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STUDY ON PARAMETERS AFFECTING DURABILITY OF POLYETHYLENE PIPES FOR HOT WATER APPLICATION

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SHORT SUMMARY
There are mainly two grades of polyethylene pipes for hot water supply and heating. The excellent molecular structure polyethylene resins having resistance to stress crack growth are used because hot water is flowed for a long time in both polyethylene pipes. Furthermore, various additives are mixed to prevent the thermal degradation of polyethylene pipes at the elevated temperature. In this study, the stress rupture test and the continuous hot water circulation test were conducted using extruded polyethylene pipes mixed two kinds of polysilane and a nucleating agent. As a test result, the time to failure of polyethylene pipes mixed two kinds of polysilane was longer than that of a polyethylene pipe without additives. The time to failure of polyethylene pipes mixed a nucleating agent was almost similar to that of a polyethylene pipe without additives. According to precise observation at the failure part, it was found that there was a small degraded colored layer at the inner surface of polyethylene pipes mixed two kinds of polysilane compared with polyethylene pipes without additives. In addition, it was found that the generation and the growth of degraded colored layer and small cracks was delayed due to diffusion of polysilane to the inner surface and to protection of adhesion of metal oxides such as copper oxide and iron oxide

KEYWORDS
polyethylene pipe, additives, polysilane, hot water circulation test, degradation, actual operating condition

ABSTRACT
Hot water distribution pipes connecting a gas cogeneration system with the water heater and terminal appliances are expected to high durability at present. There are mainly two lines for the hot water distribution system. A single pipe is used for water and hot water supply, and a double pipe for circulation of hot water is used for such as central heating, floor heating, and bath water heating.
The hot water circulation test with partial replacement of fresh water which is reflected to the actual operating condition is a useful test method. The hot water circulation test

using metal ion containing aqueous solution for central heating is also suitable as the accelerated evaluation test for polyethylene pipes. The pressurized hot water circulation test using chlorine aqueous solution for water and hot water supply is the accelerated evaluation test. It was clarified that parameters such as temperature, pressure, and concentration of aqueous solution of copper ion and/or chlorine ion affected durability of polyethylene pipes for hot water application
Various additives such as two kinds of polysilane and a nucleating agent as well as antioxidants were mixed to prevent the thermal degradation of polyethylene pipes at the elevated temperature. As a test result, the time to failure of polyethylene pipes mixed two kind of polysilane was longer than that of a polyethylene pipe without additives. The time to failure of polyethylene pipes mixed a nucleating agent was almost similar to that of a polyethylene pipe without additives.

INTRODUCTION

The polymer electrolyte fuel cells (PEFC) and the Solid Oxide Fuel Cell (SOFC) residential cogeneration systems have been commercialized in Japan. The number of commercialized both cogeneration systems have reached 200, 000 in total in 2016. The expected number of PEFC and SOFC is 1,400,000 in 2020. The waste heat can be used for hot water supply and central heating. Hot water distribution pipes connecting a gas cogeneration system with the water heater and terminal appliances are very important. As the advanced residential cogeneration system is expensive, the longer lifetime of the heating and hot water supply system is required for consumers, the reliable plastic pipes have been developed. Polyethylene of raised temperature resistance (PERT) as non-crosslinked polyethylene has been used as pipe materials for hot water supply and space heating in Japan as well as conventional cross-linked polyethylene (PEX). A resin was generally compounded with antioxidants to protect thermal degradation of a resin for extrusion and long-term use. However, in case of contact to hot water, it might not be able to keep the expected lifetime of pipes because of antioxidants being eluded and flowing out into hot water from the inner surface of a pipe. Various new additives such as two kinds of polysilane and a nucleating agent as well as conventional antioxidants were mixed to prevent the thermal degradation of polyethylene pipes at the elevated temperature. In general, the lifetime of a polyethylene pipe has been evaluated by the hydrostatic stress rupture test which is standardized in ISO 1167 and JIS K6769 (1), (2). The hot water circulation test was also conducted considering the actual operating condition.

EXPERIMENTAL

Material preparation

As shown in Figure 1, the low molecular weight polysilane, grade OGSOL SI-10-40 having methylphenyl group whose number-average molecular weight (Mn) is 610 and weight-average molecular weight (Mw) is 680 and the high molecular weight polysilane, grade OGSOL SI-10-10 whose Mn is 2100 and Mw is 12,700 produced by Osaka Gas Chemicals Co. and the nucleating agent, grade CN-001 produced by RIKEN VITAMIN Co. were prepared for additives. The pipe grade resin HE121 produced by Japan Polyethylene Corp. was extruded with above additives at 210 °C for 10mm nominal diameter pipes.

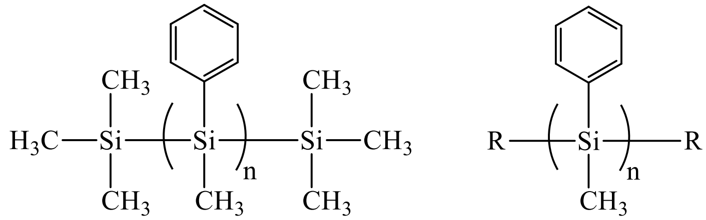


Figure 1 Molecular structure of polysilane (13)
(R: terminal group)

Hydrostatic stress rupture test

The pipes was used for 10 mm in internal diameter, 13 mm in outer diameter, and 800 mm in length, and the joints at the both ends of the pipe were made of plastic (PPS) joints. The polyethylene pipes filled with water are firstly put in the oven at the temperature of 110 °C, measuring the failure time that it takes to crack growth through the pipe thickness. The hydrostatic stress rupture test is conducted at 0.25 MPa and at 110 °C (3)

Hot water circulation test

Figure 2 shows the illustration of the device used in hot water circulation test as described in reference (11), (12). The straight and 6D bent pipes specified by ISO 10508 were used for 10 mm in inner diameter, 13 mm in outer diameter, and 2,000 mm in length, and the joints at both ends of the pipe were made of plastics. The hot water at 90, 100, and 110 °C heated by the water heater was constantly circulated. The internal pressure was 0.25 MPa which is the maximum operating pressure for heating. The circulation speed of the hot water was 2 l/min. The evaluation was continued while checking the degradation condition by taking out the samples at regular intervals. Also, keeping the constant heating circuit, the circulation water was replaced for 10 l/week in order to sustain the concentration of the dissolved oxygen in the pipe. In this study, the hot water circulation test which is reflected to the actual operating condition was conducted by using a polyethylene pipe for hot water and the correlations and differences between the hydrostatic stress rupture test and the hot water circulation test were examined.(4)～(12)

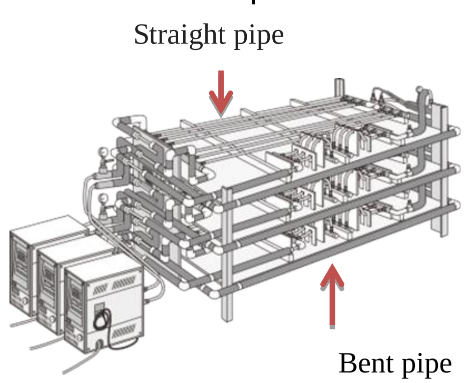


Figure 2: Hot water circulation test. (11), (12)

TEST RESULT AND DISCUSSION

Hydrostatic stress rupture test

As described in reference (13), the time to failure of the pipes with polysilane was longer than that of a pipe without polysilane after the hydrostatic stress rupture test at 0.25 MPa and at 110 °C. As comparison of low molecular weight 1.0 wt% and high molecular weight 0.3 wt% polysilane, the high molecular weight 0.3 wt% polysilane was more effective. The time to failure of a pipe with a nucleating agent was only slightly longer than that of a pipe without a nucleating agent.

Hot water circulation test

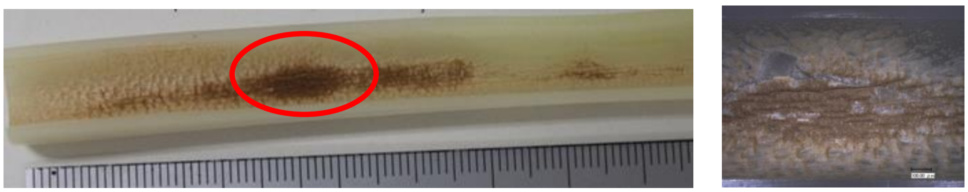
Table 1 shows the latest hot water circulation test results at 0.25 MPa and at 90, 100, and 110 °C. It was found that the time to failure of the pipes with polysilane was quite longer than that of a pipe without polysilane. As comparison of low molecular weight 1.0 wt% and high molecular weight 0.3 wt% polysilane, the high molecular weight 0.3 wt% polysilane was more effective as the same tendency of the hydrostatic stress rupture test results was obtained. The failure data were obtained for all samples at 100 °C and 110 °C and almost samples at 90 °C. The failure data on the pipes with polysilane of the high molecular weight 0.3 wt% was not still obtained at 90 °C after the testing time has reached 25,251 h.
As comparison of a straight pipe and a bent pipe, the time to failure of a bent pipe was shorter than that of a straight pipe due to the combined stress condition of hoop stress and bending stress. Although the time to failure of a pipe with polysilane of high molecular weight 0.3 wt% was longer than that of a pipe with polysilane of low molecular weight 1.0 wt% at 110 °C, there was no remarkable difference between two polysilane grades at 100 °C and 90 °C.
As comparison of results between the hydrostatic stress rupture test and the hot water circulation test, all times to failure of straight pipe after the hot water circulation test were shorter than those after the hydrostatic stress rupture test at 0.25 MPa and at 110 °C considering the actual operating condition.(13)

Table 1 Hot water circulation test results

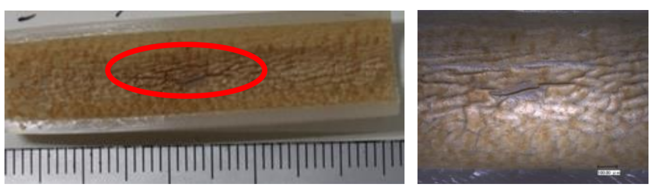
	Time to failure (h) Straight pipe			Time to failure (h) Bent pipe		
	90 °C	100 °C	110 °C	90 °C	100 °C	110 °C
HE121	18,172	8,930	3,267	17,779	5,987	3,200
HE121 with SI-10-40 1.0 wt%	21,000	15,776	5,313	21,000	11,099	4,806
HE121 with SI-10-10 0.3 wt%	25,251 or more	15,442	5,695	25,251 or more	11,445	5,335
HE121 with CN-001 2.0 wt%	18,824	8,949	3,680	18,643	6,350	3,487

Observation with optical microscope

Figure 3 shows the inner surface and crack generation at a failure part of pipes after hot water circulation test at 110 °C. There were a widely large colored and degraded layer and many longitudinal cracks at the inner surface of only HE121 at 3,267 h and HE121with CN-001 2.0 wt% at 3,680 h after the hot water circulation test. The large crack early passed through a pipe thickness due to the hoop stress. On the other hand, there was no crack generation and no colered layer at the inner surface of both HE121 with SI-10-40 1.0 wt% and HE121 with SI-10-10 0.3 wt% at 4,000 h after hot water circulation test at 110 °C as described in reference (13). There were two locally colored layers at the inner surface of HE121 with SI-10-10 0.3 wt% at 5,695 h after the hot water circulation test. It also took a relative long time that a crack passed through a pipe thickness.



HE121 (Time to failure: 3,267h)



HE121with CN-001 2.0 wt% (Time to failure: 3,680h)

Figure 3 Observation of inner surface and crack generation at failure part of pipes after hot water circulation test at 0.25 MPa and at 110°C (13)

According to precise observation at the failure part, it was found that there was a small degraded colored layer at the inner surface of polyethylene pipes mixed two kinds of polysilane compared with polyethylene pipes without additives. In addition, it was found that the generation and the growth of degraded colored layer and small cracks was delayed due to diffusion of polysilane to the inner surface and to protection of adhesion of metal oxides such as copper oxide and iron oxide at the inner surface.(13)

Water repellency

With addition of polysilane, the water repellency on surface of pipes was also improved. Although the water contact angle of only HE121 was 87 degrees, that of HE121 with SI-10-40 0.3 wt% was 91 degrees and that of HE121 with SI-10-40 1.0 wt% was 94 degrees because polysilane is hydrophobic.

Oxidation resistance

A resin was generally compounded with antioxidants to protect thermal degradation of a resin for extrusion and long-term use. However, in case of contact to hot water, it might not be able to keep the expected lifetime of pipes because of antioxidants being eluded and flowing out into hot water from the inner surface of a pipe. Various new additives such as two kinds of polysilane and nucleating agent as well as conventional antioxidants were mixed to prevent the thermal degradation of polyethylene pipes at the elevated temperature. Although there is on function of radical trapping on molecular structure of polysilane like conventional antioxidants, polysilane easily diffused to the inner surface and protected oxygen penetration and adhesion of metal oxides such as copper oxide and iron oxide at the inner surface. Oxidation induction time of HE121 with SI-10-40 1.0 wt% was 73.1 minutes although that of only HE121 was 61.2 minutes at 200 °C in air.(14)～(17)

Lifetime comparison
The lifetime of pipes of HE121 with SI-10-40 1.0 wt% was approximately 1.7 times compared with that of only HE121 at 90 °C, 100 °C and 110 °C at 0.25 MPa in the hot water circulation test.

CONCLUSIONS
The following contents are concluded from experimental tests.
- The hot water circulation test replacing water for 10 l / week was a useful test method considering the actual operating condition and elution of antioxidants in a pipe into the hot water.
- The time to failure of polyethylene pipes mixed two kinds of polysilane was longer than that of a polyethylene pipe without additives.
- The lifetime of pipes of HE121 with SI-10-40 1.0 wt% was approximately 1.7 times compared with that of only HE121 at 90 °C, 100 °C and 110 °C at 0.25 MPa in the hot water circulation test.
- With addition of polysilane, the water repellency on surface of pipes and oxidation induction time were improved.

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