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Original article

Analysis of pipe thickness reduction according to pH in FAC facility with *In situ* ultrasonic measurement real time monitoring

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A B S T R A C T

Flow accelerated corrosion (FAC) is a type of pipe corrosion in which the pipe thickness decreases depending on the fluid flow conditions. In nuclear power plants, FAC mainly occurs in the carbon steel pipes of a secondary system. However, because the temperature of a secondary system pipe is over 150 °C, *in situ* monitoring using a conventional ultrasonic non-destructive testing method is difficult. In our previous study, we developed a waveguide ultrasonic thickness measurement system. In this study, we applied a waveguide ultrasonic thickness measurement system to monitor the thinning of the pipe according to the change in pH.

The Korea Atomic Energy Research Institute installed FAC-proof facilities, enabling the monitoring of internal fluid flow conditions, which were fixed for ~1000 h to analyze the effect of the pH. The measurement system operated without failure for ~3000 h and the pipe thickness was found to be reduced by ~10% at pH 9 compared to that at pH 7. The thickness of the pipe was measured using a microscope after the experiment, and the reliability of the system was confirmed with less than 1% error. This technology is expected to also be applicable to the thickness-reduction monitoring of other high-temperature materials.

1. Introduction

FAC has damaged many facilities in thermal and nuclear power plants worldwide over the past decades. For example, the accidents current in Surry Unit 2 in 1986 and Mihama Unit 3 in 2004 resulted in numerous injuries and economic losses [1–4]. Pipe damage from FAC can seriously threaten the safety of nuclear power plants, and main pipe damage can lead to economic losses. Flow accelerated corrosion (FAC) is the most common source of rupture accidents, causing a reduction in the thickness of nuclear pipes. In such cases, corrosion and the build-up of a thin film occur owing to the flow of the internal fluids. This phenomenon occurs in hydrodynamics, hydrochemical (water chemistry) environments, and materials under certain internal fluid conditions. Major effects are due to hydrochemical (e.g., pH, dissolved oxygen, ionic concentration, impurity content, and fluid temperature) and hydrodynamic (e.g., velocity, pressure, pipe geometry, and pipe surface roughness) factors [6–9]. In the case of nuclear power plants, FAC occurs in the secondary system pipe through the interaction of these factors according to the operating environment conditions. FAC is extremely important because the plant operates for a long period of time and can be effectively mitigated using appropriate structural materials or by controlling hydroxylactic environmental factors and slowing the FAC velocity [4–7].

Studies on hydrochemical conditions are being conducted as a way to prevent FAC, and real-time monitoring using non-destructive technology is being applied to analyze the effects of such corrosion. Ultrasonic non-destructive testing methods are known to be suitable for evaluating the integrity of metal pipes. The thickness was measured using a pulse echo method with a widely known ultrasonic wave speed, or surface waves were used to detect defects. This method can be analyzed at room temperature because the probe is in direct contact with the pipe, and because a probe in direct contact with the pipe wall during measurement must be kept under the Curie temperature [9–11]. However, it is difficult to assess the integrity of the pipes using a general ultrasonic test method because the main pipe is maintained at a high temperature of more than 150 °C during the plant operation. Owing to these

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problems, the pipe thickness is generally measured using UT(ultrasonic testing) during the shutdown period of the plant at room temperature; however, owing to the difficulty of real-time monitoring, immediate action should be delayed when a problem occurs. A technology that enables continuous thickness monitoring of high-temperature pipes with a high risk of degradation has been assumed [12–15].

A waveguide system was developed as a technology for monitoring high-temperature pipes and was used to analyze the thickness reduction phenomenon owing to changes in the internal fluid flow rate in high-temperature pipes. The internal fluid velocity is an important factor in which FAC occurs, enabling a precise analysis of the phenomenon in which the thickness reduction rate increases as the flow rate increases. However, an accurate monitoring of the thickness change through signal processing should be conducted, as errors in the measurements exist and methods are needed to increase the reliability of the signals and reliably protect the transducer from high temperature [16–20].

This study validates a measurement system that can continuously monitor pipe thinning through a change in the pH, a factor that significantly affects the FAC during operation in hydrochemical (water chemistry) environments through non-destructive methods. FAC-proof facilities were used to maintain the actual nuclear power plant environment for a long period of time. In particular, the internal fluid conditions (temperature, pressure, and dissolved oxygen (DO)) affecting FAC were fixed for a comparative analysis of the thickness reduction rate caused by the pH, and the FAC-proof facilities were operated for approximately 1000 h at pH 7 and above pH 9. The reliability and verification of the measurement system were compared directly by cutting the pipe where the experiment was completed, and the thickness was measured using a microscope.

2. Flow accelerated corrosion and hydrochemistry environmental factors

Corrosion in general metallic materials is a phenomenon in which damage occurs owing to chemical reactions with materials in a state where the surrounding environment is stagnant. There are a number of ways in which such corrosion occurs, including general and crevice corrosion, occurring on a general metal surface. By contrast, FAC is a phenomenon in which corrosion is accelerated by the flow of a fluid on a surface in contact with a metal material. When the fluid flow exceeds a certain rate, the corrosion oxide film generated on the surface dissolves, the thickness becomes thin, and the base metal cannot be protected. Compared to general corrosion, the corrosion rate was relatively higher. During FAC, the dissolution rate of the oxide film and the oxide film formation rate become steady, and the corrosion rate of the base material is maintained at a constant [6].

FAC leads to a reduction in the overall thickness of the pipe, unlike general corrosion in local areas, which leads to damage owing to the operating environment of the power plant. Such damage leads to significant damage because it is destroyed in a wide area without prior warning, such as leakage caused by local damage. As shown in the figure, the surface of the carbon steel material owing to FAC grows with microscopic spots occurring at the beginning and expands to the entire surface [21].

In general, the conditions under which FAC occurs are affected by temperature, flow rate, alloy composition, redox potential, pH, and pipe shape. Oxide film and magnetite are generated on the surface, and the reaction formula is as follows [22] (see Fig. 1):

$$Fe + 2H_2O \Rightarrow Fe^{2+} + 2OH^- + H_2 \tag{1}$$

$$Fe^{2+} + 2OH^- \Leftrightarrow Fe(OH)_2 \tag{2}$$

$$Fe(OH)_2 \Rightarrow Fe_3O_4 + H_2 + 2H_2O \tag{3}$$

The soluble ferrous ions, Fe^{2+}, generated above are diffused into the solution through a porous oxide film. At the oxide film/solution interface, the magnetite oxide film is dissolved by hydrogen generated during the corrosion process, as indicated in the following:

$$1/3Fe_3O_4 + (2-b)H^+ + 1/3H_2 \Leftrightarrow Fe(OH)^{b(2-b)+} + (4/3-b)H_2O^{b\ (=0,\ 1,\ 2,\ 3)} \tag{4}$$

To suppress the occurrence of FAC, nuclear power plants in Korea maintain a secondary water chemical environment of pH 9.0–10.0. The pH of the pressurized water reactor (PWR) was initially adjusted using phosphate followed by ammonia (all volatile treatment). However, ammonia has a low basicity and is extremely volatile in each line. Therefore, the pH of the two-phase wet vapor region increases and the pH of the single-phase solution decreases. To solve this problem, ethanol amine, which has a high volatility and is effective in inhibiting FAC, was used. It is known that, at above 200 °C, the solubility of magnetite decreases when the pH is between 4.9 and 9.8, and the solubility of magnetite increases at above pH 10.3. This means that FAC is affected by the pH. The magnetite solubility, which is related to the FAC rate, is related not only to the temperature, pH, and material composition, but also to other test environments, and thus the interrelationship of these factors should always be considered [21–23].

3. High-temperature pipe thickness measurement method

The thickness measurement using a pulse-echo ultrasonic wave method measures the peak response time between the received signals of the back wall echo. This principle measures the thickness by calculating the transmission time of the ultrasonic velocity of each echo signal and converting it into the distance. The rate of reduction of the pipe thickness varies depending on the fluid condition (such as the temperature, velocity, pressure, and pH), and because surface changes occur owing to the reduction, measurement errors owing to a surface diffused reflection must be minimized. In particular, when thinning occurs from corrosion, the surface has a scallop pattern shape, and a difference in density with the base metal occurs because of the corrosion products, and thus the precision may be degraded owing to the diffused reflection of the ultrasonic signal. To solve this problem and minimize the error, the influence of various environments must first be reduced. The ultrasonic velocity varies depending on the temperature change of the test material, and the measurement errors can be minimized by keeping the experimental environment temperature constant. Second a physical error is caused by the contact conditions between the ultrasonic coupling material and the transducer. This can be minimized by consistently maintaining close contact through a custom clamp fabrication, as shown in Fig. 2. Finally, an error occurs owing to the roughness of the measurement surface. As the thickness becomes thinner and corrosives are produced, the surface is difficult to control; thus, the received signal can be processed to minimize errors [16,20].

In addition to these causes, if the test material increases to a high temperature (over 150 °C), the measuring transducer may be exposed to high temperatures. In the case of piezoelectric elements,

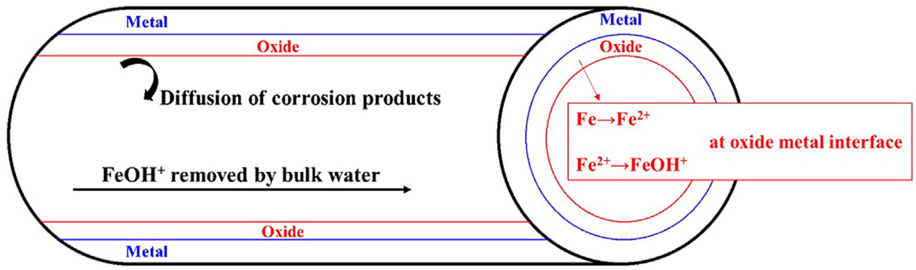

Fig. 1. Schematic diagram of the pipe thickness reduction through FAC [22].

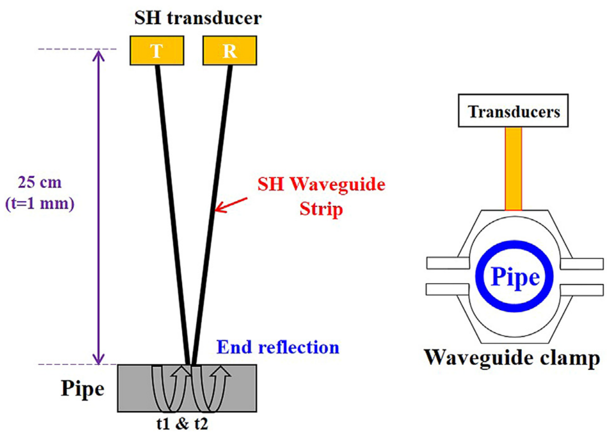

Fig. 2. (**a**) The concept of a pair of waveguide strips for high-temperature thickness monitoring and (**b**) installed waveguide clamp system for thickness monitoring on a pipe.

if the temperature rises above the Curie temperature, it leads to a depolarization of the material, which causes the piezoelectric properties to change, resulting in poor accuracy. In addition, when the temperature of the test pipe temperature increases, problems with the couplant also occur. The couplants used in ultrasonic flaw detectors evaporate when they reach a high temperature. Therefore, although there are cases in which high-temperature-only couplants are used evaporation occurs when used for a long time, and thus it is necessary to prevent heat damage by applying a solid couplant (gold plate). In this study, to satisfy both, a dedicated coupler capable of stably transmitting ultrasonic waves at 200 °C was used along with a metal couplant (gold plate). In particular, gold has a low coefficient of thermal expansion, and thus it changes less at high temperatures and can transmit ultrasonic signals without loss [18,19].

4. On-line high-temperature pipe-thickness monitoring system

A method for measuring the thickness of a high-temperature pipe surface by directly contacting the transducer was developed; however, as described above, it may cause a deformation, and therefore, a dedicated high-temperature transducer is mainly used for measurement. The high-temperature transducer operated reliably over a certain temperature range, but was unsuitable for long-term use when attached to a high-temperature surface for thermal degradation of transducer material specimens because a high-temperature specimen and a transducer are physically exposed to heat in close proximity.

To overcome this problem, we developed a waveguide method that allows the transducer to be separated from the specimen and minimize the transmission loss of the ultrasonic waves. This allows the use of a pair of stainless steel (SS) plates, as shown in Fig. 2. Stainless steel has a high corrosion resistance, and thus can maintain its integrity at high temperatures. Using a thin plate with a length of 20 cm or longer not only minimizes the effect of the heat, it also allows the transducer to be moved away from the high-temperature specimen. The aim is to protect the transducer from high temperatures by installing a test specimen at one end of the plate and a transducer at the other end. In fact, when the surface of the test object increases to approximately 150 °C, the surface of the buffer-rod type high-temperature dedicated transducer is affected by a temperature of more than 100 °C, whereas the temperature at the surface of the transducer located far from the plate is approximately 40 °C or less. The impact caused by the heat problem is reduced.

There is also a need for a method to minimize the effects of heat and minimize losses owing to an ultrasonic transmission. To measure the thickness, a longitudinal transducer is generally used; however, if a thin plate is applied, the energy transfer rate is low. The shear horizontal (SH) wave has a high transmission rate in a thin plate and has little dispersion characteristics; therefore, it is effective for an energy transfer using a thin plate. When the transducer and waveguide are in exact vertical contact, the shear horizontal wave is stably transmitted. Accordingly, a dedicated transducer for the shear horizontal modes is used, and a pitch/catch system was constructed using a pair of waveguide strips(thickness:1 mm, width:25 mm) [19]. The length of the strip was 25 cm in length to minimize the effect from the heat of the pipe. This system only receives signals transmitted from the pipe specimen, which can prevent unwanted reflected noise signals at the end of the plate, resulting in a high S/N ratio. In addition, for an accurate contact with the pipe, a dedicated clamp was manufactured, and the developed waveguide system was installed on the specimen, as shown in Fig. 2.

The changes in the specimen temperature also affect the ultrasonic velocity. The speed of the ultrasonic waves varies depending on the material, and as the temperature increases, the ultrasonic velocity decreases. In particular, for precise thickness monitoring at high temperatures, it is necessary to know the ultrasonic velocity according to the temperature change of the material. The pipe material used in this study was an SA 106 Gr.B. For a precise speed evaluation, the change in ultrasonic velocity was measured while increasing the temperature of the specimen and was used as background data. The change in ultrasonic SH wave velocity owing to changes in temperature from 0 °C to 250 °C in carbon steel pipes (SA 106) is equivalent to Equation (5), which is the result of measurements conducted through the experiments [20]:

$$V\ [m/s] = 3365.8 - 0.67728T \quad (5)$$

Through the above equation, more accurate signals can be received through signal processing after correction according to the velocity. The pitch/catch method does not have a main bang signal, but the signal-to-noise ratio of multiple reflected signals at the rear wall of the pipe is important because the signal is weak at the end of the transmission waveguide. To solve this problem, a thickness measurement program was devised allowing the gate to move in real time and accurately measure the reflected flight time. As shown in Fig. 3, the gate tracking and thickness measurement system was programmed and built using LabView. Fig. 3 (a) shows the full screen of the program and, in the upper-left corner, an oscilloscope allowing real-time signal changes to be confirmed can be seen. Under the oscilloscope screen, a setup window was produced to adjust the position of each gate to the x and y axes, and on the right side of the screen, the thickness data measured at a constant time interval can be recorded and viewed in real time. The lower part of the measurement screen is designed such that the values set for compensation according to temperature can be entered. As shown in Fig. 3 (b), the purple (gate 1) and sky blue (gate 2) gates are set to move according to the wall echo signals moving by the environment. The first signal received in Fig. 3 (b) is the surface reflection signal reflected from the end of the waveguide strip. The two subsequent signals reflect the pipe back wall signal and return signals, allowing the thickness to be evaluated based on the two signals. However, the thickness of the pipe decreases over time by FAC, causing the entire signal to move along the x-axis. In addition, as the temperature of the pipe changes, the wave speed changes, allowing the signal to move along the x-axis. To resolve this problem, a gate was set to track the signal when it was moved. In this case, even if the error of the echo signal is out of range, it can be corrected by the movement of the gate, and if the error exceeds the ability to correct it, the alarm is set to sound, allowing the user to immediately notice it. Gate 1 (purple gate) is intended to obtain the first reflection signal that came out through the pipe, and gate 2 (sky blue gate) was intended to obtain the second reflection signal. For accurate thickness data, the distance between the signal received from the pipe and the contact point of the purple and light blue gates t_1 and t_2 is automatically measured and converted into the thickness and averaged hundreds of times per second. The developed program automatically calculates the delta t value based on the peak values of gate 1 and gate 2, and calculates the thickness automatically through a calibration. The pipe thickness calculated in real time was recorded at a certain interval, as previously described, allowing an assessment of the change in thickness.

5. FAC test in proof facility

5.1. FAC-proof facility

The Korea Atomic Energy Research Institute has established an FAC demonstration facility to control the pressure and chemical components, as shown in Fig. 4, and can be checked in real time through the control monitoring room. This facility was manufactured and verified to be very similar to the nuclear power plant environment, and not only can the environmental changes of internal fluids but also control the changes in flow. The test section without the thermal insulation material was manufactured to analyze the thickness reduction effect under each test condition.

The pipes of the test section in the facility are made of carbon steel (SA 106), with dimensions of 60.4 mm in outer diameter, 50 mm inner diameter, and 5.20 mm in nominal wall thickness. The pipe material compositions are listed in Table 1. To monitor the reduction in the thickness of the pipe using an ultrasonic method, a test section with a length of 750 mm was prepared and the insulation was not covered. The proposed waveguide system was installed in the test section, as shown in Fig. 5, and the thickness of the contact point was evaluated in real time.

5.2. FAC-proof test (pH effect)

A thickness reduction from FAC can occur owing to various effects. In this study, to confirm the change in the FAC characteristics according to the pH effect, it was controlled by changing the pH of the internal fluid from 7 to 9.5 in a nuclear power plant thinning demonstration test facility. The pipe thinning test was carried out continuously at 10 m/s, 150 °C, and 10 bar for approximately 950 h

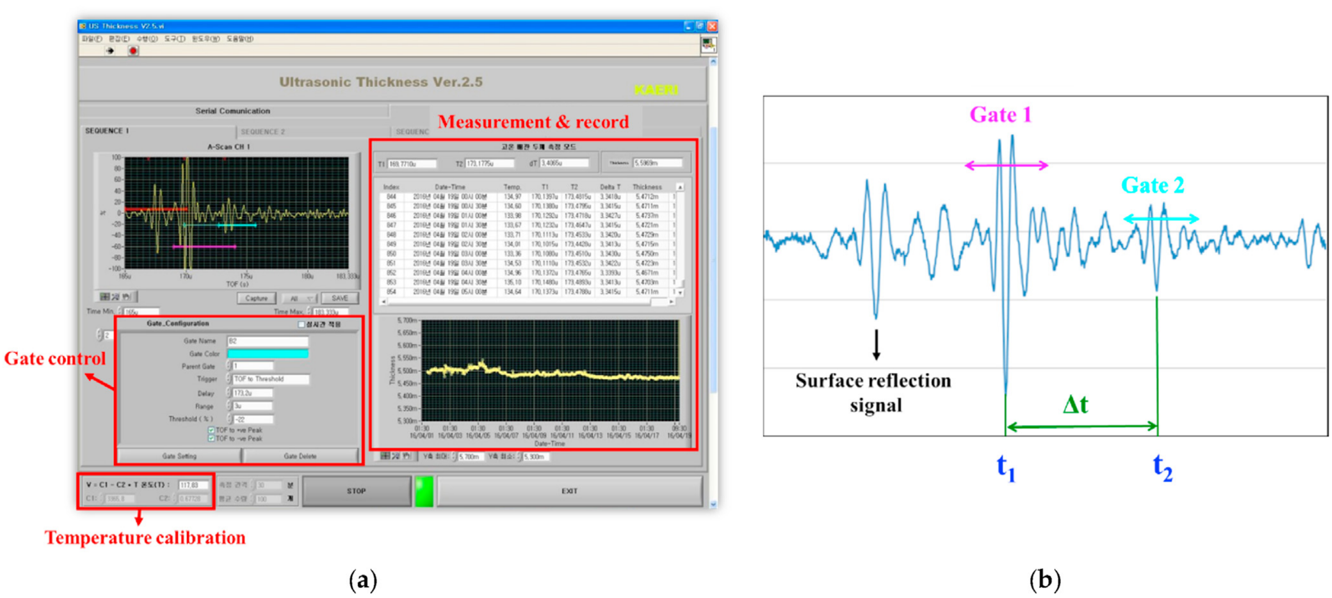

Fig. 3. The real-time thickness measurement method: **(a)** program developed by applying a moving gate and **(b)** typical reflection signal and gate setup.

(a) (b)

Fig. 4. The FAC-proof facility in Korea Atomic Energy Research Institute (KAERI).

Table 1
Chemical composition of the material (SA 106).

Material.	Cr	Mo	Cu	Mn	Ni	Si	C	P	S
SA106	0.02	0.01	0.04	0.37	0.02	0.22	0.19	0.008	0.006

at pH 7.0, and 1150 h at pH 9.1 and pH 9.5. Through heavy nitrogen gas removal, the dissolved oxygen (DO) based on the shear of the ion-exchange resin was kept below 4 ppb. Under each condition, the thickness reduction can be measured from 10 to 75 μm, as shown in Table 2. To compare the amount of thinning at the same time, the thinning rate must be quantified. The nuclear industry uses units of millimeters per year, and the conversion was conducted under the same conditions. As shown in Fig. 6, it was confirmed that the thinning rate was the fastest at pH 7, and as the pH gradually increased, a thinning of the thickness slowly occurred. The thickness reduction can only be judged by the influence of the change in pH because the internal conditions other than the pH are fixed.

It is known that FAC is suppressed as the pH increases; however, because all other environmental factors must be considered, the flow velocity inside the pipe is kept constant at 10 m/s, the fluid temperature is kept at 150 °C, inside pipe condition is constant kept dissolved oxygen and internal pressure. When the pH of the fluid was increased to 7.0, 9.1, and 9.5, the amount of thickness change owing to the FAC suppression effect was measured using an ultrasonic flow detection technique. An increase in pH also affects the reduction in thickness because the solubility of magnetite decreases. However, if rising above a certain temperature, the solubility increases even if the pH increases; therefore, a proper adjustment is needed. The amounts of thinning of the thickness of 75 μm at pH 7.0, 18 μm at pH 9.1, and 10 μm at pH 9.5, indicating that the least thinning occurred at pH 9.5. Under the condition of pH 7.0, the thickness reduction rate of the pipe was 0.68 mm/year as a result of the experiment, and it was confirmed that when the pH was increased to 9 or more, it decreased to 0.14–0.07 mm/year. The decrease in pH under the same temperature, pressure, and flow rates conditions led to an increase in the rate of thickness reduction. As a result of the FAC rate, the thickness reduction rate over time was reduced to 20% at pH 9.1, and approximately 10% at a higher rate of 9.5. Under relatively high pH conditions, the thickness reduction was 10–20 μm, which can be seen as no reduction in thickness because it is included within the measurement error range of the waveguide measurement system.

After all the FAC proof experiments were completed, the pipe thickness was directly measured using a microscope at the location (Waveguide measurement system was installed) to verify the reliability of high-temperature real-time UT measurements. The measurement area was divided according to the width of the probe (10 mm) used to measure the room-temperature UT. The thickness of the cutting pipe was measured using a microscope (SHAHE; Digital thickness gauge, 5331-10 S). The red square in Fig. 7 shows the location where the waveguide measurement system was installed and measured repeatedly over a total of five times. The average thickness measured was 5.09 mm (Fig. 7), with an error range of approximately ±0.002 mm. The average thickness of the waveguide system was approximately 5.09 mm, and the error range was ±0.007 mm, which is similar to the error rate of a typical ultrasonic thickness measurement device, confirming the reliability of the results of the development system. The measurement results

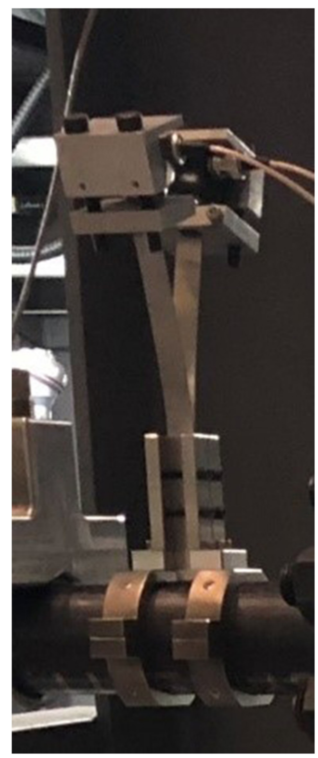

Fig. 5. A waveguide system installed in an area without insulation.

Table 2
The pipe wall-thickness monitoring of a carbon steel pipe in the flow accelerated the corrosion proof test facility: Different wall-thinning ratios observed depending on the flow velocities.

	vel	temp	Pressure	pH	Thinning	Operation time	FAC rate
Condition 1	10 m/s	150 °C	10 Bar	pH 7.0	75 μm	950 h	0.68 mm/year
Condition 2	10 m/s	150 °C	10 Bar	pH 9.1	18 μm	1150 h	0.14 mm/year
Condition 3	10 m/s	150 °C	10 Bar	pH 9.5	10 μm	1150 h	0.07 mm/year

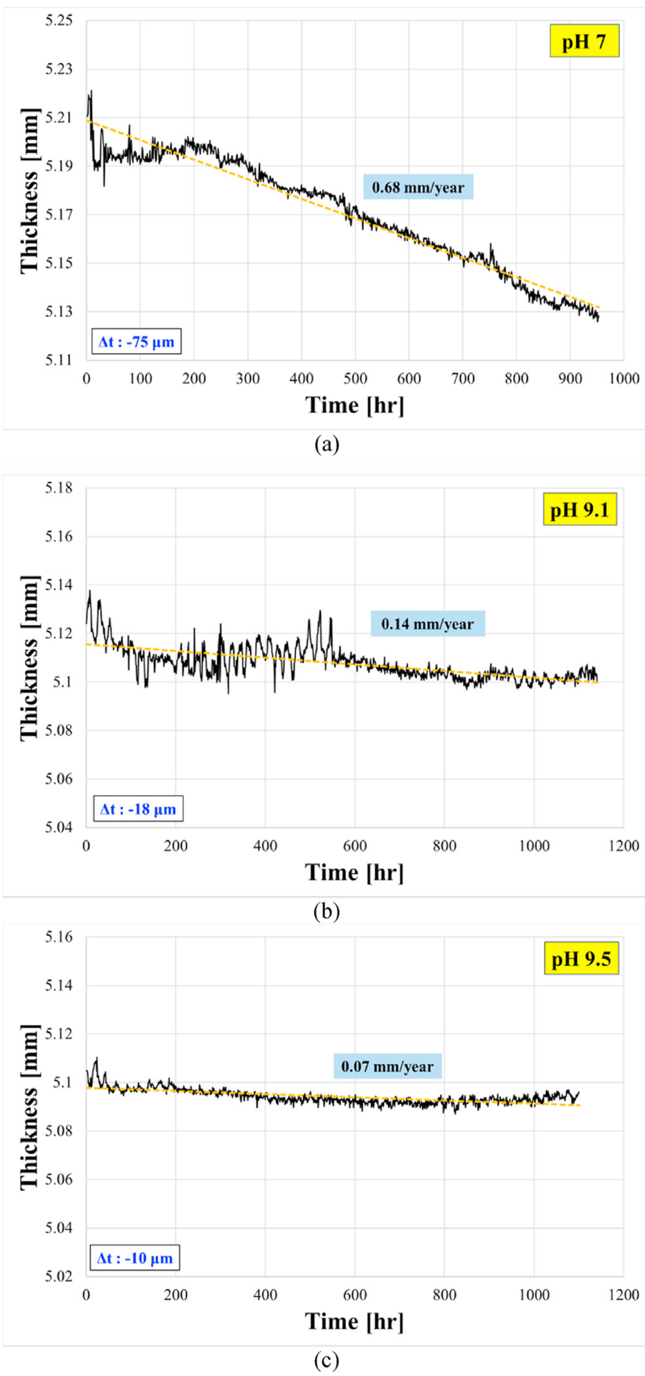

Fig. 6. Pipe wall-thickness thinning rate of carbon steel pipe in the flow accelerated corrosion proof test facility: Different wall-thinning ratios observed depending on the pH: **(a)** pH 7, **(b)** pH 9.1, and **(c)** pH 9.5.

obtained using the two methods are shown in Fig. 8. The thickness measured by the microscope was relatively narrow because the waveguide measurement point of the local area was repeatedly measured. However, each of the two methods has the same result as the error range of the equipment, and thus it can be considered an accurate measurement. Furthermore, the reliability of the measurement system was demonstrated because the measurement results using the waveguide system were similar to those measured under the microscope.

6. Results and discussions

The technique for measuring the thickness using the ultrasonic flaw detection method is generally applied at room temperature. This is because damage to the transducer may occur, causing measurement errors. However, we not only developed a technology that can measure the thickness of a pipe in real time, but also developed a technology that can reliably monitor the thickness of a pipe over a long period of time without damaging the transducer at a high temperature of 150 °C. This technology was able to measure the thickness of the pipe stably without errors for a long period of approximately 3300 h (140 days), and it was possible to measure the thickness of the transducer at high temperature without deformation or damage. Ultrasonic signal processing was conducted to obtain precise thickness measurements.

For the demonstration test of the pipe thickness reduction by FAC, evidential facilities were manufactured; the temperature, flow rate, and pressure conditions were fixed; and the pH was adjusted. The pH is one of the important factors in which the FAC and the reduction rate vary depending on the acceleration and inhibition conditions with the internal fluid. After fixing the temperature of the fluid inside the pipe at 150 °C, the pH was adjusted to a range of 7–9.5 and the thickness was measured in real time using the system, indicating that the reduction in thickness was well suppressed as the pH increased. To ensure the reliability of the measurement, the pipes were measured directly using a microscope after all experiments were completed. The error in the Waveguide system and the microscope measurements had a high confidence of less than 1%. It is believed that this technology will be useful for a thickness assessment in the nuclear industry, as well as in industries using high-temperature pipes and facilities to monitor the degradation according to the fluid conditions within the piping.

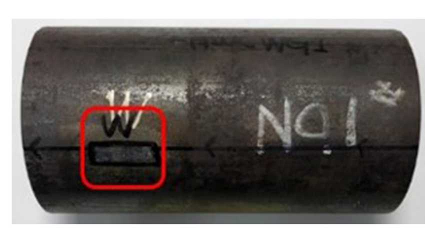

Fig. 7. Cutting pipe at the point where the waveguide system is installed.

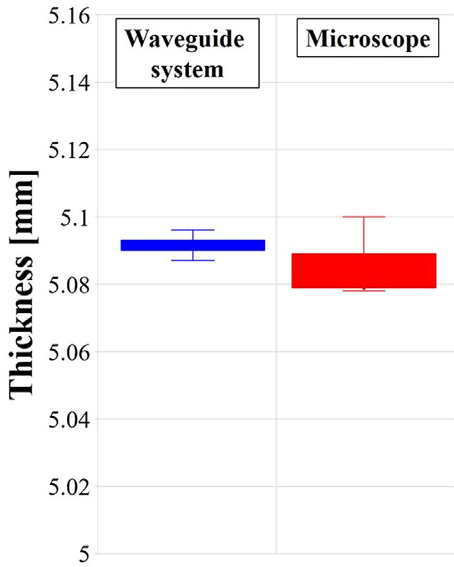

Fig. 8. Average thickness measurement results and distribution.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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