

Movement Characteristics of Hydrogen Gas Within the Ground and Its Detection at Ground Surface

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Abstract-Hydrogen gas is expected to be a clean, environmentally friendly future energy source. As a method to supply hydrogen gas to a fuel cell station, the use of buried pipelines is being studied in Japan and elsewhere. To realize this idea, ensuring safety is very important. Specifically, the understanding of hydrogen gas movement characteristics, including the extent and speed of movement within the ground in the event of a hydrogen leak, is extremely important, as is the reliability assessment of pipeline materials in order to establish the basis for the design and maintenance of safe facilities. To establish a foundation for ensuring the safety of buried hydrogen supply pipelines, full-scale experiments on underground hydrogen gas leakage and numerical simulation were performed. The release and movement characteristics of hydrogen gas within soil were investigated and measured with the objective of establishing hydrogen gas leak detection technology.

Keywords- Road Base; Buried Pipeline; Partially Saturated Soil; Hydrogen Gas Permeability

I. INTRODUCTION

A. Background and Objectives

Chemical plants and the gas industry have large buried pipelines for transporting combustible gases. In order to safely manage those buried pipelines, it is extremely important not only to evaluate the long-term reliability of the pipeline materials but also to understand the movement characteristics, including the extent and speed of movement in the ground, of the materials being transported in the event of a gas leakage. This information forms the foundation for the design and maintenance of safe facilities.

Hydrogen, which is expected to be the clean energy of the future, will be supplied to fuel cells in residences and offices via buried pipelines.

Fig. 1 is an image of the supply route of hydrogen to fuel cells via buried pipelines.

For the supply of hydrogen gas through buried pipelines, safety and maintenance are important. To investigate and study this matter, full-scale tests were carried out on the leakage of hydrogen gas, and numerical simulations of the movement of hydrogen gas in the ground were done.

Thus, the movement characteristics of hydrogen gas in the ground, including the extent and speed of movement, were determined. The characteristics of release above ground were also investigated to establish the technology for the detection of hydrogen gas leaks.

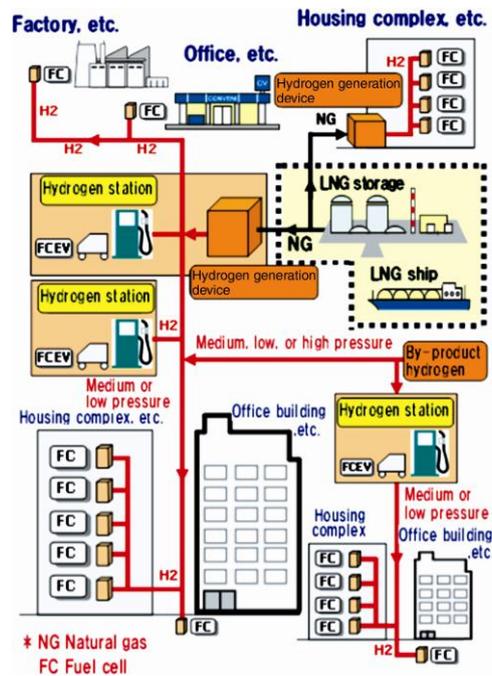


Fig. 1 Hydrogen gas supply to fuel cells via buried pipelines

B. Review of Past Research

The main topic of study, in relation to the movement through the ground of hydrogen gas that has leaked from a buried pipeline, is the spread of the gas in 3-dimensions through the gaps between soil particles under various ground conditions, thus the following items should be investigated.

a) If 100 vol% gas is released from a leak in a pipeline that is delivering the gas under pressure, the behavior due to the main factors that govern the spread of the gas through the ground, such as the differences in pressure, specific gravity and concentration, must be investigated. Because the specific gravity of hydrogen is lighter than that of air, comparative investigations must be performed.

b) The effect of the compositions of the gas and the soil, as well as the geometric boundary conditions of the ground, must be investigated.

c) Not only the extent of spread but also the local behavior at the point of leakage needs to be investigated.

To achieve those, it is necessary to carry out a three-dimensional study by full-scale verification tests, and numerical analysis based on the results of the verification tests.

A review of past research was carried out to confirm the dispersion behavior of hydrogen gas in the ground by means of full-scale leak tests, and the validity or applicability of the numerical measurement method was verified. There has been much research regarding the dispersion of gas in the atmosphere, both indoors and outdoors, but comparatively little research on the dispersion behavior of gas leaked into the ground. In order to apply the results of past research to the present study, an extensive survey of research was conducted on the behavior of fluids in the ground based on information from Japan and elsewhere. The following is a summary of the main results of that survey.

1) Studies on the flow of ground water, etc., under unsaturated and saturated conditions, due to a pressure difference, have been conducted (Hwang et al. 2004 [13]; Saito et al. 2003 [19]; Hibi 2007 [12]; Ewing et al. 1999 [7]; Faust 1985 [9]). Those studies have confirmed the behavior of gas-liquid two-phase flow and liquid phase flow for environmental pollution, penetration of rainwater, etc.

2) Gerson et al. (2003) [5] studied the behavior of water steams based on temperature and soil moisture and confirmed the behavior when the two-phase system of water steams is characterized by specific heat, thermal conductivity, and diffusion coefficient, as parameters of temperature and soil moisture content.

3) Nagai et al. (2004) [17] and Kumagai et al. (1998) [16] studied the formation and circulation of CO₂, N₂, etc., in the natural world of soil-plants-air and confirmed the formation and circulation behavior in one-dimension, vertically from the ground, and macroscopically.

4) Weerts et al. (2001) [26] studied gas diffusion based on the movement of particles and confirmed the behavior of a fluid, considered to be a collection of particles, that repeatedly impact and translate as regular particle movement (the Lattice

Boltzmann law).

5) Yan et al. (1996) [14], Arah et al. (1994) [3], Osozawa et al. (1987) [18], Egusa et al. (1994) [6] and others studied gas diffusion based on a difference in concentration. All those are studies on the molecular diffusion behavior of gases due to concentration differences.

6) Many other scholars have studied the gas movement that considers the differences in concentration, pressure and specific gravity. Those authors include Senger et al. (2007) [20], Wilson et al. (1987) [27], Fujinawa et al. (2001) [10], Akatsuka et al. (1998) [2], Kobayashi et al. (2007) [15], Slough et al. (1999) [22], Cheng et al. (1996) [4], Abriola et al. (1985) [1], Falta et al. (1992) [8], and Sleep et al. (1993) [21]. These are theoretical studies primarily on the movement of gas due to advection and dispersion using Darcy's law and Fick's law; however, some examples of full-scale verification tests and analysis using city gas considered the conditions of the differences in pressure, specific gravity, and concentration.

As described above, various studies have been done in the agricultural and civil engineering fields on the movement of water and dissolved substances and gases through the ground. In addition, in recent years, studies on the behavior of hydrogen generated by corrosion of nuclear waste containers in clay strata have been focusing on the movement of gases through the ground, in the soil contamination and other environmental soil-engineering fields. However, no full-scale tests and numerical simulations have been conducted, focusing on hydrogen gas to confirm the distribution of concentration to investigate its behavior within the ground under pressure difference, specific gravity difference, and concentration difference.

C. Research Policy

One of the main methods for ensuring the safety of buried natural gas pipelines, etc., is periodic leakage inspection, which is required by law (Gas Business Act, Examples of Interpretation of Technical Standards for Gas Products 113). It is required for leakage inspection to be conducted at least once every 40 months after the burial of pipelines by boring to a depth of about 50 cm at 5 m interval on the road surface, or by using a gas leak detector at the ground level. In the event that a leak is detected, the release of the leaked gas to the ground surface becomes the issue. It is difficult to identify the leaked gas if it has difficulty rising to the ground surface, and the leakage and movement of the gas through the ground will continue; thus, a dangerous situation could develop.

Using this detection procedure of city gas as a reference, it was decided that the basic policy of this research should consist of two phases as shown below.

In phase 1, a full-scale leak test of hydrogen gas within the ground in the ground condition equivalent to the basic condition of buried pipelines was carried out to measure the basic extent and speed of movement as well as to investigate the gas released to the ground surface using a gas leak detector. In addition, it was verified whether the result of the test can be reproduced by theoretical numerical analysis as well as the validity or applicability of such reproduction based on the comparison with the actual result.

Next, in phase 2, another full-scale leak test of hydrogen gas was performed; the space for the pipe alone was excavated and backfilled in the road, to simulate a real road burial site in the road of an urban area for the investigation of the characteristics of hydrogen gas movement and rising to the ground surface in the actual ground condition. In the actual ground, pipelines are buried within trenches of approximately 60 cm in width excavated from the ground surface. Thus, at the time of gas leakage, gas is expected to move through the boundary between the backfilling in the trench and the ground outside the trench. The purpose of this study is to confirm the conditions of such movements in tests as well as to investigate the possibility to reproduce them by the numerical analysis described above.

In the simulation using the numerical analysis, it is necessary to consider all the gas movement factors, such as pressure, specific gravity, and concentration differences, because 100 vol% gas, which is lighter than air, is released into the ground under pressure. Therefore, since it was assumed that the analysis method for advection–dispersion using Darcy's law and Fick's law, as stated in the sixth survey result above, could be theoretically applied, this method was adopted.

II. INVESTIGATION OF THE MOVEMENT CHARACTERISTICS IN THE GROUND OF LEAKED HYDROGEN GAS USING FULL-SCALE TESTS (PHASE I)

A. Experimental Investigation

1) Outline of the Test

General ground conditions to bury a hydrogen pipeline were postulated and a full-scale test field, where crushed stone and asphalt are laid on a uniform surface layer of pit sand, was prepared, as shown in Fig. 2. For safety concern, a 10 m × 10 m × 2 m test ground was prepared within a tent. To properly investigate the result of the test, the origin of the co-ordinate axes was considered the point on the ground surface directly above the hydrogen gas leak point, and directions shown in the Fig. 2 were assigned to x-, y-, and z-axes, respectively.

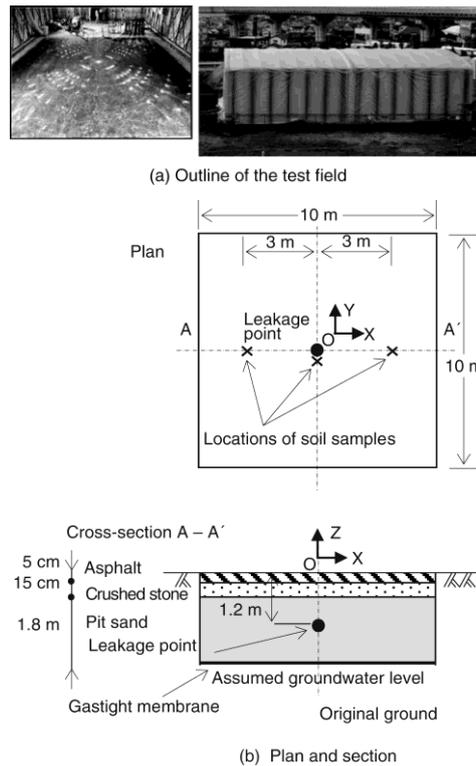


Fig. 2 Outline of the test field and composition of the test ground (Phase 1)

As shown in Fig. 3, the hydrogen gas was fed from a cylinder through a flow rate control device and risen up through a pipe in the center of the test field in which the piping has been laid in advance below a gastight membrane, which is described later. A porous ceramic sphere was fitted to the end of the pipe as the leak point.

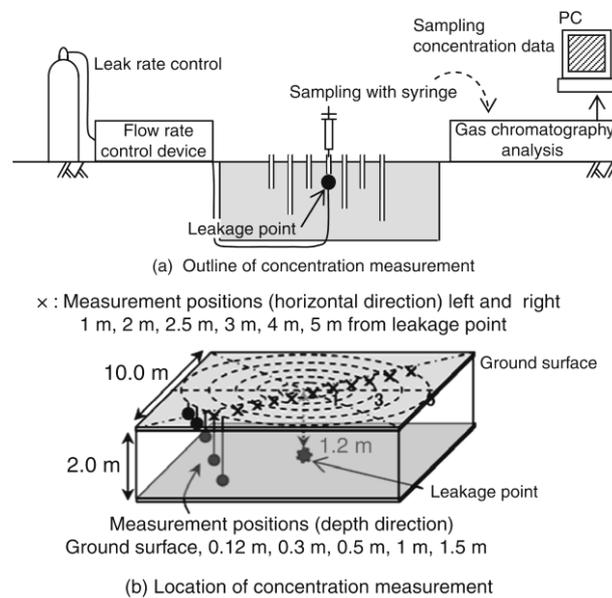


Fig. 3 Measurement of hydrogen gas concentration (Phase 1)

The tests were performed at a leakage pressure of 0.2 kPa, which is the same as for a leak at the supply pressure of city gas, as shown in Table 1. The leakage flow rate was 1000 cc/min, which corresponds to the value of the leakage flow rate from a corrosion hole or joint in the ground in the past.

TABLE 1 TEST CONDITIONS (PHASE 1)

Gas type	Hydrogen (specific gravity 0.07)
Leak rate	1000 (cc/min)
Leak pressure	0.2 (kPa)

Leak position	GL -1.2 (m)		
Postulated groundwater level	GL -2.0 (m)		
Test ground composition and properties * Asphalt is the dry density		Porosity (vol%)	Wet density (g/cm ³)
	Asphalt	7.5 ± 2.5	2.33 ± 0.01
	Crushed stone	22.1 ± 1.0	1.94 ± 0.01
	Pit sand	19.5 ± 3.0	1.91 ± 0.09
	Original ground	18.8 ± 3.0	1.92 ± 0.15

Assuming normal road construction, the test field used No. 14 pit sand (soil particle density 2.66 g/cm³, $D_{50} = 0.9$ mm, average water content 9.2%) that is commonly used for backfilling pipes. Compaction was carried out every 30 cm of backfill in the test field where the leakage point was installed. The sub grade was 15 cm of crushed stone ($D_{50} = 6.2$ mm), and the top layer was 5 cm of asphalt.

As the boundary condition for the movement of hydrogen gas through the ground, a rubber gastight membrane, through which water and gas cannot pass, was laid as a measure against fluctuations in the groundwater level at the bottom of the test field due to rain on the surrounding test site, and its position was considered the groundwater level. Based on a nationwide survey of groundwater levels for representative buried pipes, the average depth at which the membrane was laid was 2 m. The solubility of most combustible gases in water is small, and in the case of hydrogen used in the tests, the solubility is about 0.01 ml/1 ml in water (20 °C), which is negligible. Therefore, it was considered that it was possible to simulate the presence of the groundwater level by using the impermeable membrane. The side surfaces were considered in contact with the original ground and gas can pass through the sides.

In addition, to confirm the uniformity of the compaction of the ground, boreholes were made at the leakage point and at +3.0 and -3.0 m in the X-direction from the leakage point, and soil samples were taken for test at depths of $Z = -0.2, -0.5, -1.1,$ and -1.7 m. The results are shown in Table 1.

The concentration of hydrogen in the ground was measured by using several fine tubes (1-mm internal diameter stainless steel tubes with porous filters (10 μ m) fitted to their tips) placed in advance at specific positions and depths in the field, as shown in Fig. 3. In particular, at 3, 6, 9, 12, and 24 h after the start of leakage, and thereafter at 24-h interval, a small quantity of hydrogen gas was sampled from the measuring tubes using a syringe. Those samples were analyzed by gas chromatography (detection concentration limit for hydrogen gas 0.1 ppm). The sample quantity was about 10 ml, in order to be able to analyze accurately with gas chromatography, and to avoid affecting the gas movement.

The concentration of hydrogen gas released from the ground surface was measured using a cart-type general-purpose gas detector/ NEW COSMOS ELECTRIC CO., LTD (heat type semiconductor type/detection concentration limit for hydrogen gas 0.008 vol%), as shown in Fig. 4. The detector was positioned about 2 cm from the ground, traveling at a speed of 4 km/h, which is the same as that for the detection of city gas.

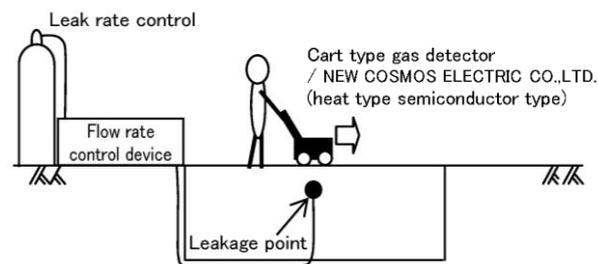


Fig. 4 Measurement of hydrogen gas concentration at the ground surface (Phase 1, 2)

2) Test Results

Change of hydrogen gas concentration in the ground with time

Fig. 5(b) shows the change in hydrogen gas concentration in the ground over time, as measured at test points 1 to 3 on cross section AA' in the test field, as shown in Fig. 5(a) (co-ordinates $(X, Z) = (0.0, -1.0) (-1.0, 0.5) (-2.0, -1.5)$).

Because the specific gravity of hydrogen is lower than that of air, it can easily move upward; therefore, at about 50 h, the leak rate of hydrogen gas into the ground and the rate at which it escapes into the atmosphere are balanced, and steady state

occurs.

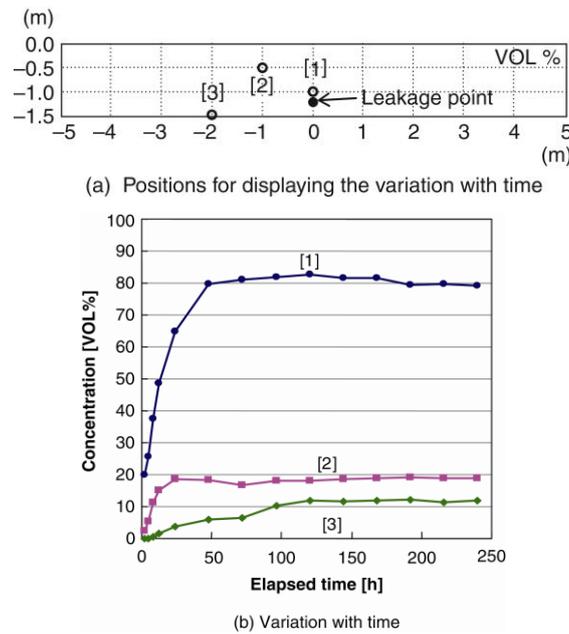


Fig. 5 Variation of the concentration of hydrogen gas in the ground with time (Phase 1)

Distribution of hydrogen gas concentration in the ground

Fig. 6 shows the distribution of hydrogen gas concentration in the ground at cross section AA' during the steady state condition after 240 h since the start of hydrogen leakage. The concentration distribution shows a symmetrical spread to the left and right with the leakage point as the center, reflecting the fact that the test field was uniform. In addition, since the specific gravity of hydrogen gas is much smaller than that of air, it can easily diffuse from the asphalt surface into the atmosphere, thus the concentrations in the upper layers of the test field and to the sides of the leakage point were comparatively low. Because of the effect of the gastight membrane that replicated the effect of groundwater at the bottom layer of the test field, the hydrogen was unable to move downward, so the concentration was comparatively high. The area where the concentration of hydrogen gas in the ground was 10 vol% or higher was distributed within ± 2 m of the point of leakage.

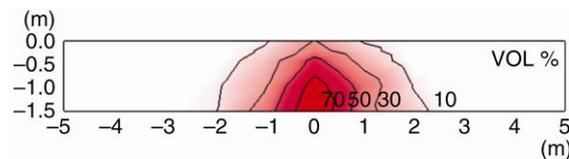


Fig. 6 Distribution of hydrogen gas concentration in the ground (Phase 1, after 240 hours)

Distribution of hydrogen gas released from the ground surface

Fig. 7 shows the distribution of hydrogen gas concentrations in the X-axis direction on the surface, measured with the cart-type gas detector as shown in Fig. 6 under steady state condition after 240 h from the start of leakage.

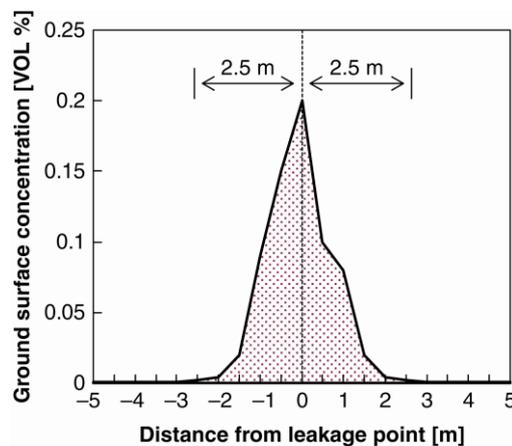


Fig. 7 Distribution of hydrogen gas concentration at the ground surface (Phase 1, after 240 hours)

The distribution was symmetric about the center directly above the leak point. Similar to the distribution in the ground, the concentration directly above the leak point was high at 0.2 vol%, and the concentration becomes lower the further the distance from the leak point. The hydrogen gas released from the ground was distributed about ± 2.5 m about the leak point at the center, and the cart-type gas detector, whose hydrogen gas concentration detection limit is 0.008 vol%, was capable of detecting the hydrogen within this area. This result indicates that it is possible to detect released hydrogen gas by periodic leak inspection using gas detectors on the surface of the road, as required by the Gas Business Act, as previously stated.

From the above results of the full-scale tests, the extent and speed of the movement and rising to the ground surface of hydrogen gas, when leaked under ground conditions equivalent to general pipeline burial conditions, were confirmed.

B. Numerical Simulation of the Movement Characteristics of Hydrogen Gas in the Ground

1) Basic Equations

As stated previously, the analysis method based on Darcy's law and Fick's law is applicable to advection-diffusion problems of gases through the gaps in a porous medium such as soils, so it is considered to be theoretically applicable in reproducing the full-scale test conditions in this research, where there are differences in pressure, specific gravity, and concentration.

For the flow of a fluid through the voids in soil, the equations of motion, law of conservation of mass, and so on apply. However, the shapes of the voids in soil are extremely complex; therefore, the equations of motion and the law of conservation were set up for volume elements, as shown in Fig. 8, with integration averaged volumes that are sufficiently larger than the local size of the voids and sufficiently smaller than the overall size of the ground being considered.

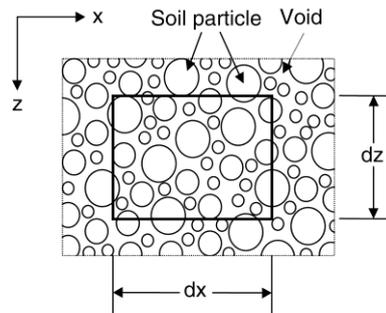


Fig. 8 Small volume element set in the soil

In addition, this research was concerned with low pressures of around 0.2 kPa, which is similar to leakage due to the supply pressure of city gas. It is considered that the movement of water due to such leakage pressure is extremely small, so the problem was to set it as a movement problem, through the voids of soil of a two-phase mixture, of hydrogen and air. Furthermore, the equations of state of an ideal gas were incorporated as the basic equations to consider the effect of thermodynamics.

Within the test field, No. 14 pit sand with an average water content of 9.2% was used, backfilled, and compacted every 30 cm. Therefore, it was considered that the soil was sufficiently uniformly compacted, and it was assumed in the numerical simulation that the volume of the soil did not vary.

The basic equations required for the numerical simulation are as follows:

Equations of motion

The equations of motion that describe the movement of a gas mixture through the ground in Cartesian co-ordinates are expressed by Eq. (1).

$$\rho \frac{\partial \vec{v}}{\partial t} + \rho(\vec{v} \cdot \nabla) \vec{v} = -\nabla p - [\nabla \cdot \vec{\tau}] + \rho \vec{g} + \vec{F}, \quad (1)$$

where

p : Mixed gas pressure [Pa],

\vec{g} : Acceleration due to gravity [m/s^2],

$\vec{\tau}$: Shear stress [N/m^2],

ρ : Mixed gas density [Kg/m^3],

\vec{F} : External force applied to mixed gas per unit volume,

\vec{v} : Mixed gas velocity [m/s].

The small-volume elements can be considered an assembly of curved and bent circular tubes with various sizes of cross section. Here, steady state laminar flow of gas with Newtonian viscosity through a circular tube of diameter d was considered, thus the average flow velocity in the direction of flow (for example, the Z -direction) can be expressed by Eq. (2). Furthermore, considering the assembly of curved and bent circular tubes that require various sizes of cross section, gas permeability was introduced for the average cross section, thus Eq. (3), which is equivalent to Darcy's law, was obtained. Eq. (3) indicates that the Darcy velocity of a gas in a porous medium can be obtained from the pressure distribution of the gas within the voids.

$$v = \frac{d^2}{32\mu} \left(-\frac{dp}{dz} + \rho \bar{g} \right), \quad (2)$$

$$\vec{v}_s = \varepsilon \vec{v} = \frac{\kappa}{\mu} (-\nabla p + \rho \bar{g}), \quad (3)$$

where

\vec{v}_s : Darcy velocity [m/s],

ε : Void ratio of soil,

μ : Viscosity of the gas mixture [Pa·s],

κ : Gas permeability [m²].

Equation of conservation of mass for gas i

Here, Eq. (4) was used as the law of conservation of mass for gas i ($i = 1$, air; $i = 2$, hydrogen gas), taking into account the advection–diffusion and hydrogen gas leakage (leakage into the calculation area).

$$\frac{\partial}{\partial t} (\varepsilon \rho C_i) + \nabla \cdot (\rho C_i \vec{v}_s) = \nabla \cdot (D_{ij} \nabla \rho C_i) + \rho_i Q_i, \quad (4)$$

where

ρ_i : Density of gas component i [kg/m³],

C_i : Concentration of gas component i [wt%],

Q_i : Leakage flow rate of gas component i [1/s],

D_{ij} : Effective diffusion coefficient based on Fick's equation when

gas component i diffuses into gas component j in the porous medium [m²/s].

In Eq. (4), if $D_{12} = D_{21} = D$ and the equation is written for hydrogen gas ($i = 2$), then Eq. (5) is obtained, and this equation governs the variation in concentration of hydrogen gas.

$$\frac{\partial}{\partial t} (\varepsilon \rho C_2) + \nabla \cdot (\rho C_2 \vec{v}_s) = \nabla \cdot (D \nabla \rho C_2) + \rho_2 Q_2, \quad (5)$$

Equation of conservation of mass of the mixed gases

The equation of conservation of mass for the mixed gases was obtained by taking the sum of Eq. (4) for hydrogen gas and air, and is expressed by Eq. (6).

$$\frac{\partial}{\partial t} (\varepsilon \rho) + \nabla \cdot (\rho \vec{v}_s) = \nabla \cdot (D \nabla \rho) + \sum_i \rho_i Q_i, \quad (6)$$

Equation of state

In order to perform movement calculations that consider the thermodynamic properties of the gas mixture, the gas in the voids of the soil was assumed an ideal gas, and Eq. (7) was introduced as the equation of state. Hence, the differences in the thermodynamic properties of the mixed gases can be reflected in the relationship between pressure and velocity in Eq. (3).

$$p = \sum_i \frac{\rho_i RT}{M_i} = \sum_i \frac{C_i \rho RT}{M_i} = \frac{\rho RT}{M}, \quad (7)$$

where

T : Absolute temperature [K],

R : Gas constant [J/mol K],

M_i : Molecular weight of gas component i [kg/mol],

M : Average molecular weight of the gas mixture [kg/mol].

Finally, by substituting Eq. (3) and (7) into Eq. (6), Eq. (8) for pressure p can be obtained.

$$\frac{\partial}{\partial t} \left(\frac{\varepsilon M}{RT} p \right) - \nabla \cdot \left\{ \frac{\rho \kappa}{\mu} (\nabla p - \rho \vec{g}) \right\} = \nabla (D \nabla \rho) + \sum_i \rho_i Q_i \quad (8)$$

When solving Eq. (8), rather than dealing with the absolute value of the pressure p , it is more convenient to deal with the pressure difference of p' from a certain standard value (atmospheric pressure p_0). Therefore, Eq. (8) was converted into Eq. (10) by using the pressure difference p' through Eq. (9).

$$p = p' + p_0 + \rho_s (\vec{g} \cdot \vec{r}), \quad (9)$$

where

r : Position vector from a standard point, ρ_s : Density of air.

$$\frac{\varepsilon M}{RT} \frac{\partial}{\partial t} (p') + \frac{\varepsilon p}{RT} \frac{\partial}{\partial t} (M) - \nabla \cdot \left[\frac{\rho \kappa}{\mu} \{ \nabla p' - (\rho - \rho_s) \vec{g} \} \right] = \nabla (D \nabla \rho) + \sum_i \rho_i Q_i \quad (10)$$

Viscosity of gas mixture

Here, the viscosity of the gas mixture was estimated from Wilke's equation (Society for Chemical Engineers 1978 [23]) in Eq. (11).

$$\mu = \sum_i \frac{\mu_i}{1 + \sum_{j \neq i} \phi_{i,j} x_i/x_j}, \quad (11)$$

where

$$\phi_{i,j} = \frac{\left\{ 1 + (\mu_i/\mu_j)^{1/2} (M_j/M_i)^{1/4} \right\}^2}{2\sqrt{2(1 + M_i/M_j)^{1/2}}}$$

x_i : Mol fraction of gas component i [mol/mol],

μ_i : Viscosity of gas component i [Pa.s].

From the above, by solving the governing equation Eq. (10) using the finite difference method, the hydrogen gas concentration and pressure distribution were obtained.

2) Analysis Conditions

The object for the simulation analysis was the test field for the full-scale tests, 10 m square in the XY-plane, as shown in Fig. 2, to which 16 m of the surrounding original ground was added to give a 42-m square, as shown in Fig. 9, and the depth in the Z-direction was 4.8 m. This design was because that although the 10 m \times 10 m gastight membrane was laid at a depth of 2.0 m and was assumed to be the groundwater level in the test, there was a possibility that a very small amount of hydrogen gas could move into the original ground outside the test ground area, and into the area below the membrane surface.

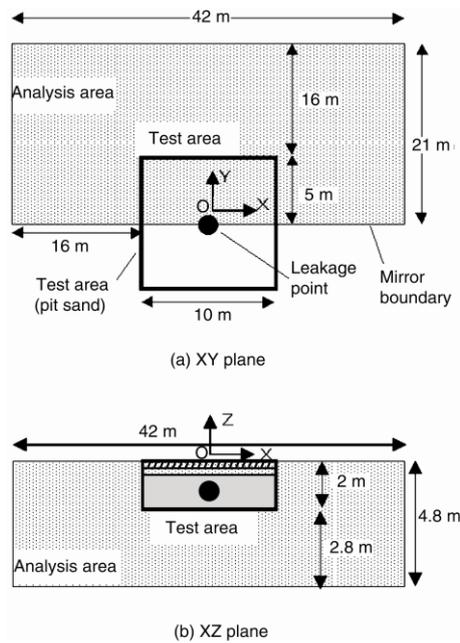


Fig. 9 Area subject to analysis (Phase 1)

Regarding the boundary conditions, the XZ plane that included the leakage point was considered a mirror boundary, thus $\partial C_2/\partial Y = 0$, $\partial p'/\partial Y = 0$. At the other side surfaces in the analysis area, it was assumed that $C_2 = 0$, $p' = 0$, at the other surfaces $\partial C_2/\partial Z = 5.0 C_2$, $\partial p'/\partial Z = 0$, and at the bottom surface $\partial C_2/\partial Z = 0$, $\partial p'/\partial Z = 0$. The number of analysis mesh was set at 92 in the X-direction, 46 in the Y-direction, and 34 in the Z-direction. A non-uniform mesh with a mesh size of 1.25–2 cm near the leak point and 15–40 cm elsewhere was used. The initial time interval in the analysis was 1 min, and there were 500 steps up to 400 h.

TABLE 2 PROPERTIES AND PARAMETERS USED IN THE ANALYSIS (PHASE 1)

(a) Soil Properties and parameters

Soil layer	Porosity [vol%]	Gas permeability [m ²]	Hydrogen diffusion coefficient [m ² /s]
Asphalt layer (0.0–0.05 m)	7.5	3.59×10^{-12}	1.00×10^{-6}
Crushed stone layer (0.05–0.2 m)	22.1	4.07×10^{-11}	6.82×10^{-6}
Pit sand layer (0.2–0.4 m)	18.3	6.19×10^{-12}	4.26×10^{-6}
Pit sand layer (0.4–0.8 m)	19.0	1.19×10^{-11}	4.42×10^{-6}
Pit sand layer (0.8–1.4 m)	22.6	1.40×10^{-11}	5.96×10^{-6}
Pit sand layer (1.4–2.0 m)	18.1	1.31×10^{-11}	3.65×10^{-6}
Original ground	18.8	3.23×10^{-11}	3.14×10^{-6}

(b) Properties of air and hydrogen gas

Gas	Mass per 1 mol [kg/mol]	Viscosity [Pa·s]
Air	2.90×10^{-2}	1.80×10^{-5}
Hydrogen	2.00×10^{-3}	8.80×10^{-6}

In addition, the property parameters required for the analysis are as shown in Tables 2(a) and (b), the actual values obtained in the full-scale tests were used as the soil property parameters. The gas permeability was obtained by passing pressurized air

through a sample of soil in a cylindrical container under the same temperature conditions as the ground, in accordance with the normal measurement method (Yasuda 1976 [24]), and calculating it from the measured pressure difference and flow rate. The diffusion coefficient was obtained according to the normal measurement method (Hamada et al. 2006 [11]; Troeh et al. 1982 [25]) by diffusing hydrogen gas in a soil sample in a cylindrical container and calculating it from the variation in concentration.

3) Analysis Results

Variation in the hydrogen gas concentration through the ground with time

Fig. 10 is a comparison of the variations over time of the hydrogen gas concentrations in the ground, obtained from the analysis and test, for the positions corresponding to points [1] to [3] in the test field shown in Fig. 5(a).

Comparing the variations over time of the hydrogen gas concentrations for analysis and test, it can be seen that for point [1] directly above the leak point, the steady state condition was reached sooner in the analysis results; however, at the other two points, the variations of the concentrations over time were reproduced well. In addition, the concentrations at steady state at those two points were values that almost correspond to the test result.

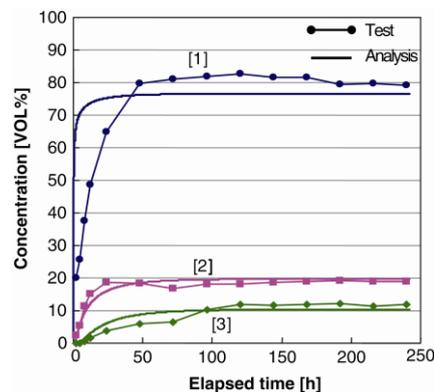


Fig. 10 Comparison of the variations of the hydrogen concentrations in the ground with time (Phase 1)

As a fact of the result that the steady state was reached sooner in the analysis than in the tests in the high-concentration area near point [1], the difference between the porosities of the test ground measured when preparing the test field and after the leak test was paid attention. At point GL -1.1 m directly above the leak point, the porosity after the test was 21.7 vol% while it was 17.0 vol% before the test. At this point, the porosity increased by 4.7 vol%, though the porosity changes at points other than this were within ± 3.0 vol%, that is, within the range of deviation of the porosity. This was postulated due to that dry hydrogen gas had passed though the void within the ground directly above the leak point for a long time, and soil moisture also moved according to the flow of hydrogen gas.

In the simulation, the void rate after the test was used. This means the void directly above the leak point was estimated rather bigger for the calculation of the value during leakage; thus, it was assumed that the result shows a condition in which advection–diffusion was easier. Consequently, the concentration in the high-concentration area directly above the leak point is to be underrated in the simulation.

Distribution of hydrogen gas concentration in the ground

Fig. 11 is a comparison of the distributions of hydrogen gas concentration in the ground from the analysis and from the test at steady state conditions after 240 h from the start of leakage. Comparing these results, it can be seen that, for example, the 10 vol% contour line was at ± 2 m to the side of the leak point, so the shapes of the distributions were almost identical.

Therefore, the result of the verification above demonstrated that the method of numerical simulation selected here was valid enough for the purpose of this research, the analysis of the movement of hydrogen gas in the ground, and it can be judged feasible to apply to these problems.

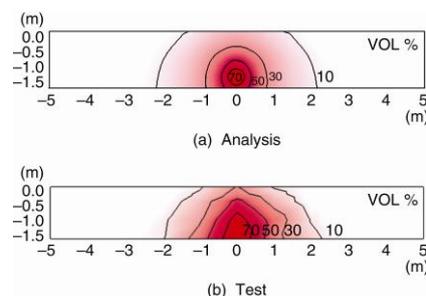


Fig. 11 Comparison of the distributions of the hydrogen gas concentrations in the ground for analysis and test (Phase 1, after 240 hours)

III. INVESTIGATION OF THE MOVEMENT CHARACTERISTICS OF LEAKED HYDROGEN GAS IN THE GROUND BY FULL-SCALE TEST CONSIDERING THE PIPELINE BACKFILL CONDITION (PHASE 2)

A. Experimental Investigation

1) Test Outline

In phase 2, another full-scale leak test of hydrogen gas was performed for where the space for the pipe alone was excavated and backfilled in the road, as shown in Fig. 12. This test was to simulate the real road burial site in the road of an urban area to investigate the characteristics of hydrogen gas movement and rising to the ground surface in the actual ground condition. In actual ground, pipelines are buried within trenches of approximately 60 cm in width excavated from the ground surface. Thus, in the event of a gas leakage, gas is expected to move through the boundary between the backfilling in the trench and the original ground outside the trench. In addition to the fact that the condition of gas movement was to be verified by the test, it was aimed to confirm whether the numerical simulation method, the validity of which was demonstrated in Chapter 2 above, is capable of reproducing such a condition.

Regarding the boundary conditions for hydrogen gas movement in the ground, an actual situation was assumed for the groundwater level at the bottom of the test field, and the rubber gastight membrane was not laid. The other conditions were the same as in test phase 1.

In addition, to confirm the uniformity of the compaction of the ground, boreholes were made at the leakage point and at +3.0 and -3.0 m in the Y-direction, and soil samples were taken for test at depths of $Z = -0.2, -0.5, -1.1,$ and -1.5 m. The results are shown in Table 3.

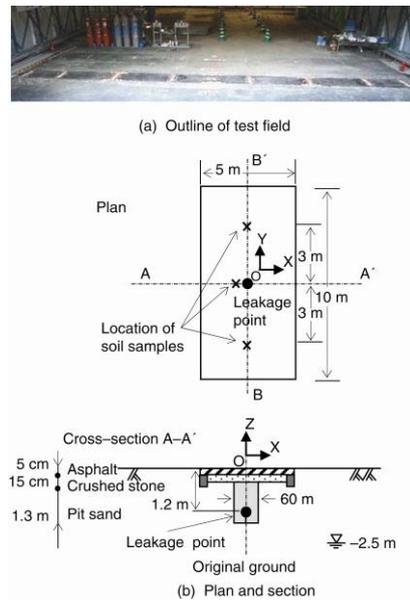


Fig. 12 Outline of the test field and composition of the test ground (Phase 2)

TABLE 3 TEST CONDITIONS (PHASE 2)

Gas type	Hydrogen (specific gravity 0.07)		
Leak rate	1000 (cc/min)		
Leak pressure	0.2 (kPa)		
Leak position	GL -1.2 (m)		
Postulated groundwater level	GL -2.5 (m)		
Test ground composition and properties * For Asphalt, dry density		Porosity (vol%)	Wet density (g/cm ³)
	Asphalt	6.9 ± 1.0	2.26 ± 0.01
	Crushed stone	16.9 ± 1.0	1.89 ± 0.03
	Pit sand	13.9 ± 2.6	2.02 ± 0.03
	Original ground	10.6 ± 3.0	2.05 ± 0.09

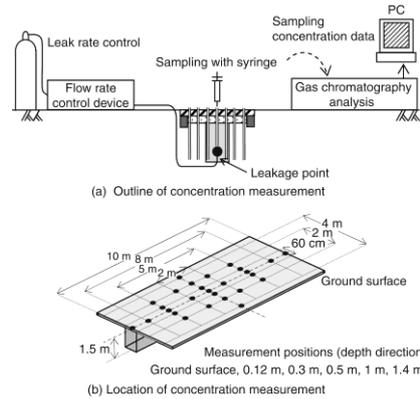


Fig. 13 Measurement of hydrogen gas concentration (Phase 2)

The hydrogen gas concentration in the ground was measured at the specific locations and depths shown in Fig. 13. The concentration of hydrogen gas released from the ground surface was also measured using the cart-type general-purpose gas detector shown in Fig. 4, as in phase 1.

2) Test Results

Change of hydrogen gas concentration in the ground with time

Fig. 14(b) shows the changes in hydrogen gas concentrations in the ground over time obtained in the tests at points [1] to [3] on cross section BB' in the test field as shown in Fig. 14(a) (co-ordinates (Y, Z) = (0.0, -1.0) (-1.0, -0.3) (-2.5, -0.12)).

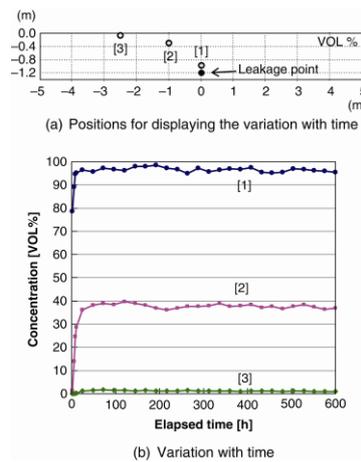


Fig. 14 Variations of the concentrations of hydrogen gas in the ground with time (Phase 2)

As with phase 1, the hydrogen can easily move upward. Thus, after about 50 h, the leak rate of hydrogen gas into the ground and the rate at which it escapes into the atmosphere are balanced, and steady state occurs.

Distribution of hydrogen gas concentration in the ground

Fig. 15 shows the distribution of hydrogen gas concentration in the ground on cross section AA' and cross section BB' during the steady state condition after 600 h from the start of hydrogen leakage. The concentration distribution shows a generally symmetrical spread to the left and right about the leakage point, reflecting the fact that the test field was made uniform.

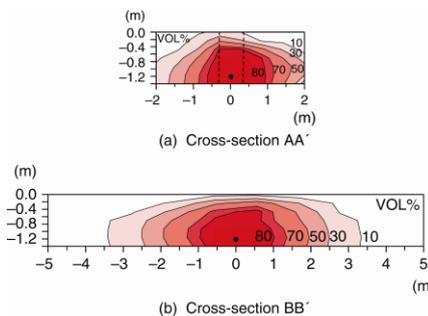


Fig. 15 Distributions of hydrogen gas concentrations in the ground (Phase 2, after 600 hours)

In addition, as in phase 1, the hydrogen gas can easily diffuse from the asphalt surface into the atmosphere, so the concentrations in the upper layers of the test field and to the sides of the leakage point were comparatively low. The area where the hydrogen gas concentration in the ground was 10 vol% or higher was distributed within about ± 2 m in the X-direction with the leakage point as center, and likewise about ± 3 m in the Y-direction.

Distribution of hydrogen gas concentration released to the ground surface

Fig. 16 shows the distribution of hydrogen gas concentration in the X- and Y-axis directions on the surface, measured with the cart-type gas detector under steady state conditions after 600 h had elapsed from the start of leakage.

The concentration distribution spread symmetrically left and right around the center directly above the hydrogen gas leak point. In the X-direction, the concentration was high at 1.0 vol% since in this direction the pipe space had been excavated to a 60 cm width, backfilled, and the asphalt was jointed. The concentration reduced with distance. In the Y-direction, the distribution was similar to the concentration in the ground and directly above the leak point, and it was high at 0.2 vol%. The concentration reduced with distance from the leakage point. The hydrogen gas released from the ground was distributed about ± 2.5 m about the leak point, and the cart-type gas detector, whose hydrogen gas concentration detection limit is 0.008 vol%, was capable of detecting the hydrogen within this area. This result indicates that it is possible to detect the released hydrogen gas by periodic leak inspection using gas detectors on the surface of the road, as required by the aforementioned Gas Business Act.

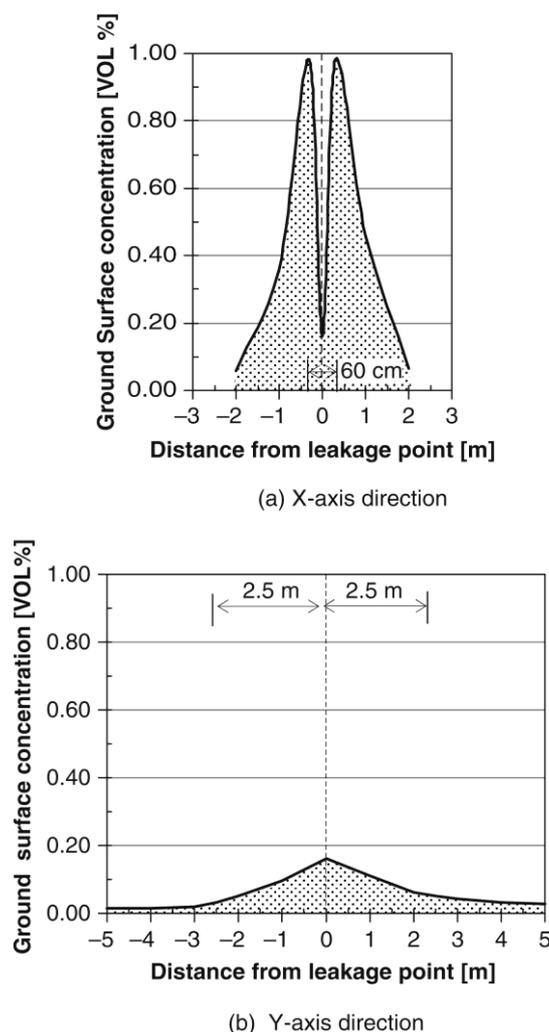


Fig. 16 Distribution of hydrogen gas concentration at the ground surface (Phase 2, after 600 hours)

B. Verification of the Numerical Simulation Method on the Movement of Hydrogen Gas in the Ground

1) Analysis Conditions

The numerical simulation analysis area was the 5 m \times 10 m area in the XY plane of the full-scale test field shown in Fig. 12 to which 3 and 5.5 m of the surrounding original ground were added, as shown in Fig. 17, to give a total of 16 m square, and the depth in the Z-direction was 2.5 m.

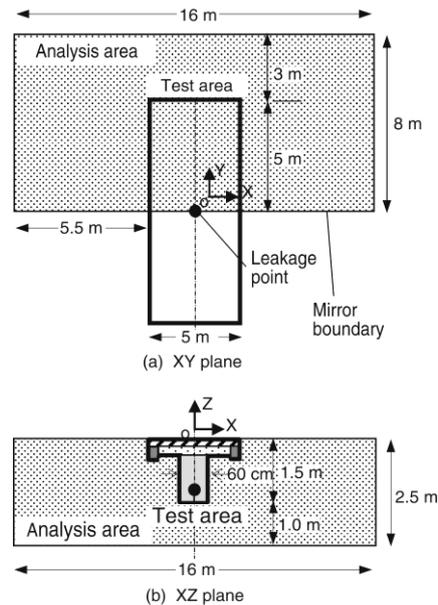


Fig. 17 Area subject to analysis (Phase 2)

Regarding the boundary conditions, the XZ plane that included the leakage point was taken to be a mirror boundary, so $\partial C_2 / \partial Y = 0$, $\partial p' / \partial Y = 0$. It was assumed that at the other side surfaces in the analysis area $C_2 = 0$, $p' = 0$, at the other surfaces $\partial C_2 / \partial Z = 5.0 C_2$, $\partial p' / \partial Z = 0$, and at the bottom surface $\partial C_2 / \partial Z = 0$, $\partial p' / \partial Z = 0$. The number of analysis mesh was set at 56 in the X-direction, 56 in the Y-direction, and 67 in the Z-direction. A non-uniform mesh with a mesh size of 2 cm near the leakage point and 2–50 cm elsewhere was used. The initial time interval in the analysis was 1 min, and there were 800 steps up to 672 h.

In addition, the property parameters required for the analysis are as shown in Table 4, the actual values obtained in the full-scale tests were used as the soil property parameters.

TABLE 4 PROPERTIES AND PARAMETERS OF SOIL USED IN THE ANALYSIS (PHASE 2)

Soil layer	Porosity [vol.%]	Gas permeability [m ²]	Hydrogen diffusion coefficient [m ² /s]
Asphalt layer (0.0–0.05 m)	6.9	3.04×10^{-12}	4.33×10^{-7}
Crushed stone layer (0.05–0.2 m)	16.9	1.94×10^{-11}	3.71×10^{-6}
Pit sand layer (0.2–0.5 m)	14.1	2.11×10^{-12}	2.43×10^{-6}
Pit sand layer (0.5–1.1 m)	14.1	7.01×10^{-12}	1.93×10^{-6}
Pit sand layer (0.1–1.5 m)	13.5	6.02×10^{-12}	1.89×10^{-6}
Original ground	10.6	8.50×10^{-13}	7.78×10^{-7}

2) Analysis Results

Variation in the hydrogen gas concentration through the ground with time

Fig. 18 is a comparison of the variations over time of the hydrogen gas concentrations in the ground, obtained from the analysis and the test, for the positions corresponding to points [1] to [3] in the test field as shown in Fig. 14(a).

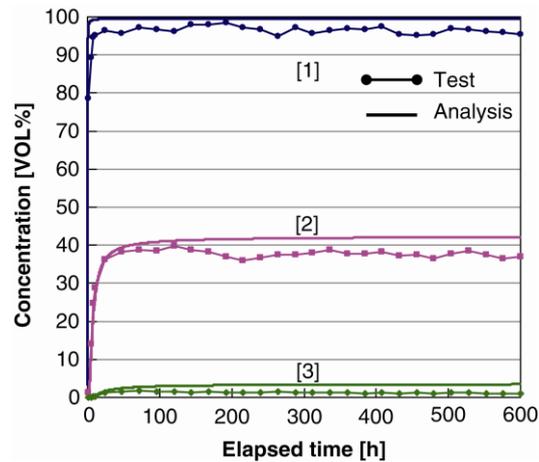


Fig. 18 Comparison of the variation in hydrogen gas concentration in the ground with time (Phase 2)

As seen in Fig. 18, from the low-concentration area to the high-concentration area, the variations in concentrations over time were reproduced well. In addition, the concentrations at steady state were values that almost correspond to the test result.

Here, in phase 2 (Fig. 18), when compared with phase 1 (Fig. 10), the difference between the results of the test and the numerical simulation was not significant even at the point [1] near the leakage point. It was assumed that since the compaction condition of the test ground in phase 2 was better when compared with that in phase 1, the porosity in phase 2 was smaller, which resulted in the effect of porosity variation due to permeation of dry hydrogen being less.

Distribution of hydrogen gas concentration in the ground

Fig. 19 is a comparison of the distributions of hydrogen gas concentrations in the ground at cross sections AA' and BB' from the analysis and the test at steady state conditions after 600 h from the start of leakage. It can be seen that the shapes of the distributions are in virtual agreement with the distributions of hydrogen gas concentrations at steady state conditions in both transverse and longitudinal sections.

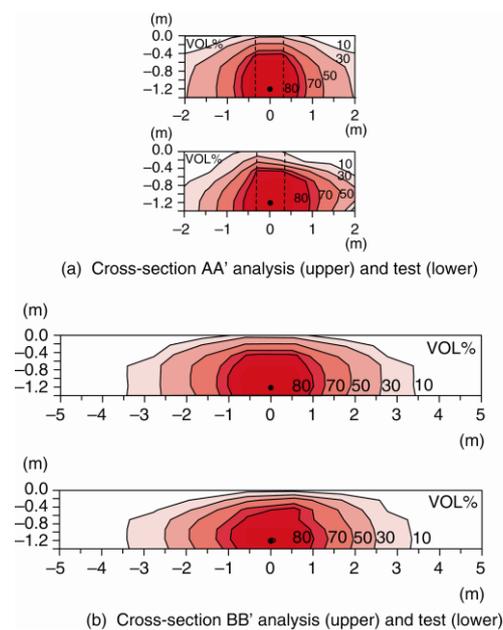


Fig. 19 Comparison of the distribution of the hydrogen gas concentration in the ground for analysis and test (Phase 2, after 600 hours)

Based on the test results as described above, the numerical simulation method used was concluded to provide generally valid outcome in the condition similar to the real road burial site in the road of an urban area, where it is expected that gas moves through the boundary between backfill within the trench and the ground outside the trench in a potential leakage incident.

In general, for a buried gas pipeline, the soil conditions of the backfill soil used around the pipe are specified by a government organization, and the water content, porosity, and particle size are measured through soil tests. Several methods have been proposed for estimating the gas permeability and the diffusion coefficient from these measured results (Yasuda 1976 [24]; Hamada et al. 2006 [11]; Troeh et al. 1982 [25]). Using these input parameters to conduct a numerical simulation, as was

applied in this study, it is possible to conveniently obtain the extent of movement of leaked gas and other items useful for maintaining safety.

However, if a higher supply pressure is assumed, it is desirable that the movement of water be considered, such as a water–hydrogen–air two-phase, three-component model (gas–liquid, two-phase model).

IV. CONCLUSIONS

In this study, full-scale hydrogen gas in-ground leakage tests and numerical simulations of the movement of hydrogen gas through the ground were performed with the objective of studying and researching safety and maintenance issues, which are important when investigating the supply of hydrogen gas through buried pipelines.

The results obtained in this study can be summarized as follows.

- 1) The specific gravity of hydrogen is smaller than that of air, so it can easily travel upward. At about 50 h after the start of leakage, a steady state is achieved with a balance between the rate of leakage of hydrogen gas into the ground and the rate at which it is released into the atmosphere.
- 2) The area where the hydrogen gas concentration in the ground was 10 vol% or higher was distributed within an area about ± 2 to 3 m with the leak point as the center. It was considered that the potential of diffusion over a wide area and ingress into houses from the road boundary is low.
- 3) Distribution of hydrogen gas released from the ground in concentration of 0.008 vol% or higher, which the cart-type gas detector is able to detect, was about ± 2.5 m of a point directly above the leak point. This result indicates that it is possible to detect leakage by periodic leak inspection using a gas detector on the surface of the road, as required by the Gas Business Act, in the occasion of accidental hydrogen gas leakage.
- 4) The numerical simulation of the movement of hydrogen gas in the ground in this study, based on Darcy's law and Fick's law, was generally capable of reproducing the states of leakage of hydrogen gas into the ground at full scale under various boundary conditions, thus it was considered valid. Therefore, by performing numerical simulations, as adopted in this study, using suitable input parameters, it is possible to conveniently obtain the extent of movement of the hydrogen gas and other information that is useful for ensuring safety.

ACKNOWLEDGMENT

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