

# TESTING SPOOLABLE REINFORCED FLEXIBLE PIPES AND LINER MATERIAL FOR HIGH-PRESSURE HYDROGEN TRANSPORT

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## SUMMARY

This paper describes the high-pressure hydrogen permeation testing of a spoolable reinforced thermoplastic pipe system and the testing of the chemical resistance to hydrogen of the piping materials. Two different approaches were used to determine the permeation rate: 1) combining single layer permeation measurements, and 2) permeation measurements on the entire system.

## KEYWORDS

*HDPE, permeation, chemical resistance, hydrogen, H<sub>2</sub>, high-pressure testing, spoolable reinforced thermoplastic pipeline systems, RTP.*

## ABSTRACT

*This paper describes two different test methods used to determine the permeation rate of hydrogen through a reinforced thermoplastic pipe (RTP) system with a HDPE liner pipe and a gas-tight layer at a pressure of 42 bar(g) hydrogen and ambient temperature. One method involved combining the permeation resistance of each individual layer, while the other involved determining the permeation rate of the entire pipe (with an outside diameter of 150 mm), including the inline couplers and end fittings. The two permeation rates obtained for the pipe correspond very well, and are extremely small in comparison to the permeation of hydrogen through a solid wall PE pipe.*

*This paper also describes the chemical resistance check, which confirms that the piping materials are suitable for the intended application. In this test, the individual pipe materials were immersed in a 42 bar(g) hydrogen environment for 95 days, after which no significant change in mechanical properties, weight or volume were observed.*

*The results of the aforementioned tests have been combined with existing certifications, which has led to a covenant between the certification agency and producer. Based on this covenant, the actual application of RTP as the backbone of a hydrogen gas network in the northern Netherlands will be established.*

## INTRODUCTION

### Transport of hydrogen gas

The use of hydrogen gas is widely seen as a key component of the current energy transition. Hydrogen can be used as a renewable energy feedstock for industrial processes or as a substitute for fossil fuels. The increasing demand for hydrogen gas results in new opportunities, including the transport and distribution of this gas.

The use of polymers rather than steel pipes is often favored, due to the lower costs of construction and maintenance. Mature products for the high-pressure transport of gases and fluids include spoolable reinforced thermoplastic pipes (RTP, also known as Thermoplastic Composite Pipes (TCP) or Flexible Composite Pipes).

Using RTP pipes for the transport of hydrogen gas requires reconsideration and determination of the properties of the product when exposed to hydrogen, including the permeation resistance of the product and the chemical resistance of the piping materials.

## THEORETICAL BACKGROUND

### Permeation through a homogenous pipe

The movement of a permeate through a material is a process driven by a difference in concentration of the permeate. Permeation is often described by a combination of Fick's law of diffusion and Henry's law, which correlates the concentration of dissolved gas inside a material to the partial pressure. The transport of a gas through a homogeneous polymer pipe is dependent on [1] [2] [3]:

- The difference in the concentration of the permeate between two phases. At equal temperature, this correlates to the difference in partial pressure of the gas on the inside and outside of the pipe.
- The rate at which the permeate moves through the material, known as the permeation coefficient.
- The dimensions of the object separating both phases, in this case a cylindrical pipe.

The permeation of a gas through a homogenous pipe can be derived via [4] [5]:

$$P_C = \frac{Q_P \times \ln\left(\frac{r_L}{r_L - e_L}\right)}{2 \times \pi \times L \times \Delta P} \approx \frac{Q_P \times e_L}{(2 * r_L - e_L) \times \pi \times L \times \Delta P} \quad (1)$$

Where:

- $P_C$  is the permeation coefficient in [(ml\*mm)/(m<sup>2</sup>\*bar\*day)]
- $Q_P$  is the flow of permeated gas in [ml/day]
- $r_L$  is the outer radius of the pipe in [mm]
- $e_L$  is the wall thickness of the pipe in [mm]

- L is the length of the pipe in [m]
- $\Delta P$  is the difference in partial pressure in [bara]

### Permeation through a multilayer pipe

The studied spoolable reinforced thermoplastic pipe is a multilayer pipe produced by SoluForce and coded as M570 H2T (see Figure 1). From the inside out, the pipe consists of the following elements:

- 1) Fluid-tight, corrosion-resistant liner
- 2) Aluminium gas-tight layer
- 3) Synthetic fiber reinforcement for strength
- 4) Ambient resistant white high-density polyethylene cover to protect against UV, abrasion and solar heating



Figure 1: The SoluForce H2T

The permeation process in a multilayer system needs to be considered and equation (1) needs to be modified accordingly.

The movement of the permeate ( $Q_P$ ) depends on the driving force ( $\Delta P$ ) and the resistance of the pipe ( $R_P$ ). This can be expressed as [4]:

$$\Delta P = Q_p \times R_p \quad (2)$$

The dependence of the flow of an entity on both a driving force and a resistance is well-known in several scientific fields, for example:

- Electricity: where the electric current ( $I$ ) is dependent on the difference in voltage ( $\Delta V$ ) and the resistance ( $R$ ) via:

$$\Delta V = I \times R \quad (3)$$

- Thermodynamics: where the heat flow ( $Q$ ) is dependent on the difference in temperature ( $\Delta T$ ) and the thermal resistance ( $R_\theta$ ) via:

$$\Delta T = Q \times R_\theta \quad (4)$$

These fields have combined the influence of multiple resistances on the overall flow through a series circuit. This is also applicable to permeation, as shown by the equations below. A similar approach can be applied to parallel circuits.

Electricity:  $\Delta V = I \times (R_1 + R_2) \quad (5)$

Heat:  $\Delta T = Q \times (R_{\theta,1} + R_{\theta,2}) \quad (6)$

Permeation:  $\Delta P = Q_P \times (R_{P,1} + R_{P,2}) \quad (7)$

The movement of a gas through a multilayer pipe with  $n$  layers can be derived by combining equation (1) and (7) via:

$$Q_P = \Delta P \times \left( \sum_{i=1}^n R_{P,i} \right) \approx \Delta P \times \left( \sum_{i=1}^n \frac{e_L}{(2 * r_L - e_L) * \pi * L * P_{C,i}} \right) \quad (8)$$

By combining the resistance of each individual layer, one can thus determine the permeation rate of the entire multilayer pipe. The resistance of each individual layer can be estimated by consulting literature or determined by performing permeation experiments on each individual layer. However, it is also possible to perform a permeation experiment on the entire multilayer pipe. Both the combined permeation resistance of each individual layer and the permeation of the entire system were used to determine the permeation rate for the RTP.

#### Permeation through accessories

The RTP system tested includes not only the multilayer pipe but also the accessories used to make inline connections and end fittings to connect the system to various facilities. Due to the irregular shape of these accessories, it is not possible to accurately determine their permeation rate using only the permeation resistance of each individual material. As such, the complete inline coupling and end fittings were used in the permeation experiments.

#### Chemical resistance

Literature sources such as [6] and [7] indicate the chemical resistance of the piping materials to hydrogen, with no change in material properties being expected. To provide additional assurance, the change in material properties due to the influence of hydrogen gas was determined. The performed exposure test served only as a chemical resistance check.

#### High-pressure permeation

The permeation coefficient is corrected for the partial pressure. One would therefore expect that the permeation coefficient would be independent of the pressure. However, pressures much higher than standard conditions do not automatically obey Henry's law of proportionality [1] [8] [9]. This includes this case, in which the RTP system is intended to transport hydrogen at a pressure of 42 bar(g). This high pressure can affect the material properties and influence the permeability of the layers via two conflicting processes [10]:

1. The high pressure compresses the polymer and reduces the segmental motion. This reduces the diffusion rate of the gas through the polymer material and thus reduces the permeation rate.
2. The gas enters the polymer and enlarges the matrix, thus increasing the free volume. This results in an increase in segmental motion and therefore an increase in diffusion rate, and therefore an increase in permeation rate.

It is not known which process is dominant. The experiments were performed at the designed operating pressure of 42 bar(g) to exclude possible influences from pressure.

## EXPERIMENTAL

### Permeation of the individual layers

Part of the permeation behavior of the RTP materials was measured in an earlier project and is known. The unknown material was also tested. A disk-shaped specimen with a diameter of 60 mm and a thickness of 1 mm was placed inside a metal container. The disk was pressurized in the container on one side (primary side) with hydrogen at 42 bar(g). The other side (secondary side) was filled with air at atmospheric pressure. To prevent bending of the disk, the disk was supported in the middle by a labyrinth (see Figure 2) with an open surface area of 11.95 cm<sup>2</sup>. Because the volume on the secondary side of the disk was kept constant, the pressure on the secondary side increased due to permeation. This increase in pressure was measured with a pressure sensor. At a certain point, the pressure was relieved via a safety valve, after which the pressure increased again.

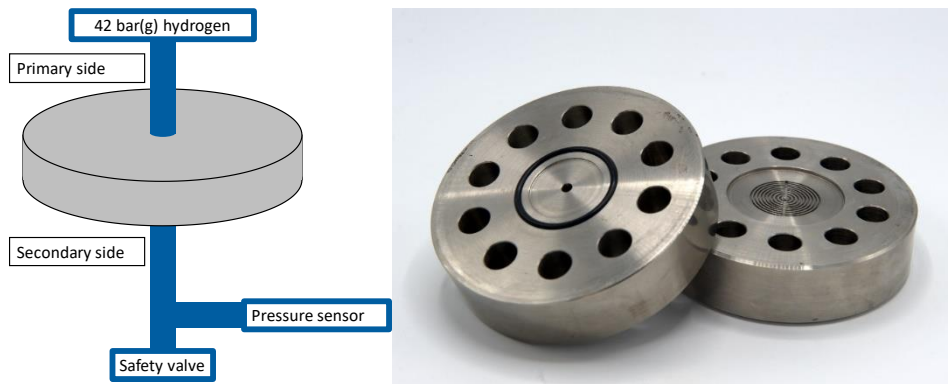


Figure 2. Test setup for the permeation of the disks. Left: a schematic diagram of the method, showing 42 bar(g) hydrogen on top (primary side), with the pressure difference due to permeation being measured on the bottom (secondary side). Right: one opened container with labyrinth to support the disk (right), and a hole to pressurize the disk (left).

The measurement was performed five times. The volume at atmospheric pressure and ambient temperature was calculated from the pressure by means of separate calibration measurements. By considering the thickness of the disk ( $e_A$ ), the open surface area of the labyrinth ( $A_M$ ), the partial pressure difference ( $\Delta P$ ) and the flow of permeated gas ( $Q_{P,A}$ ), the permeation coefficient ( $P_{C,A}$ ) can be calculated as follows:

$$P_{C,A} = \frac{Q_{P,A} \times e_A}{A_M \times \Delta P} \quad (13)$$

This test setup has similarities to the test setups described in [1] and [3].

### Permeation of the multilayer pipe and the accessories

The permeation rate of three different components of the RTP piping system was measured for:

1. The spoolable pipe
2. The inline coupling, where an inline connection between two pipes is made
3. The end fitting, i.e. the end connection between the RTP pipe and the facility, for example

Each component was tested in duplicate.

Jacket pipes were installed around each component (see Figure 3) and flushed with 99.999% pure nitrogen and left at a small overpressure. The piping systems were subsequently flushed with pure hydrogen and pressurized to 42 bar(g).

Because the internal volume of the jacket pipes and the external volume of the components was known, the annular volume was also known. The concentration of hydrogen in the annular volume was measured at specific times using a mass spectrometer. A short piece of spoolable pipe was present in the jacket pipe of both the inline coupling and the end fitting. The permeation rate of the component was corrected for this. This means that the given permeation rates are for the inline coupling and end fitting only.

After an initial phase without any permeation (the time lag), accumulation inside the metal jacket pipe started after some days. After somewhat more time, a stationary permeation phase was reached in which a linear increase in concentration occurred over time. The permeation rate (in [ml] hydrogen per [day] at 42 [bar(g)]) was calculated from the slope of this part of the curve. The given volume was applicable at ambient temperature and atmospheric pressure. This permeation rate was applicable for one end fitting or inline coupling. The permeation rate for spoolable pipes was corrected for the length of the pipes (in [ml] hydrogen per [day] per [meter] at 42 [bar(g)]).



Figure 3. Overview of the test setup for two piping systems (A and B). Jacket pipes 1 and 4 contain the end fittings. Jacket pipes 2 and 5 contain the inline couplers. Jacket pipes 3 and 6 contain the pipe itself.

### Chemical compatibility

Dog-bone-shaped specimens of the liner material and gas-tight layer and two batches of synthetic fibers were placed inside two similar compartments. Both compartments were flushed and filled with pure hydrogen gas and pressurized to 42 bar(g).

After 2,300 hours (95 days), the samples were removed from the compartments. The samples were removed in three phases to ensure that the samples were tested within six hours of removal from the 42 bar(g) hydrogen environment.

The test specimens of the liner material and gas-tight layer were weighed, the dimensions were measured, and the tensile properties were determined. The synthetic fibers were tested using a tensile test. The material properties were compared to the properties of non-exposed reference specimens.

## RESULTS

### Permeation of the individual layers

The accumulated hydrogen volume (calculated from the pressure) in the secondary side of the test setup of a single RTP material specimen is given in Figure 4. For measurement 1, a few datapoints are clear outliers, because a high volume (pressure) was measured, after which the volume (pressure) dropped again. These high data values have been omitted. No straight slope could be obtained for measurement 2. This entire measurement has therefore been omitted. For measurement 5, the datapoints up to the point at which the pressure dropped below 40 bar(g) on the primary side have been used.

The data in Figure 4 is corrected for the actual partial pressure during the measurement, thickness of the test specimen, and surface area. The square of the Pearson product moment correlation coefficient (RSQ) of all slopes in Figure 4 (permeation coefficient) is favorably high, between 0.999 and 1.000 (very close to the maximum 1, indicating a perfect correlation).

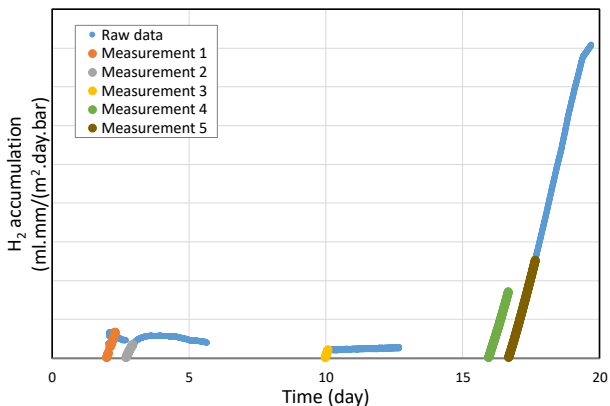


Figure 4. The accumulated hydrogen in the test setup over time, corrected for the partial pressure, thickness of the disk and surface area of the disk. The non-blue lines are datapoints used for the calculation of the permeation coefficient.

The RTP pipes are delivered on rolls of 400 m in length. Using the permeation coefficients (partly obtained from literature) and dimensions of the individual piping layers, and equation (8), one can derive the permeated volume of hydrogen through 400 m RTP at 42 bar(g) during a period of one year. This is equal to 9.9 liters of hydrogen at ambient pressure and temperature.

Permeation through the multilayer pipe and accessories

The accumulated hydrogen volume in the metal jacket pipes is presented in Figure 5. Figure 6 shows the same graph but with a different accumulation scale.

The end fitting in test setup B had much less accumulation in the jacket pipe than the end fitting in test setup A. The accumulation in both the inline coupling and the spoolable pipe was very similar in both test setups. A visual inspection was performed after disassembly. No differences between the two test setups, nor the end fittings, that could contribute to the observed difference could be found.

The steady state appears to be reached after 40 days. The permeation rate (slope in Figure 5 and Figure 6) and the square of the Pearson product moment correlation coefficient (RSQ) of the slope is given in Table 1. These values are valid at ambient temperature and 42 bar(g) internal pressure. The RSQ of all slopes is favorably high (very close to the maximum 1, indicating a perfect correlation).

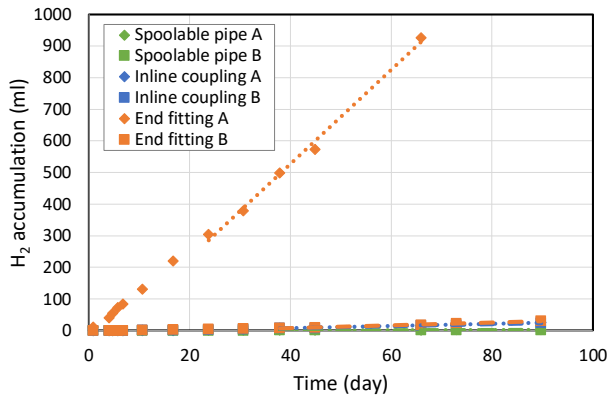


Figure 5. The accumulated hydrogen in the metal jacket pipes over time. The dashed lines are a linear regression over the five last measured points.

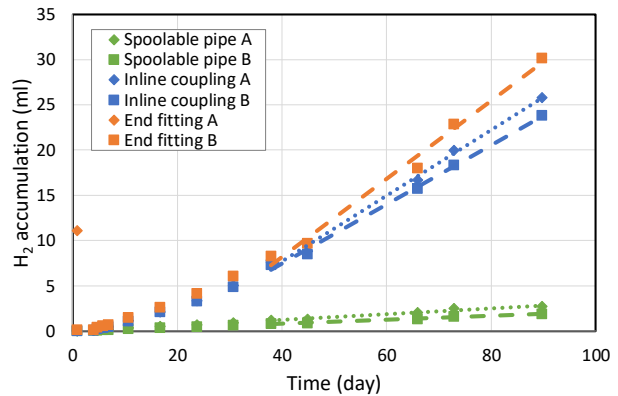


Figure 6. As Figure 5, but with adjusted vertical axis. The accumulated hydrogen in the metal jacket pipes over time. The dashed lines are a linear regression over the five last measured points.

Table 1. The permeation coefficient and the RSQ of the different components (see Figure 5 and Figure 6).

Component	Permeation rate (ml/day)	RSQ of the slope
Spoolable pipe A	0.0525*	0.960
Spoolable pipe B	0.0357*	0.983
Inline coupling A	0.363	0.995
Inline coupling B	0.326	0.997
End fitting A	14.9	0.994
End fitting B	0.432	0.989

\* These values have been corrected for the length of the measured pipe (which was 610 mm), so the unit is in fact ml/(m·day).



Using the permeation rate for spoolable pipe A, one can derive the permeated volume of hydrogen through 400 m RTP at 42 bar(g) during a period of one year. This is equal to 7.7 liters at ambient pressure and temperature.

This value, derived using a completely different method of measuring, is very similar to the value found by combining the permeation resistance of the individual layers (9.9 liters of hydrogen per year over 400 meters).

### Chemical compatibility

Figure 7 shows the ultimate strength of the liner material and gas-tight layer, and the breaking force of the gas-tight layer and both batches of the synthetic fibers relative to their respective reference measurements (reference is 100%). No statistically significant difference, based on the 95% two-tailed T-test, can be observed between the reference measurements and the measurements after exposure. This also applies to the other measured material properties, including the E-modulus, strain at break, yield stress, dimensional measurements and weight measurements (Data for these tests is not shown in this paper).

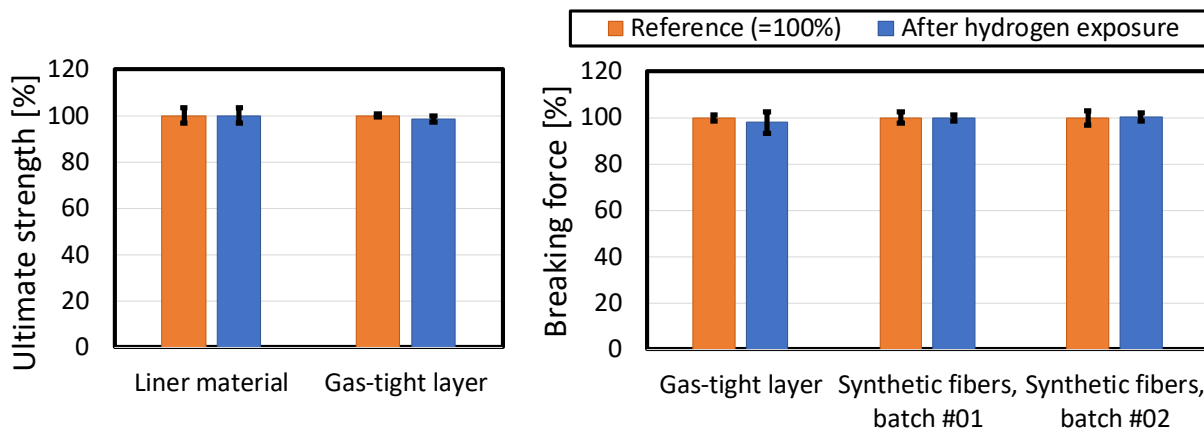


Figure 7. Histogram with the ultimate strength of the liner material and gas-tight layer specimens and breaking force of the gas-tight layer and the synthetic fiber specimens relative to their respective reference measurements. The standard deviation is indicated as a black bar.

### PERMEATION COMPARISON

By using the highest values in Table 1, the permeation volume of hydrogen of various designs of the RTP system can be calculated. As an example:

One kilometer of the piping system contains two inline connectors and two end fittings. Over a period of one year, 30.3 liters of hydrogen will permeate over the entire system:  $(0.0525 \cdot 1000 + 0.363 \cdot 2 + 14.9 \cdot 2) \cdot 365 / 1000 = 30.3$  liters.

The permeation of hydrogen through a PE 100-RC pipe was measured in an earlier study [11]. Using this permeation coefficient of 126.8 (ml·mm)/(m<sup>2</sup>·day·bar) and a ø110 mm SDR 11 pipe at 2 bar(g), 1.745 liters of hydrogen would permeate through a pipe of 400 meters annually. This is about 175 times more than an RTP at 42 bar(g).

## CONCLUSION

### Hydrogen permeation

The permeation of three components of the RTP system was measured in duplicate. The highest measured permeation rate is as follows:

- Spoolable pipe: 0.0525 ml/(m·day)
- Inline coupling: 0.363 ml/day
- End fitting: 14.9 ml/day

The value for the spoolable pipe corresponds very closely to the theoretical values when calculating the permeation rate using the permeation coefficients of each individual layer.

Permeation in this high-pressure resistant gas-tight system at 42 bar(g) will be 175 times less than in a normal polyethylene piping system at 2 bar(g).

### Chemical compatibility

Specimens of the liner pipe, the gas-tight layer and the synthetic reinforcement fibers were exposed to a 42 bar(g) hydrogen environment for 2,300 hours (95 days). The material properties were determined after exposure and compared to the material properties of reference specimens. No statistically significant difference was observed in respect of the specimen weight, dimensions or mechanical properties between the exposed and non-exposed specimens.

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