, a symmetrical bifurcated flow wake of a

certain length was formed in the downstream of the tank (shown in the red circle).

Fig.7 shows the distribution of wind speed streamline near the storage tank. It could be seen that there were vortices on the windward and leeward sides of the tank. Besides, two symmetrical vortices were formed at 70m in the x -axis behind the horizontal of the tank after the atmosphere bypassed the tank (Fig.7, a). In the process of the wind flowing downstream along both sides of the tank, the wind speed decreased continuously and the wind direction changed to produce backflow. When the wind moved to the central axis of the storage tank, its speed was close to zero, and a small cavity zone was formed on the back of the storage tank (Fig.7,b). However, the vortex and low wind speed areas were very close to the storage tank. When the wind was away from the storage tank, the streamline returned to normal and the wind movement also stabilized.

Fig.6 The wind speed in calculation area

Fig.7 The distribution of wind speed streamline near the storage tank

3.2.2 Liquid phase leakage diffusion process of LNG storage tank

The average wind speed was assumed to be 4m/s, and LNG leaked at arate of 105.5kg/s for

400s. The expansion of liquid LNG after leakage is shown in Fig.8. It can be seen that the pressure difference between the inside and outside of the tank caused the LNG to continuously spray from the leakage port to the ground in the form of parabola. The amount of LNG leakage was large, but the limited heat of the surrounding environment was hard to provide enough heat for the entire LNG to vaporize. Therefore, part of LNG absorbed heat from the surrounding environment and evaporated into a low temperature gas cloud, and others formed a liquid pool on the ground. During the landing process, part of the atomized LNG droplets absorbed heat from the air and evaporate into a gas state, resulting in a higher concentration of LNG leaking from the leakage hole and a lower concentration of LNG in the surface liquid pool (Fig.8, c). Under the action of initial kinetic energy and gravity, the liquid LNG diffused around the landing point which was 7m away from the storage tank to form a thin "round" liquid pool (Fig.8, b).

(a) Three-dimensional view of the liquid pool (b) Expansion of liquid pool at Y=0m (c) LNG injection at Z=0m

Fig.8 The distribution of LNG liquid pool

Fig.9 is a three-dimensional perspective view of gas clouds with different methane volume fractions at different leakage moments, showing the movement and diffusion process of LNG low-temperature steam cloud with leaking. At the initial stage of leakage, the density of the low-temperature vapor cloud formed by flash evaporation was greater than that of the surrounding air, so the height of gas cloud with the volume fraction of methane greater than 1%,

5% and 15% were extremely low due to the gravitational settling. As the leakage time increased to 120s, the gas cloud with a volume fraction greater than 15% was still close to the ground with a "hole" inside, while the gas cloud with a volume fraction greater than 1% and 5% rose slightly. When the leakage time reached 320s, the whole gas cloud presented the phenomenon of "leaf like bifurcation" on both sides. However, gas cloud with volume fraction above 15% and 5% were of low height, while the height of gas cloud with volume fraction above 1% was relatively high, with a large amount of light methane floating over the tank (shown in the red box). The whole process of diffusion change fully reflected the accumulation of LNG in the form of heavy gas cloud after leakage, and the mixing with air to absorb and transfer heat, resulting in the gradual narrowing of the difference between gas cloud density and air density, and finally the transformation of heavy methane into light methane in the periphery of the gas cloud.

Fig.9 Three-dimensional perspectives of gas clouds with different methane volume concentrations at different leakage moments

In order to reveal the spatial distribution characteristics of the LNG vapor cloud near the storage tank, methane concentration contours were selected from the xy plane, xz plane, and yz plane for analysis. Considering that the low height of the gas cloud bifurcated gas cloud along

the z axis on both sides of the tank, $x = 57$ m, $z = 30$ m, $y = 0.5$ m were selected as the observation surface. Fig.10 shows the methane gas cloud concentration distribution under different planes. As shown in Fig. 10(a), at the plane $y = 0.5$ m, the overall shape of the gas cloud was "fan-shaped" (shown in white box), accompanied by a cavity with a radius of about 17 m on the back. High concentrations of methane were deposited on both sides of the cloud, while low concentrations of methane were distributed in the middle of the cloud. As the leakage time went on, the low concentration methane in the middle was preferentially diluted by air, resulting in a "hole" in the middle of the gas cloud (shown in white box). After the leak continued for some time, the "hole" area expanded from the middle to the tail, and the gas cloud split into two parts. One part was a heavy gas cloud, which was stacked behind the storage tank in the form of "leaf-like bifurcation" (shown in white box), and the other part was a light gas cloud (shown in a white round frame), spreading further with the wind. Throughout the leakage process, the gas cloud gradually developed from a complete "fan shape" to a front-end "leaf-shaped bifurcation. Due to the disturbance effect of the storage tank on the atmospheric movement, the detention zone and low wind speed region behind the storage tank restrained the downwind expansion of the middle part of the gas cloud to some extent. When the low temperature LNG vapor mixed with the atmosphere, the movement of the vapor cloud also diverged laterally along the streamline development at the back of the tank, resulting in a large amount of methane accumulation on both sides and forming a leaf-shaped bifurcation.

In Fig.10(b), it can be seen that the gas cloud is divided into different concentration layers along the vertical direction at plane $z = 30$ m, and the methane volume fraction decreased with height. Among them, the methane concentration near the ground was high (shown in white box),

and the methane concentration far away from the ground was low(shown in white round frame). The reason was that a large amount of highly concentrated methane accumulated near the storage tank during the leakage process, which was difficult to dilute and dissipate. However, the heavy methane in the outermost part of the gas cloud continuously absorbed and transferred heat with air to form light methane with low concentration, and then spread to higher and farther places. In Fig.10(c), the gas cloud after leakage was symmetrically distributed behind the storage tank at 57m on the x direction. With the increase of leakage time, the width and height of vapor cloud in this area increased slightly. The vapor cloud appeared as "low in the middle and high at both ends" (shown in a white circle).

According to the results of numerical simulation and relevant heavy gas diffusion theory^[27], the macroscopic diffusion behavior of LNG vapor cloud could be roughly divided into three stages for the continuous leakage of LNG tank studied in this paper.

(1) Initial stage of diffusion (heavy gas accumulation): This stage was a period of heavy gas accumulation and diffusion. As shown in Fig.10, from the beginning of the leakage to 50s, the vapor cloud was in the shape of "fan leaf", and its internal concentration of the vapor cloud was in an unstable state. As methane concentration increased over time, the radial size of the vapor cloud increased, too. Due to the difference in density between the low-temperature gas cloud and the air, the heavy gas collapsed, making the height of gas cloud extremely low. In this stage, it was the turbulence caused by gravity collapse that played a dominant role in the shape and concentration distribution of the cloud, while the atmospheric turbulence played an auxiliary role.

(2) Mid-stage of diffusion (Transitional levitation): This stage was a period of transition

from heavy gas to light gas. From 120s to 160s, the development of gas cloud was in a neutral state and the whole gas cloud was still in a "fan leaf shape". The methane concentration inside the gas cloud increased to a peak. Meanwhile, when the wind bypasses the storage tank during the diffusion process, a lateral divergence was formed with the methane moving with the wind. High concentrations of methane accumulated on both sides of the vapor cloud, while lower concentrations of methane were distributed in the middle of the vapor cloud. When LNG vapor cloud exchanged heat with air and ground, the volume fraction of methane in the middle of gas cloud decreased rapidly, rising to tens of meters under the action of buoyancy. As the leakage time went on, the methane in the middle of the gas cloud is continuously diluted, resulting in a "hole" in the gas cloud. In addition, methane in the outermost part of the gas cloud was most affected by the wind, making the diffusion speed on both sides of the gas cloud significantly higher than the middle part, therefore, the vapor cloud presented the characteristics of "low in the middle and high on both ends".

(3) Post diffusion stage (Light gas drift): This was the period when light gas entered into passive diffusion. After 210s of leakage, the development of vapor cloud was in a stable state, the width of gas cloud remained unchanged, but the length and height of vapor cloud slowly increased. As the "hole" area inside the vapor cloud continued to expand, the contact area between the gas cloud and the surrounding air increased, which led to the rise of temperature and the decrease of methane density at the tail of the gas cloud. When the methane with higher concentration in the middle of the gas cloud was converted to light methane, the gas cloud split into two parts from the whole. One part was a heavy gas cloud, which was piled up behind the storage tank in the form of "leaf-shaped bifurcation". The other part was a light gas cloud, which

diffused with the wind in the downwind direction and finally entered the passive diffusion stage.

At the same time, under the influence of wind, methane in the outermost part of the cloud was still diluted the fastest, making the cloud still behave as "low in the middle and high at both ends".

(a) Distribution diagram of methane concentration at plane $y = 0.5$ m

(b) Distribution diagram of methane concentration at plane $z = 30$ m

(c) Distribution diagram of methane concentration at plane $x = 57$ m

Fig.10 Distribution of methane concentration in different planes

3.3 Effect of leakage aperture on leakage and diffusion of LNG storage tanks

3.3.1 Influence of leakage aperture on LNG liquid pool expansion

The effect of leakage rate was studied to further investigate the impact of leakage aperture on LNG liquid pool expansion and vapor diffusion. According to fluid mechanics, the leakage rate of liquid phase in storage tank can be calculated by equation (9).

$$Q_l = C_d A \rho \sqrt{2gh + \frac{2(P - P_0)}{\rho}} - \frac{\rho g C_d A}{A_0} t \quad (9)$$

Where, Q_l is the liquid phase rate and C_d is the liquid phase leakage coefficient which is taken as 0.6. A and A_0 are respectively the leakage hole area and liquid cross-sectional area in storage tank. Furthermore, since the leak area of the hole leakage is much smaller than the surface area of the tank, the time correlation term on the right side of equation (9) is zero. By maintaining all the other conditions the same, cases of leakage aperture of 0.1 m, 0.13 m, 0.15 m, 0.18 m and 0.2 m were tested in the multiphase model. The corresponding leakage rate are 62.45 kg/s, 105.54 kg/s, 140.51 kg/s, 202.34 kg/s and 249.81 kg/s, respectively.

Fig.11 shows the expansion diagram of the liquid pool of the storage tank after 180s leakage under different leakage apertures. It could be seen from the comparison that the LNG concentration and the liquid pool area on the ground both increased with the increase of leakage aperture. The instability of liquid pool expansion resulted in the unsmooth contour and irregular shape of liquid pool. However, the sensitivity of liquid pool expansion to different leakage apertures was also different. When the leakage aperture was less than 0.13 m, the area of the liquid pool which was tardy to the change of the leakage aperture increased slightly with the increase of the leakage aperture. When the leakage aperture was more than 0.13 m, the area of the liquid pool which was extremely sensitive to the change of the leakage aperture increased rapidly

with the increase of the leakage aperture.

Fig.12 was the comparison of the maximum diameter of the liquid pool with time under different leakage apertures. It can be seen from Fig. 12 that with the increase of leakage area, the growth rate of LNG liquid pool accelerated and the stabilization time to reach the maximum diameter also increased. For example, when the leakage aperture was respectively set as 0.1m, 0.15m and 0.2m, accordingly, the liquid pool reached a maximum diameter of 8.8m in 50s, 42m in 120s and 83m in 160s. It meant that the increase in leakage would change the pool area by affecting the heat transfer between the LNG and the ground. At the initial stage of LNG leakage to the ground, due to the large temperature difference between LNG and the ground, the heat exchange between the two was close to forced convection. LNG would quickly boil and evaporate, forming a continuous vapor film between the ground and LNG. Subsequently, the gas film broke due to the decrease of ground temperature, resulting in the transition boiling of LNG in direct contact with the ground. Finally, the heat transfer between LNG and the ground stabilized, and nuclear boiling occurred between LNG and ground. In this process, if the leakage rate increased, more bubbles would be generated to cover the ground, which limited the heat flux between the LNG and the ground, therefore, it cost more time to form a relatively stable boiling rate.

Fig.11 Expansion of liquid pool under different leakage holes

Fig.12 The change of liquid pool diameter with time under different leakage aperture

3.3.2 Influence of leakage aperture on LNG vapor cloud diffusion

Fig.13 shows the change in the morphology of LNG vapor cloud with time at 0.5m on the y= axis under five kinds of leakage apertures. When the leakage lasted for 60s which belong to the initial stage of diffusion, the vapor cloud was in the shape of "fan leaf" with the similar downwind diffusion speed under different leakage aperture. As the leakage aperture increased, the volume concentration of methane in the gas cloud kept rising, and the width of the gas cloud increased slightly. Compared with the situation at 60s, the gas cloud had different degrees of

holes inside at 180s which was at the middle stage of diffusion. However, the area of the hole in the gas cloud decreased with the leakage aperture increasing (shown in white box). When the leak lasted for 320 s, it reached the late stage of diffusion, the heavy gas in the vapor cloud was accumulated behind the storage tank in the form of "leaf like bifurcation", while the light gas at the tail of the vapor cloud was diluted with the wind. With the increase of the leakage aperture, the width of the heavy gas cloud became larger and the methane volume concentration of the light gas in the tail increased (shown in white round frame), which made it more difficult to be diluted. According to the LNG gas cloud diffusion under different leakage conditions, it could be demonstrated that the trend of the LNG vapor diffusion under different leakage apertures had similar characteristics. The change in the size of the leakage aperture would affect the coverage and concentration of the gas cloud, and thus delay the development of the gas cloud into the next diffusion stage. The motion trajectory of the vapor cloud was still determined by the wind field behind the tank.

Fig.13 Variation of gas cloud concentration distribution at $y=0.5\text{m}$ plane under different leakage apertures

As shown in Fig.14, the comparison of changes in methane volume fraction in the diffusion distance in the downwind direction at 100s, 200s and 300s leakage was made. Overall, the volume fraction of methane at all times was increased with the increase in leakage aperture at the same location, which was attributed to the increase of leakage per unit time. Besides, within the range of leakage aperture variation, the volume fraction of methane in the range of 200m from the storage tank to the downwind direction was the largest, which reached the peak value. In other words, when the LNG storage tank leaked, the vaporized LNG was preferentially stacked vertically within the range of 200m, causing the methane concentration to rise rapidly and to form a local high concentration area. The results indicated that the diffusion process of LNG met the theory of heavy gas accumulation. Due to the effect of gravity, the leaked LNG would first diffuse to the horizontal direction, which increased the concentration of methane in the horizontal direction and the drag. When the drag increased close to the cloud gravity, the horizontal diffusion velocity decreased. Heavy gas accumulated inside the soft diffusion boundary, causing methane concentrations to rise rapidly and stratify. As the leakage continued, when the air soft boundary cannot support the cloud gravity, the methane in the cloud continued to climb along the boundary layer, forming a new diffusion zone of light methane. This was why the area near the tank had a high concentration of methane, and the area far away from the tank had a low concentration of methane.

Fig.14 Variation of methane volume fraction with downwind diffusion distance under different leakage times

According to NFPA 59A, the distance where the concentration of natural gas was lower than 50% of the lower flammability limit could be regarded as the safety distance. Therefore, the area with the volume fraction of methane from 2.5% to 15% belongs to the explosion risk area. The maximum explosion range of methane and the change of combustible gas cloud volume with leakage time are shown in Fig.15. The increase of the leakage aperture would promote the diffusion speed of the vapor cloud in the downwind direction. As the leakage aperture increased, the maximum explosion range of methane and the volume of flammable clouds increased rapidly. For example, when the leakage aperture increased from 0.1m to 0.2m, the maximum diffusion distance of methane 0.5LFL in Fig.15(a) increased from 531m to 948m, with a growth rate of 78.5%, and the volume of flammable vapor cloud in Fig.15(c) enlarged from 13563.44m³ to

458 53642.89m³, with a growth rate of 295%. However, there was some difference, as shown in
459 Fig.15(b). When the leakage aperture was 0.1 m, the gas cloud with a concentration of 0.5LFL
460 had the largest width on the z - axis at 243m. When the leakage aperture increased from 0.13m to
461 0.2m, the largest width of methane 0.5LFL increased from 194.6m to 238.6m, with an increase
462 of 22.6%. This was because when the leakage pore size was 0.1m, due to the small leakage
463 volume, the methane density in the late stage of diffusion was close to the air and the gas cloud
464 diffused faster in the horizontal direction, resulting in the farthest diffusion distance of the gas
465 cloud along the z axis. As leakage aperture increased, the leakage and vaporization of LNG
466 increased, and a larger volume of combustible gas clouds increased too. However, the dilution
467 ability of air was limited, and the gas cloud rapidly accumulated and diffused along the
468 downwind distance, resulting in a larger diffusion distance along the x and z axes. Therefore,
469 after the LNG leaked, the leakage source should be cut off or blocked in time to reduce the
470 amount of LNG leakage.

Fig.15 Variation of the farthest moving distance and volume of flammable vapor clouds with leakage time under different leakage apertures

4. Conclusions

A three-dimensional numerical model was established to describe the leakage and diffusion process of LNG storage tank by using the realizable k - ϵ turbulence model and the Eulerian model. The conclusions were drawn as follows.

- (1) After the storage tank leaked, LNG is sprayed to the ground to form a circular liquid pool and continuously exchanges heat with air to evaporate into low-temperature steam. The diameter of liquid pool increases first and then remains unchanged with the leakage time, while the gas cloud diffusion state is divided into three stages due to the cylindrical turbulence of the tank. In these three stages, the LNG gas cloud experienced heavy gas accumulation, entrainment heat transfer and light gas drift, with the shape gradually developing from a complete "fan blade"

to a "leaf bifurcation" of heavy methane at the front end.

(2) Leakage aperture greatly affects the heat transfer between LNG and the surrounding environment. It delays the development of liquid pool and gas cloud to a stable state. In the five cases, heavy methane presented the characteristics of "leaf bifurcation". The increase of leakage aperture quantitatively affects the expansion process of LNG liquid pool and the distribution of vapor cloud across LNG dispersion routes. The liquid pool area is increased by 10.3 times, the length, the width and the volume of flammable vapor cloud is respectively increased by 78.5%, 22.6% and 249%. Besides, within the variation range of leakage aperture, there will always be a local high concentration area within 200m downstream of the storage tank. In the field near the storage tank, the clouds settle and accumulate towards the ground in the state of gas-liquid two-phase flow, and the density of the cloud is gradually lower than the air in the far field, manifesting as light gas diffusion. The methane concentration in this area is high and lasts for a long time, so it should be the focus area of alarm prediction.

Data Availability

All data, models, and code generated or used during the study appear in the submitted article. All data from top-down analysis are publicly available.

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508 **Ethics declarations**

509 Ethics approval and consent to participate

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513 Competing interests

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594 **Contributions**

595 Xue Li: review, drafting; numerical simulation; Bing Chen: review; Vamegh Rasouli: review,
596 language polish; Ning Zhou: review; Qian Zhang: data sorting; Xuanya Liu: modeling
597 optimization; Weiqiu Huang: model analysis and review
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Figures

Figure 1

The geometry and boundary settings of the large-scale LNG storage tank. a: geometric schematic of the tank; b: the boundary settings of the tank

Figure 2

Meshing of computational watershed

Figure 3

The meshing of LNG leakage diffusion experiment

Figure 4

Comparison of experimental and simulated values of methane volume concentration at a vertical height of 1 m (a)Burro 8 test measured value (b) Fluent simulation results (c) Comparison of test and simulation

Figure 5

Comparison of experimental and simulated values of methane volume concentration at 57m in (a)Burro 8 test measured value (b) Fluent simulation results (c) Comparison of test and simulation

Figure 6

The wind speed in calculation area (a) Cross wind direction $z=0\text{m}$ (b) Vertical height $y = 10\text{m}$

Figure 7

The distribution of wind speed streamline near the storage tank (a) Cross wind direction $z = 0\text{m}$ (b) Vertical height $y = 1.5\text{m}$

Figure 8

The distribution of LNG liquid pool (a)Three-dimensional view of the liquid pool (b) Expansion of liquid pool at Y=0m(c) LNG injection at Z=0m

Figure 9

Three-dimensional perspectives of gas clouds with different methane volume concentrations at different leakage moments

(a) Distribution diagram of methane concentration at plane $y = 0.5$ m

(b) Distribution diagram of methane concentration at plane $z = 30$ m

(c) Distribution diagram of methane concentration at plane $x = 57$ m

Figure 10

Distribution of methane concentration in different planes

Figure 11

Expansion of liquid pool under different leakage holes

Figure 12

The change of liquid pool diameter with time under different leakage aperture

Figure 13

Variation of gas cloud concentration distribution at $y = 0.5\text{m}$ plane under different leakage apertures

Figure 14

Variation of methane volume fraction with downwind diffusion distance under different leakage times

(a) Variation of the farthest diffusion length of 2.5% methane volume fraction (0.5LFL) with leakage aperture

(b) Variation of the farthest diffusion width of 2.5% methane volume fraction (0.5LFL) with leakage aperture

(c) Changes in the volume of flammable vapor cloud

Figure 15

Variation of the farthest moving distance and volume of flammable vapor clouds with leakage time under different leakage apertures