Experimental and Numerical Prediction Model for the Dangerous Radius of Natural Gas Leakage in Soil

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ABSTRACT: Prediction of the dangerous radius of natural gas plays an important role in reducing the hazards of a buried natural gas pipeline after leakage. The factors affecting the diffusion law of natural gas in soil after leakage are mainly divided into the pipe, soil, and environmental sides. Previous studies focused on the effects of leakage pressure, leakage aperture, and leakage direction on the pipe side and porosity and water content on the soil side. In this paper, experiments and numerical simulations are conducted for further investigating the effects on the diffusion of natural gas in soil of the soil type (porosity and granule diameter) and layered structure among the soil side factors and soil temperature as environmental side factors. The contour radius corresponding to 5% volume concentration (the lower limit of natural gas explosion in soil) is defined as the natural gas dangerous radius for analysis. Based on comprehensive analysis of the effects of the factors, a prediction model is proposed for the dangerous radius of natural gas in soil with leakage pressure, leakage aperture, porosity, and granule diameter as the dominant influencing factors, which is of great significance for locating the source of the leakage.

1. INTRODUCTION

With the continuous growth of natural gas consumption, buried pipelines, as the main transportation method of natural gas, may leak due to corrosion, aging, or human damage during transportation.¹,² Similar problems also exist in the subsea pipeline transportation.³⁻⁶ The complex soil environment around the pipeline makes it difficult to detect natural gas leakage in time, which may cause serious economic losses and casualties.⁷⁻¹¹

The dangerous radius of natural gas is related to the gas diffusion law in soil after the leakage of buried pipelines. According to the different influencing factors, it can be divided into pipe, soil, and environmental side factors.

Pipe side factors mainly include leakage pressure, leakage aperture, leakage direction, pipe buried depth, and so on.¹²⁻¹⁵ When the leakage pressure or leakage aperture is larger, the methane concentration in soil increases faster and the time for methane diffusion to reach the steady state is shorter.¹⁶,¹⁷ The leak diffusion law of methane in soil is the same when leaking in the horizontal direction (leftward and rightward), and it is of great difference for vertical (upward and downward) leakage with the maximum amount of methane released to the atmosphere when leaking upward.¹⁶,¹⁷ The increase of the pipeline buried depth will increase the diffusion distance of methane in the vertical direction, thereby increasing the time for methane to diffuse to the soil surface.¹⁸

Soil side factors mainly include porosity, granule diameter, water content, permeability, tortuosity, and so on. Among them, porosity, granule diameter, and water content directly affect permeability and tortuosity. In the soil, the diffusion rate of natural gas becomes faster, the time for diffusion to reach the steady state is shortened, and the gas concentration

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Increases when the porosity or granule diameter increases.\textsuperscript{19–21} The increase of water content will indirectly reduce the number of aeration pores in soil, which will impede the diffusion of methane and reduce its diffusion rate. Also, the uneven distribution of water in soil will also lead to irregular distribution of methane.\textsuperscript{22,23} During long distance transportation, natural gas pipelines pass through different geological structures, in suburban areas, of a porous media consisting of soil, gravel, and asphalt near towns, and of backfill, soil, gravel, and cement concrete or asphalt in towns. The diffusion of methane in the layered structure composed of sand, gravel, and asphalt has been investigated by large-scale experiments.\textsuperscript{24} It is found that the pressure, velocity, and concentration distributions of methane differed significantly from those in the single soil structure due to the difference in the diffusion rate in different porous media, and methane will accumulate near the surface due to the obstructive effect of asphalt. Because of the high population density in towns and cities, the hazards of natural gas pipeline leakage inside town roads are even greater, but there are fewer studies on the diffusion of natural gas inside town roads with backfill.

Environmental side factors affect the diffusion of methane both in soil and in the atmosphere. However, the current research mainly focuses on the effects on the diffusion of methane in the atmosphere, including wind speed, surface temperature, and so on.\textsuperscript{25,26} However, there are few studies of the effects on internal diffusion in soil. The environmental side factors affecting the diffusion of methane in soil mainly include environmental humidity, environmental temperature, and so on. Among them, environmental humidity affects soil moisture content, which in turn affects the diffusion of methane.\textsuperscript{27,28} Also, the change of environmental temperature will affect the internal temperature of soil, thereby affecting the diffusion of methane, but its effect needs to be further studied.

In summary, previous research on pipe side factors such as leakage pressure, leakage aperture, leakage direction, and pipe buried depth, as well as porosity and water content on the soil side, has been relatively completed. This paper further investigates the effects of soil type (porosity and granule diameter), layered structure among the soil side factors, and soil temperature among the environmental side factors on the methane leak diffusion law in soil by using a combination of experimental and numerical simulations. Also, studying the influence of multiple factors on the leak diffusion law of methane in soil can identify the main factors affecting diffusion and provide a basis for the next analysis of the influence of each factor on the dangerous radius of methane. Based on this, a prediction model for the dangerous radius of methane with leakage pressure, leakage aperture, porosity, and granule diameter as the dominant influencing factors is proposed by comprehensively analyzing the factors affecting the leak diffusion law of methane in soil. In engineering practice, an in-depth analysis of the factors affecting the dangerous radius and its prediction is of great significance to locate the leakage source as soon as possible and reduce the hazard of pipeline leakage.

2. RESEARCH METHODS

2.1. Experimental Method. 2.1.1. Experimental System.

In order to investigate the effects of various factors on the leak diffusion law of methane in soil, small-scale experiments are conducted in a wooden box with dimensions of 1.6 m × 1 m × 1.2 m (L × W × H). The schematic diagram of the experimental system is shown in Figure 1.

It can be seen from Figure 1 that the experimental system consists of a gas supply system, a soil domain, a pipeline leakage system, gas concentration sensors, and a data collection system. The gas flows out from a 40 L high-pressure gas cylinder (10.5 Mpa), passes through a pressure reducing valve (set to 0.1 Mpa) and a volume flowmeter (set to 10 L/min), and is supplied by a hose into the pipe. In order to withstand the weight of the soil, the leakage pipe across the soil domain is made of steel with a hole of diameter 3 mm in the center of the pipe. Also, to ensure gas tightness, the gap between the pipe and the wooden box is plugged with clay and sealed with waterproof tape.

In the experiment, the laser sensor probe is directly buried in the soil, and the sensor parameters are shown in Table 1. For
this experimental bench, sensors are uniformly distributed with a distance of 0.3 m because the uniform distribution of sensors can save costs and maximize the detection range of the methane and will not affect the soil structure and gas diffusion process. The optimal number and layout of sensor points are shown in Figure 2.

2.1.2. Experimental Contents and Working Conditions. Since natural gas is flammable and explosive and the test bench is placed in the experimental workshop, methane in nitrogen (4% methane and 96% nitrogen) is used as the experimental gas in order to prevent accidents. The measured gas parameters are shown in Table 2.

In order to ensure the homogeneity of the soil and reduce the experimental error, manual compaction is performed when the soil is boxed. Different soils are applied for the experiments, including sand and clay after sieving to remove impurities. In addition, the experiments include a study of the layered structure, in which the parameters of cement concrete and gravel are compared with those of the soil, as shown in Table 3.

The leak diffusion law of methane in soil is studied considering various experimental conditions, which can be found in Table 4.

2.2. Numerical Simulation Method. 2.2.1. Numerical Model Establishment. In order to conveniently verify the correctness of the numerical simulation, a small-scale model consistent with the dimensions of the experimental bench is established, with the parameters described previously, and the leakage gas is methane in nitrogen, called model 1. After verifying the correctness of the numerical model, in order to obtain the dangerous radius when the methane diffusion reaches the steady state after leakage, the small-scale model is expanded to a large-scale model 15 m in length, 15 m in width, and 5 m in height according to the technical code for city gas, with a pipeline buried depth of 1.5 m, a pipe diameter of 150 mm, a leakage hole of 5 mm, and the leakage gas as 100% methane, called model 2. The sketch of numerical model 2 is shown in Figure 3. The nonlinear Darcy’s law, diluted species transport, and heat transfer in porous media in COMSOL are used to solve the leak and diffusion process of methane in soil. The version of COMSOL software is 5.6. In this paper, only homogeneous material is considered, neglecting the heterogeneity of the material or underground sediments in the current case.

To simplify the model, some necessary assumptions are made: (1) the soil is an isotropic homogeneous porous medium and the spatial structure does not change; (2) methane does not react chemically with the surrounding soil;

Table 1. Sensor Parameters

| dimension (mm) | accuracy | detection method | measurement range (VOL %) | working temperature (°C) | working humidity |
|---------------|----------|------------------|---------------------------|--------------------------|-----------------|
| 90 × 38 × 38  | %3       | TDLAS            | 0–5%                      | 0–55                     | 0–98%RH         |

Figure 2. Arrangement and number of sensors.

Table 2. Properties of the Considered Gas

| components   | volume fraction (−) | M (g/mol) | ρ (kg/m³) | M (× 10⁻⁶ Pa·s) |
|--------------|---------------------|-----------|-----------|-----------------|
| methane      | 0.04                | 16        | 0.716     | 11,067          |
| nitrogen     | 0.96                | 28        | 1.250     | 17,805          |

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The leak diffusion law of methane in soil is studied considering various experimental conditions, which can be found in Table 4.

Table 3. Soil Parameters

| soil type       | mean granule diameter (mm) | porosity (VOL %) | density (kg/m³) | Absolute permeability (m²) | coefficient of diffusion (mL/s) |
|-----------------|-----------------------------|------------------|-----------------|---------------------------|---------------------------------|
| clay            | 0.05                        | 35               | 1531            | 6.8 × 10⁻¹⁵               | 4.7 × 10⁻⁸                     |
| sand            | 0.3                         | 30               | 1650            | 3.3 × 10⁻¹¹               | 3.8 × 10⁻⁸                     |
| crushed stone   | 5                            | 20               | 2350            | 1.7 × 10⁻⁹               | 2.2 × 10⁻⁸                     |
| cement concrete | 3                            | 5                | 1400            | 6.9 × 10⁻¹²               | 3.5 × 10⁻⁷                     |

Table 4. Experimental Conditions

| scenario | soil temperature (°C) | soil type     |
|----------|-----------------------|---------------|
| case 1   | 5                     | sand          |
| case 2   | 5                     | clay          |
| case 3   | 15                    | clay          |
| case 4   | 25                    | clay          |
| case 5   | 25                    | clay with sand backfill |
| case 6   | 25                    | layered structure |
and (3) the temperature of methane at the leakage hole is constant.

2.2.1. Nonlinear Darcy’s Law. The nonlinear Darcy’s law in COMSOL gives the mass conservation equation and the velocity expression as follows

\[
\frac{\partial (\varepsilon \rho)}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = Q_m
\]

(1)

\[
\nabla p = -\frac{\mu}{k} \mathbf{u} - \frac{C_F}{\sqrt{k}} \rho \mu_l \mathbf{u}
\]

(2)

\[
C_F = \frac{1.75}{\sqrt{150 \cdot \varepsilon}}
\]

(3)

where \( \varepsilon \) is the porosity; \( \rho \) is the density of the mixed gas \([\text{kg/m}^3]\); \( \mu \) is the slow rate \([\text{m/s}]\); \( Q_m \) is the mass flow rate \([\text{kg/s}]\); \( k \) is the permeability \([\text{m}^2] \); \( \nabla p \) is the pressure gradient \([\text{Pa}] \); \( \mu_l \) is the viscosity \([\text{Pa}\cdot\text{s}] \); and \( C_F \) is a dimensionless friction coefficient.\(^{30}\)

The density of the mixed gas can be calculated using the following equation

\[
\rho_{\text{mix}} = \sum_{i=1}^{n} \frac{p(M_i \varepsilon_i)}{RT}
\]

(4)

where \( p \) is the pressure \([\text{Pa}] \); \( M_i \) is the molar mass of the gas component \( i \) \([\text{g/mol}] \); \( \varepsilon_i \) is the molar fraction of the gas component \( i \); \( R \) is the gas constant \([\text{J/(mol\cdot K)}]\); and \( T \) is the gas temperature \([\text{K}]\).

The viscosity of the mixed gas can be calculated using Wilke’s equation:\(^{31}\)

\[
\mu_{\text{mix}} = \sum_{i=1}^{n} \frac{x_i \mu_i}{\sum_{i=1}^{n} x_i \phi_i}
\]

(5)

\[
\phi_i = \frac{1}{\sqrt{8}} \left( 1 + \left( \frac{M_i}{M_j} \right)^{1/2} \left( \frac{\mu_i}{\mu_j} \right)^{1/4} \right)^2
\]

(6)

where \( \mu_i \) is the viscosity of the gas component \( i \) \([\mu\text{Pa}\cdot\text{s}]\).

The gas permeability can be calculated using the Ergun empirical equation:\(^{32}\)

\[
K = \frac{\varepsilon^3 \rho^2 p^2}{180(1 - \varepsilon)^2}
\]

(7)

where \( d_p \) is the soil granule diameter \([\text{mm}]\).

2.2.1.2. Diluted Species Transport in Porous Media. The diluted species transport in porous media in COMSOL is used to describe the gas transportation in the soil:

\[
\frac{\partial (\varepsilon C_i)}{\partial t} + \mathbf{u} \cdot \nabla C_i + \nabla \cdot (D_{\text{eff}} \nabla C_i) = Q_i
\]

(8)

\[
D_{\text{eff}} = \frac{\varepsilon}{\tau} D_0
\]

(9)

where \( C_i \) is the concentration of the gas component \( i \) \([\text{mol/m}^3]\); \( D_{\text{eff}} \) is the gas diffusion coefficient in soil \([\text{m}^2/\text{s}]\); \( Q_i \) is the molar mass source term; \( \tau \) is the tortuosity; and \( D_0 \) is the methane–air diffusion coefficient \([\text{m}^2]\).

Since the model takes into account the effect of temperature on gas diffusion, the methane–air diffusion coefficient at different temperatures can be calculated using the Fuller equation:\(^{33}\)

\[
D_0 = 0.101 T^{1.75} \left( \frac{1}{M_A} + \frac{1}{M_B} \right)^{1/3} P \left[ \sum vA \right]^{1/3}
\]

(10)

where \( M_A \) is the molar mass of gas component \( A \) \([\text{g/mol}]\); \( \sum vA \) is the molecular diffusion volume of gas component \( A \) \([\text{cm}^3/\text{mol}]\); the molecular diffusion volume of gas component...
A (methane) is $25.14 \text{ cm}^3/\text{mol}$; and the molecular diffusion volume of gas component B (air) is $19.7 \text{ cm}^3/\text{mol}$.

The tortuosity $\tau$ in eq 9 is solved using the Millington Quirk model

$$\tau = e^{-\frac{1}{3}}$$

2.2.1.3. Heat Transfer in Porous Media. The variation of the temperature in soil is solved using the heat transfer in porous media in COMSOL

$$\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot (-k \nabla T) = Q$$

$$\rho C_p = \epsilon ps C_p s$$

$$k = \epsilon k_s$$

where $\rho_s$ is the soil density [$\text{kg/m}^3$]; $C_p_s$ is the soil specific heat capacity [$\text{J/(kg} \cdot \text{K)}$]; $k_s$ is the soil thermal conductivity [$\text{W/(m} \cdot \text{K)}$]; and $Q$ is the heat source term.

2.2.1.4. Boundary Conditions. The leakage hole is set as the flow inlet boundary, the upper surface of the soil domain is set as the atmospheric pressure boundary, and the rest of the surface is set as the wall boundary. The upper surface of the soil domain is set as the convective heat transfer boundary, the heat transfer coefficient $h$ is $15 \text{ W/(m}^2 \cdot \text{K)}$, and the other surfaces are the adiabatic boundary. The initial methane concentration in the soil domain is set to 0.

2.2.2. Mesh Segmentation and Independence Verification. According to the model to be solved in the numerical simulation, the mesh around the leakage hole is locally refined, and the rest is divided using a tetrahedral mesh. For model 1 and model 2, concentrations are monitored at three locations: top, bottom, and on the side of the leakage hole at 30 min to verify grid independence, as shown in Figure 4. From Figure 4a, it can be found that the methane concentration basically does not change when the grid number continues to increase from 430,000, so the grid number for the small-scale model is chosen to be 430,000. According to Figure 4b, the grid number of the large-scale model is 800,000 based on the same way.

2.2.3. Model Verification. For the model 1, a physical model in the numerical simulation is consistent with the size of the experimental bench for verifying the correctness of the numerical simulation. Figure 5 shows the comparison between the experimental and simulation results of the concentration with time, where the soil type is sand. It can be seen from the figure that the concentration error is within 5%, and the simulation results are in good agreement with the experimental ones, which verifies the correctness of the numerical simulation.

3. RESULTS AND DISCUSSION

In this section, both the small-scale experimental results and the large-scale numerical results are shown.

3.1. Verification of the Sensor Arrangement. In the experiment, the methane concentration curve of sensors placed separately and all placed are compared to verify whether the sensors have any effect on the leak diffusion process of methane. As shown in Figure 6, the difference in the concentration between the two conditions is less than 0.05%, so the sensors do not have a significant effect on the methane leak diffusion process.

3.2. Soil Temperature. 3.2.1. Experiment. In order to investigate the effect of soil temperature variation on methane diffusion, representative sensor points 4, 9, and 11 are selected from near the leakage hole, away from the leakage hole, and the soil surface. The sensor coordinates are shown in Figure 2. In the experiment, when the change of methane volume concentration is less than 0.05% in 30 min, the gas diffusion is considered to reach the steady state.

It can be seen from Figure 7 that the temperature change has little impact on sensor 4 near the leakage hole. The concentration of methane when it diffuses to reach the steady state is basically the same at different temperatures, and the increase in temperature mainly shortens the time it takes for its diffusion to reach the steady state.

Since sensor 9 is far away from the leakage hole, it is obviously affected by temperature. When the temperature increases, it can be found that the concentration curves of the three conditions have the same trend, but the concentration at the steady state has obvious differences. When the temperature increases by 20°C, the volume concentration of methane at the steady state increases from 3 to 3.35%, which increases by...
0.1 times. Sensor 11 at the surface is the most affected by temperature, and when the temperature increases by 20 °C, the methane concentration increases from 0.82 to 1.16% and the concentration increases by 0.4 times.

The diffusion forms of gas in soil are mainly divided into two types. Due to the high pressure at sensor 4 near the leakage hole, methane flows outward mainly due to the pressure difference with high velocity. At sensors 9 and 11, which are far away from the leakage hole, the gas mainly diffuses due to the concentration difference, the diffusion speed is slow, and the increase of temperature will speed up the movement of gas molecules and thus the diffusion of methane, so the increase of temperature here will increase the diffusion rate of methane more obviously.

3.2.2. Simulation. Due to the limitation of experimental conditions, the temperature range studied in the experiment is limited. Therefore, more in-depth research is carried out by means of numerical simulation to improve the variation range of soil temperature. After temperature monitoring, the soil temperature is generally about 5 °C lower than the ambient temperature in summer and 5 °C higher in winter in the experimental plant.

Two operating conditions are discussed below: when the ambient temperature is 0 °C and the soil temperature is 5 °C and when the ambient temperature is 45 °C and the soil temperature is 40 °C.

As can be seen from Figure 8, the effect of soil temperature variation on the methane leak diffusion process is not significant. Even if the temperature difference reaches 35 °C, the methane concentration cloud map range increases slightly.

After methane leakage reaches the surface, it is more dangerous to mix with air. Therefore, a point is set at the surface directly above the leakage hole to monitor the mole flux at the surface and observe the mole flux at the surface under different temperature conditions, as shown in Figure 9. From the figure, it can be found that the magnitudes of the mole flux at the surface at 1000 min are $6.03 \times 10^{-6}$ mol/(m²·s) and $8.56 \times 10^{-6}$ mol/(m²·s) for the soil temperatures of 5 °C and 40 °C, respectively. This shows that when the internal temperature of soil increases, the movement of gas molecules will accelerate and the diffusion speed will increase so that the
time of methane diffusion to the surface will be shortened and the amount of gas diffusion to the atmosphere will increase.

The effect of soil temperature on the dangerous radius is further studied. It can be found from Figure 10 that when the soil temperature changes by 35 °C, even after 1000 min of diffusion, the dangerous radius only differs by 0.11 m, increasing to 0.04 times, indicating that the effect of soil temperature on the dangerous radius is a relatively minor factor.

3.3. Soil Type. 3.3.1. Experiment. Sand and clay are common soil types in engineering. The similarities and differences of methane leak diffusion law between the two soil types are investigated experimentally. As shown in Figure 11, representative sensor points 4, 9, and 11 are still selected. Because sensor 4 is close to the leakage hole, the change trend of the concentration curve under the two conditions is basically the same. The difference is that the time for clay to reach the steady state is relatively delayed compared with that for sand, and the methane concentration in clay at the steady state is slightly lower than that in sand. At sensor 9, which is far from the leakage hole, the methane is still in the increasing concentration stage in the clay at 120 min with a concentration size of 2.22%, while diffusion in the sand reached a steady state with a concentration size of 3.93%, an increase of 0.77 times compared to the former. Compared with the concentration curve of sensor 11 at the surface, it can be found that at 120 min, the methane concentration at the surface under the clay condition is 0.74% and that under the sand condition is 1.96%, which is an increase of 1.65 times. Further, it can be seen from Figure 11 that when methane diffuses in sand, the diffusion time to the surface is about 20 min, and when methane diffuses in clay, the diffusion time to the surface is about 60 min, which is shortened by a factor of 2. It shows that when methane diffuses in the vertical direction, it is much more hindered in clay.

Under the condition of approximate porosity, the granule diameters of clay are small, the pores are dense, and the blocking effect of upward diffusion of methane is strong. When methane diffuses in sand, the granule diameters are large,
Figure 11. Concentration in different soils (sand and clay).

Figure 12. Fitting curve of vertical diffusion distance in different soils.

Figure 13. Methane mole flux through the ground surface for different porosities.

Figure 14. Dangerous radius for different porosities.

Figure 15. Methane mole flux through the ground surface for different granule diameters.

Figure 16. Dangerous radius for different granule diameters.
internal pores are relatively sparse, and the gas is less impeded in upward diffusion, which leads to different methane diffusion laws in two soils.

Figure 12 shows a fitting curve of gas diffusion distance and time in the vertical direction in sand and clay. From the figure, it can be found that the slope of the curve gradually decreases with time when methane diffuses in both soil types, which

Figure 17. Side view of the clay with sand backfill structure.

Figure 18. Comparison of concentrations for clay with sand backfill and clay.

Figure 20. Concentration of clay with sand backfill and the layered structure with sand backfill.

Figure 21. Fitting curve of vertical diffusion distance for different pipe laying methods.

Figure 19. Side view of the layered structure with backfill.
proves that the diffusion rate decays gradually when the gas diffuses upward in two soils, especially in clay. Also, according to the graph, the average diffusion velocity of the two soils in the vertical direction can be obtained, that is, $6.48 \times 10^{-4}$ m/s for sand and $1.92 \times 10^{-4}$ m/s for clay. The comparison shows that the diffusion velocity of methane in the vertical direction in sand is greater than that in clay.

3.3.2. Simulation. It is found that the difference of gas leak diffusion law between two soils is mainly caused by the difference of basic parameters of porosity and granule diameter. Therefore, the effects of changing porosity and granule diameter on the gas leak diffusion law and dangerous

Table 5. Parameter Setting of the Material$^{23,29,34}$

| material type | mean granule diameter (mm) | porosity (vol %) | absolute permeability$^a$ (m$^2$) | diffusion coefficient$^b$ (m$^2$/s) |
|---------------|---------------------------|------------------|-----------------------------------|-----------------------------------|
| cement concrete | 3                         | 0.02             | $3.09 \times 10^{-13}$            | $1.03 \times 10^{-7}$             |
| road base     | 5                         | 0.03             | $4.78 \times 10^{-12}$            | $1.77 \times 10^{-7}$             |
| subgrade layers | 0.05                     | 0.1              | $2.06 \times 10^{-14}$            | $8.82 \times 10^{-7}$             |
| backfill      | 0.5                       | 0.05             | $2.31 \times 10^{-15}$            | $3.5 \times 10^{-7}$              |

$^a$Calculated using eq 7. $^b$Calculated using eq 9.

Figure 22. Concentration contours of methane over time in the layered structure.
radius are further studied with the help of numerical simulation software. The effect of varying the porosity on gas diffusion is investigated using the granule diameter determined. From Figure 13, it can be seen that when the granule diameter is 0.1 mm, the change law of methane molar flux at the surface increases rapidly in the first period, and then, the rate gradually slows down and finally approaches a stable value. When the porosity is 0.1, the methane cannot diffuse far outward due to the high resistance of the soil and only accumulates near the leakage hole, so the molar flux at the surface is 0. When the porosity continues to increase, the molar flux value at the surface in the steady state gradually increases to a maximum of 0.002967 mol/(m²·s). Except for the case of a porosity of 0.1, the time for methane to reach the surface decreases with the increase of porosity, and when the porosity is 0.5, methane diffuses to the surface in 60 min, while when the porosity is 0.2, it takes 420 min to diffuse to the surface.

Figure 14 shows the dangerous radius variation curves for different porosities. It can be found that the trend of the dangerous radius change with time is that it increases rapidly first and then tends to level off gradually. With the increase of porosity, the dangerous radius of methane gradually increases, but the increase of the dangerous radius decreases with the increase of porosity. When the porosity increases from 0.1 to 0.5, the dangerous radius at 1000 min increases by 2.87 m, which is 8.3 times larger, indicating that porosity is an important factor affecting the dangerous radius of methane. Furthermore, the effect of the change of soil granule diameter on methane diffusion is studied by fixing the porosity. Figure 15 shows the comparison of methane molar flux at the surface under different soil granule diameters when the porosity is 0.5. It can be seen from the figure that when the soil granule diameter is 0.01 mm, it is difficult for methane to break through the resistance, so methane is not detected at the surface. When the soil granule diameter continues to increase, the molar flux at the surface gradually increases, with the
maximum of 0.007649 mol/(m$^2$·s). The time for methane to reach the surface decreases with the increase of soil granule diameter. When the soil granule diameter is 0.5 mm, methane diffuses to the surface in 20 min, while when the soil granule diameter is 0.05 mm, methane diffuses to the surface in 100 min.

The change of the dangerous radius with time for different soil granule diameters is the same as that for different porosity conditions, as shown in Figure 16; that is, both of them rapidly increase in the early stage and then gradually level off. With the increase of the soil granule diameter, the dangerous radius of methane also increases gradually. The increase of the dangerous radius decreases with the increase of soil granule diameter, and when the granule diameter increases from 0.01 mm to 0.5 mm, the dangerous radius increases by 2.71 m at 1000 min, which is increase of 3 times, indicating that it is also an important factor affecting the dangerous radius.

3.4. Layered Structure. 3.4.1. Clay with Sand Backfill.

During the laying of underground pipelines, sand is backfilled near the pipeline to prevent damage or settlement of the pipeline. Therefore, in this paper, sand is filled in each of the upper and lower parts of the pipeline and the rest is clay to study the effect of backfill on the methane leak diffusion law, and the side view of the clay with sand backfill structure is shown in Figure 17.

For the soil structure with backfill, the methane diffusion is compared with that of a single soil structure, as shown in Figure 18, and representative sensor placements 4, 9, and 11.
are still selected. As can be seen from the figure, for sensor 4 near the leakage hole, the methane under the backfill condition is able to reach the steady state earlier because the sand with a larger granule diameter is buried near the pipe, so the leak and diffusion rate is faster, and the concentration magnitude at the steady state is basically the same for both conditions. At sensor 9, the trend of methane concentration over time is the same for both conditions, but the overall concentration under the backfill condition is higher than that under the single clay condition, and the concentration increases by 1.04 times at the steady state. At sensor 11 at the surface, the time under the backfill condition for methane diffusion to reach the surface is greatly reduced and the concentration magnitude at the steady state is increased by a factor of 0.7, from 1.1 to 1.87%. The reason for this phenomenon is that sensor 11 is far away from the leakage hole, where the gas diffusion is mainly driven by the concentration gradient, and the impact of sand backfill is more obvious. Therefore, the methane in the clay with sand backfill condition will accumulate at the surface to reach a higher concentration, which leads to a large difference in the methane concentration at the surface of the two working conditions at the steady state.

3.4.2. Layered Structure with Backfill. 3.4.2.1. Experiment. Based on the previous work, the layered structure is extended to the case with backfill. In the experiment, the side view of the layered structure with backfill is shown in Figure 19, where the pavement structure is simplified, with cement concrete slabs used for the surface material to replace the cement concrete pavement scene, crushed stone for the base material, clay for the roadbed material, and medium coarse sand for the backfill material.

Figure 20 compares the similarities and differences of the methane concentration variation law with time in the single soil structure with backfill and the layered structure with backfill. For sensor 4 near the leakage hole, both conditions are filled with sand of a larger granule diameter near the pipe, so the change trends of both the filling methods here and the concentration magnitude at the steady state are the same. At sensor 9, there is a big difference between the concentration change curves of the two filling methods. The methane concentration of the soil structure with backfill at the steady state is 3.46%, while the methane concentration of the layered structure is 3.95%, which is an increase of 0.14 times. The concentration difference between the two filling methods at the surface is the largest. Under the layered structure condition, because the porosity and permeability of the gravel and cement are low, the gas cannot effectively diffuse into the air, resulting in the rapid accumulation of methane near the surface. Its steady-state concentration value is 1.1 times that of the soil structure with backfill. In general, all locations under the layered structure with backfill condition reach the upper limit of concentration, which indicates that the gravel and cement layer on the top layer will greatly hinder the gas diffusion, resulting in the methane concentration accumulated in the soil being higher and more dangerous.

Figure 21 shows the fitting diagram of diffusion distance and time in vertical direction for different filling methods. From the overall view, the diffusion times to the surface for clay with backfill and the layered structure with backfill are not much different, and the methane in the clay takes the longest time to diffuse to the surface. The average diffusion velocity in the vertical direction for the three filling conditions can be obtained according to the diagram, that is, $1.92 \times 10^{-4}$ m/s for clay, $6.65 \times 10^{-4}$ m/s for the soil structure with backfill, and $7 \times 10^{-4}$ m/s for the layered structure with backfill, which indicates that the backfill has a great enhancement on the diffusion velocity of gas.

3.4.2.2. Simulation. Due to the limitation of experimental conditions, it is difficult to obtain the concentration distribution of methane in the layered structure with backfill, so the study is continued with the help of the numerical simulation method. In practice, the compaction rate of the pavement is above 90%, so in order to be close to the reality, the compaction of different materials is set higher, and the parameter setting of the material is shown in Table 5.

The cloud diagram of the concentration distribution of the methane diffusion process in the layered structure with backfill is shown in Figure 22. From the figure, it can be seen that at the early stage of leakage, the methane will mainly diffuse along the backfill laterally because of the presence of backfill (medium and coarse sand) near the pipeline, and it will also diffuse in other directions, but the lateral diffusion speed is greater than the diffusion speed in other directions, so the diffusion cloud will show a long ellipse shape. Afterward, when the methane diffusion has continued over a certain time (240 min), the diffusion rate starts to slow down because at this time, the methane will accumulate near the surface due to the obstructing effect of the cement concrete and gravel at the top; thus, it can be found that the contour at the surface will bend outward, which proves that the diffusion rate will be reduced due to the obstruction when the gas diffuses to this place. When the lateral diffusion reaches a certain level, the methane will continue to diffuse along other directions due to the concentration difference and the obstruction of cement concrete and gravel at the top until the diffusion shape gradually changes to a semicircular shape at the steady state. Therefore, when the pipeline is buried under the town road, it will have a great impact on the methane concentration cloud pattern, and the contour arrangement will change from concentric circles to irregular shapes, which will make it more difficult to find the location of the leakage point.

According to Figure 23, at about 240 min, a relatively obvious concentration change is detected at the surface. The concentration at the center is the highest with a size of about 25%. The methane dangerous radius increases from 1.23 m (240 min) to 4.39 m (3000 min), an increase of 2.6 times. Due to the existence of backfill, the shape of the cloud map of methane at the surface is a flat ellipse, but the shape gradually changes to concentric circles with time.

3.5. Prediction Model of the Dangerous Radius. The dangerous radius of methane in soil is influenced by various factors, including leakage pressure, leakage aperture, leakage direction, and pipe buried depth among pipe side factors, porosity, granule diameter, and water content among soil side factors, and soil temperature and ambient humidity among environmental side factors. Previous studies on pipe side factors have been relatively completed, among which the impact of pressure and leak aperture will have a greater influence on the dangerous radius of methane. Further, based on the in-depth study of porosity and granule diameter in soil side factors and soil temperature in environmental side factors in this paper, it is concluded that the dominant factors affecting methane diffusion in soil are leakage pressure $P$, leakage aperture $d$, porosity $\epsilon$, and soil granule diameter $d_p$, while soil temperature in environmental side factors as a secondary factor affecting.
The relationship between each influencing factor and the dangerous radius of methane at the steady state is shown in Figures 24, 25, 26, and 27. Figures 24 and 25 show the effect of changing the leakage pressure or leakage aperture on the dangerous radius of methane at the steady state when fixing the porosity and soil granule diameter in the soil side factors, and it can be found that the curve of the dangerous radius of methane changes more gently as the leakage pressure or leakage aperture increases. Figures 26 and 27 show the effect of varying the porosity and soil granule diameter on the dangerous radius of methane at the steady state by fixing the leakage pressure or leakage aperture, and it can be found that the effect of porosity and granule diameter on the dangerous radius of methane at the steady state is greater than that of leakage pressure and leakage aperture.

When any of the four influencing factors is zero, the size of the dangerous radius at the steady state is 0 m. Therefore, the form of the fitting formula can be expressed as the product of the influencing factors, and the nonlinear regression method is carried out according to the simulation results to obtain the predicted relationship between the dangerous radius and various influencing factors when the methane diffusion reaches the steady state, where the coefficient of determination $R^2$ is 0.91. The formula is shown as

$$R = 15.68 · P^{0.12} · d^{0.27} · ε^{0.34} · d_p^{0.21}$$  \hspace{1cm} (15)

Among them, the leakage pressure $P$ is in the range of 0.05—0.1 Mpa, the leakage aperture $d$ is in the range of 5—15 mm, the porosity $ε$ is in the range of 0.1—0.5, and the soil granule diameter $d_p$ is in the range of 0.05—0.5 mm.

As can be seen from Figure 28, when the diffusion radius is small, the fitted results are more different from the experimental results, the relative error ranges from −10 to 25%, and the error decreases when the diffusion radius increases. The overall average relative error is 7.3%, which proves that the accuracy of the prediction formula can be guaranteed.

4. CONCLUSIONS

In this paper, the effects of soil parameters (such as temperature, porosity, granule diameter, and layered structure) on the diffusion law of natural gas leakage in the soil are investigated, and a prediction model for the dangerous radius is proposed by using a small-scale experimental bench combined with small-scale and large-scale numerical simulations in COMSOL. The following conclusions are obtained:

1. The increase of soil temperature enables the methane to diffuse faster and increases the methane concentration at the steady state. When the temperature is changed by 35 °C, the dangerous radius at 1000 min is increased only by 0.04 times, so the effect of soil temperature on the dangerous radius of methane is a secondary factor.

2. Methane diffuses faster laterally when diffusing in clay and vertically when diffusing in sand. The difference in gas diffusion in two soils is mainly due to the difference in porosity and granule diameter. When the granule diameter is 0.1 mm and the porosity increases from 0.1 to 0.5, the dangerous radius at 1000 min increases by 8.3 times. When the porosity is 0.5 and the particle diameter increases from 0.01 to 0.5 mm, the dangerous radius at 1000 min increases by 3 times.

3. Comparing the soil structure with sand backfill with clay, it is found that the methane diffusion will be less hindered and its diffusion rate will be accelerated because of the sand filling near the pipe. Also for the layered structure with backfill, the methane will mainly diffuse along the backfill at the beginning and will accumulate near the surface due to the obstructive effect of cement concrete and gravel, resulting in the methane reaching a higher concentration at the steady state, which is more dangerous.

4. All the influencing factors affecting the diffusion of methane in soil are comprehensively analyzed, among which leakage pressure, leakage aperture, porosity, and soil granule diameter are observed to be the dominant factors. Based on this, this paper proposes a nonlinear predictive equation for the dangerous radius associated with these four influencing factors when methane diffusion reaches the steady state, with a coefficient of determination of 0.91 and an overall average relative error of 7.3%, which is an important guidance value for identifying the leakage source of buried natural gas pipelines as soon as possible and reducing the hazard after leakage.

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Notes

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■ NOMENCLATURE

$M$ molar mass (g/mol)
$M_i$ molar mass of component $i$ (g/mol)
$x_i$ molar fraction of gas component $i$
$T$ gas temperature (K)
$\rho$ density of gas (kg/m$^3$)
$\rho_s$ density of soil (kg/m$^3$)
$\mu$ dynamic viscosity (Pa·s)
$\mu_i$ dynamic viscosity of gas component $i$ (Pa·s)
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