Performance Evaluation of Improved EPDM Material under High-temperature Environments

1. Introduction

In the case of a severe accident (hereafter, SA), the primary containment vessel (PCV) is surrounded by high-temperature steam at 200°C and high radiation dose of several hundred kGy. Therefore, the PCV requires a sealing material that can maintain sealing performance even under an SA environment. For such cases, we evaluated the performance of our improved EPDM material (No. H3070) under high-temperature environments.

Previously, silicone rubber and fluororubber, which generally have excellent heat resistance, have been used as sealing materials for PCV. However, in an environment with high-temperature steam of 200°C and high radiation dose of several hundred kGy, the structural weaknesses of polymers cause the progression of reduction in molecular weight due to hydrolysis and due to exposure to high energies including γ rays. This is likely to lead to the loss of sealing performance, and so a new material was considered necessary.[1)]

The improved EPDM material was developed on the assumption that it would be used in high-temperature steam, and we have proposed the new material for such applications. We have been conducting various evaluations to confirm its sealing performance, using a steam environment at a high temperature of 200°C and atmospheric pressure of 0.854 MPa as basic conditions which properly allow a PCV to contain radioactive material in the event of SA. Various tests were conducted under these conditions and the improved EPDM material was confirmed to have adequate sealing performance.[2)]

Figure1 schematically illustrates the characteristics of the improved EPDM material and existing silicone rubber and fluororubber sealing materials, based on previous evaluation results of each material. The horizontal axis represents steam temperature and the vertical axis radiation dose. The figure shows that silicone rubber and fluororubber cannot maintain sealing performance in the event of SA; on the other hand, the new EPDM material maintains sealing performance even in high-temperature steam of over 200°C and γ ray irradiation of over 800 kGy.

Thus, the improved EPDM material was confirmed to offer adequate environmental performance in the event of SA, and was determined to be used as a new sealing material for PCV. For this application, it was crucial to confirm the following two matters: confirmation of the limit conditions under which the improved EPDM material can maintain sealing performance, and verification of the improved EPDM material' s superiority under SA environments. Therefore, an evaluation under a high-temperature

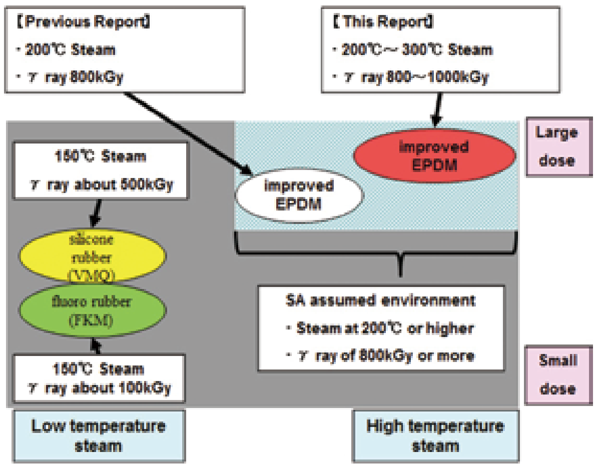

Figure1　General characteristics of sealing material

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environment in which the temperature significantly exceeded SA temperatures was conducted.

Regarding the evaluation method, the following tests were conducted to confirm sealing performance: compression-set tests, which are effective for verifying the lifespan of the seal, and leakage tests using small flanges, which imitate flanges installed on actual equipment.

2. Evaluation

2-1) Preliminary test to set the evaluation temperature

This evaluation aimed to confirm the limitation of sealing performance of the improved EPDM material; the evaluation was conducted at temperatures up to the temperature at which the improved EPDM material showed no significant material changes. Thermogravimetric analysis was used as the evaluation method. In Figure2, the purple line represents the rate of weight loss due to heat. Weight loss begins to occur at a temperature of over 300°C. No major change was found to develop in the main backbone structure at temperatures up to 300°C. Therefore, the performance was evaluated at temperatures up to 300°C.

2-2) Evaluation method

2-2-1) Compression-set test

Two evaluation methods were used to confirm the performance at 300°C, one of which was the compression-set test, which is very effective for confirming the lifespan of a seal.

The method of measurement conformed to JIS K 6262:2013 "Rubber, vulcanized or thermoplastic – Determination of compression-set at ambient, elevated or low temperatures." The JIS large specimen (cylindrical specimen with a diameter of ϕ29 mm and thickness of 12.5 mm) was used as the sample shape; the compression rate was 25%.

Plastic deformation which occurs at high temperatures and under compressed conditions (but not only high-temperature conditions) was quantified. The quantification was defined as follows: 0%: the original shape is completely restored; 100%: no shape restoration occurs. Rubber deterioration can be considered to be a chemical reaction. Therefore, the time taken to reach a given deformation can be estimated by plotting a graph with time on the horizontal axis and compression-set rate on the vertical axis as shown in Figure3.

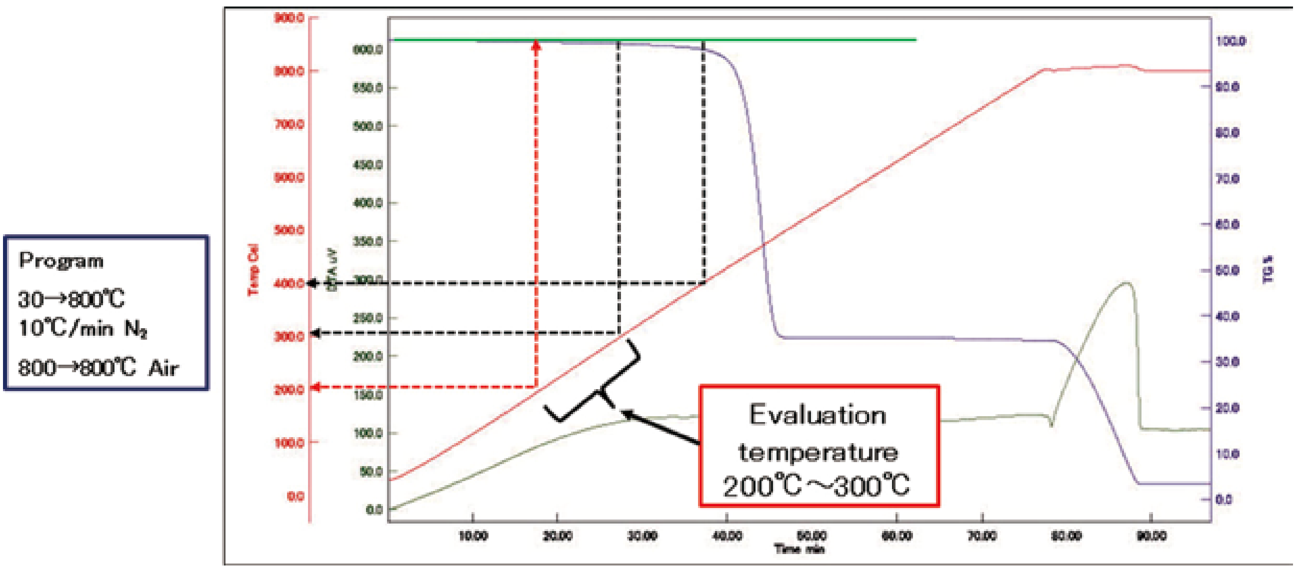

Figure2　Results of thermogravimetric analyses

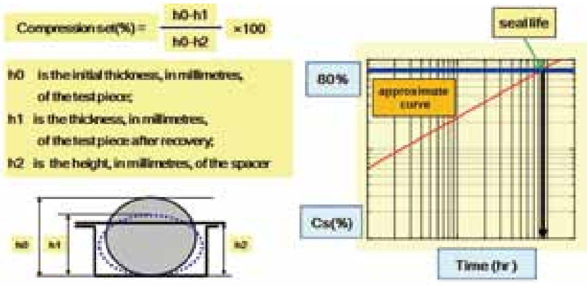

Figure3　Compression-set test and expected lifespan

Table1 shows the evaluation conditions. To take the conditions in the event of SA into consideration, the evaluation was conducted under the following conditions: atmosphere: saturated steam environment, exposure dose of γ rays: 800 to 1000 kGy, temperature: 200 to 300°C. Regarding the environment within a PCV in the event of SA, the maximum atmospheric pressure was assumed to be 0.854 MPa. On the other hand, this evaluation was conducted under a saturated steam environment, resulting in much greater pressure applied to the specimens (saturated steam pressure at 200°C: 1.6 MPa, at 300°C: 8.6 MPa). Therefore, the environment is much more severe than in a PCV in the event of SA.

2-2-2) Leakage tests using small flanges

Leakage tests using small flanges were conducted as the second evaluation method. Small flanges, which imitate the flanges installed on actual equipment, were used to confirm whether the sealing performance could be maintained at the temperatures and pressures of SA. Figure4 schematically illustrates the small flange and Table2 shows their sizes. The cross-sectional shape and size of the flange's groove are equivalent to those of the actual equipment; the cross-sectional shape and size of gaskets were also equivalent to those of the actual equipment. In the event of SA, the flanges of the PCV open due to inner pressure. To imitate this phenomenon, an adjusting shim was inserted into the smooth part in the small flange, and the opening area was then adjusted. After the adjustment, He gas was used to apply pressure to the inside of the small flange. This pressurization enabled leakage tests to be conducted under conditions imitating those in the actual equipment.

As Table3 shows, the test conditions were as follows:

Table1　Conditions for compression-set tests

Test	Radiation dose	Environ	Temp	Time
No.	kGy	-	℃	hr
1-1	800	Steam	200	24
1-2	800	Steam	200	72
1-3	800	Steam	200	168
2-1	800	Steam	225	24
2-2	800	Steam	225	72
2-3	800	Steam	225	168
3-1	800	Steam	250	24
3-2	800	Steam	250	72
3-3	800	Steam	250	168
4-1	1000	Steam	265	72
4-2	1000	Steam	265	168
4-3	1000	Steam	265	336
5-1	1000	Steam	280	168
6-1	1000	Steam	300	24
6-2	1000	Steam	300	72
6-3	1000	Steam	300	168

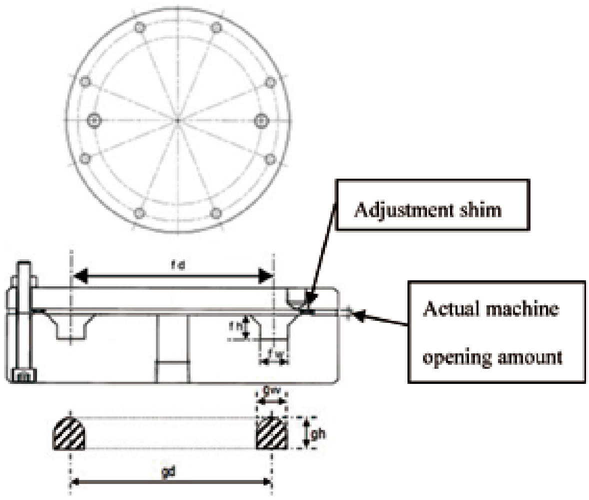

Figure4　Outline of a small flange

Table2　Sizes of small flanges

	Seal material dimensions			Groove dimensions			Opening amount
	Center diameter (gd)	Width (gw)	Height (gh)	Center diameter (fd)	Width (fw)	Height (fh)	
Small flange	Shrink actual machine	Same dimension as actual machine	Same dimension as actual machine	Shrink actual machine	Same dimension as actual machine	Same dimension as actual machine	Actual machine opening amount

Table3　Conditions for leakage tests

Test No.	Radiation dose	Heating				
		Exposure environment			Test environment	
		Environ	Temp	Time	Environ	Temp
	kGy	-	℃	hr	-	℃
7-1	1000	Steam	280	168	Dry	280
8-1	1000	Steam	300	168	Dry	300

After the flange temperature reached a given temperature, He atmosphere of 1 MPa was used to pressurize the inside of the flange for 10 minutes. After the pressurization, the presence of leakage was confirmed. If He gas leaks in this test, the leakage can be confirmed from the outside of the flange. Therefore, the leaked He gas was collected through water as a substitute to calculate the hourly leakage rate of He. Figure 5 outlines the test equipment.

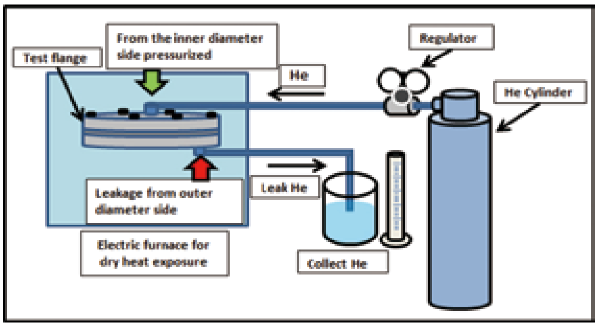

Figure5 Outline of test equipment to determine leakage from a small flange

3. Evaluation results

3-1) Compression-set test

Table4 shows the results of the compression-set tests. In the table, Cs represents the compression-set rate. The following trends were confirmed: under a constant temperature, the compression-set rate increases over time; within a given time period, the compression-set rate increases with increasing temperature.

Table 4 Results of compression-set tests

Test No.	Radiation dose kGy	Environ -	Temp ℃	Time hr	Cs %
1-1	800	Steam	200	24	10
1-2	800	Steam	200	72	11
1-3	800	Steam	200	168	16
2-1	800	Steam	225	24	10
2-2	800	Steam	225	72	15
2-3	800	Steam	225	168	18
3-1	800	Steam	250	24	13
3-2	800	Steam	250	72	16
3-3	800	Steam	250	168	22
4-1	1000	Steam	265	72	25
4-2	1000	Steam	265	168	34
4-3	1000	Steam	265	336	38
5-1	1000	Steam	280	168	53
6-1	1000	Steam	300	24	47
6-2	1000	Steam	300	72	67
6-3	1000	Steam	300	168	84

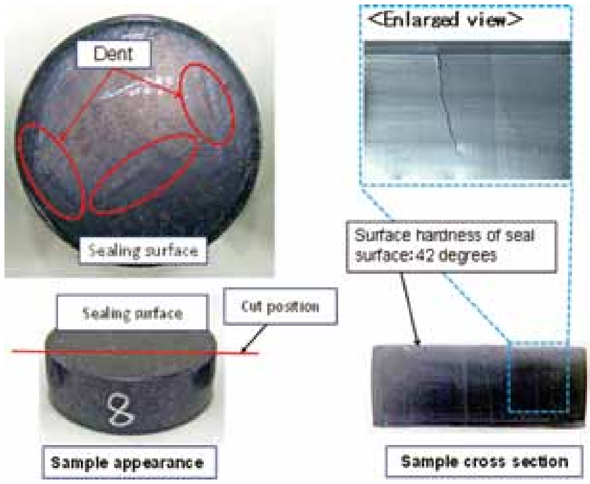

Figure6 Specimen after Test No. 6-3 test

Figure6 shows an external view of a specimen tested under the most severe conditions with a temperature of 300°C and duration of 168 hours (Test No. 6-3). Although no abnormality was found on the side face, dents were found around the center of the sealing surface. Therefore, the specimen was cut and the cross section was checked. Although the dents were caused by a crack that had developed inside, the crack was confirmed not to have penetrated into the sealing surface. The cause of the crack appeared to be a rapid expansion of compressed steam while the pressure was being reduced under highly-pressurized saturated steam. To be more specific, the crack is considered to have developed as follows: 1) Steam pressure of 8.6 MPa was applied to flanges for the evaluation under a saturated steam environment at 300°C. 2) The steam pressure also compressed steam contained in the rubber. 3) After the evaluation was completed, the highly-pressurized steam was released from the pressure container. 4) During steam release, rapid decompression occurred. 5) The rapid decompression caused steam, which had been compressed in the rubber, to expand rapidly. 6) The rapid expansion caused the crack in the rubber.

In this evaluation, the crack was caused by a physical factor stemming from the evaluation equipment. On the other hand, in the event of SA, the maximum pressure is expected to be 0.854 MPa, and so it is considered that no crack will develop in actual

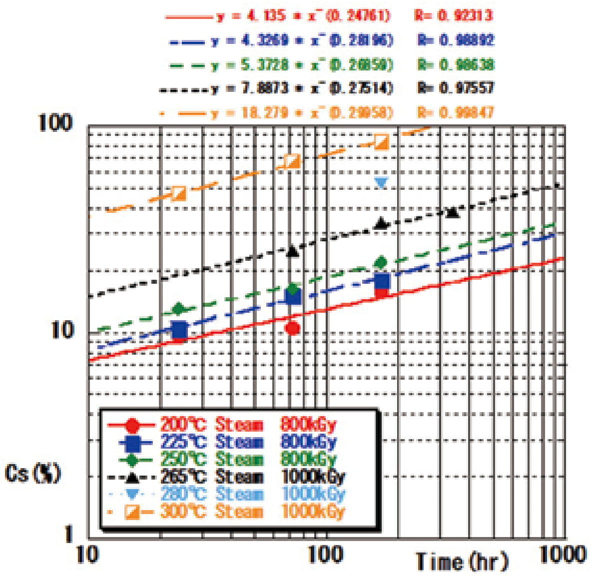

Figure7 A plot of the results of compression-set tests

equipment in the event of SA.

Figure7 illustrates the results of the compression-set tests. The horizontal axis represents time; the vertical axis represents compression-set rate. Under such conditions, the resulting data were plotted to produce an approximation curve for each temperature. As Figure7 shows, each curve shows a very high correlation coefficient, and the slopes of the respective approximation curves are approximately the same. Figure8 is an Arrhenius plot of the results of the Arrhenius equation under the following conditions: 1) Sealing performance can be secured at the

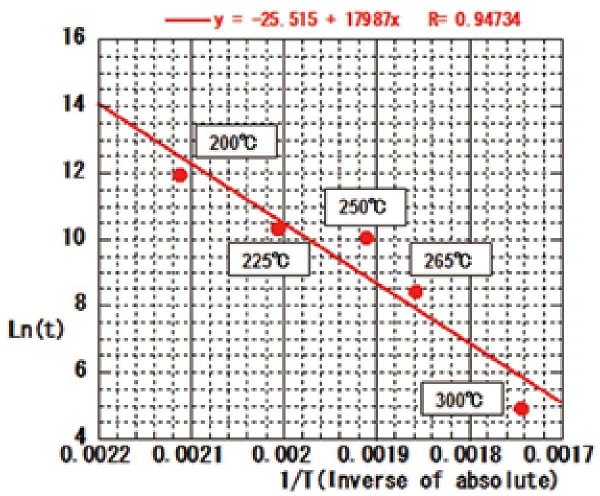

Figure8 Arrhenius plot based on data at temperatures of 200 to 300°C

compression-set rate of 80%.[3)] 2) Time duration was set such that a compression-set rate of 80% could be achieved within the time duration. 3) Each temperature was arranged according to the time duration. As a result, the plot demonstrates that a collinear approximation can be made with very high correlation coefficients.

Since thermal deterioration is a chemical reaction, the progress of deterioration varies depending on the amount of thermal energy which is applied to the material. Therefore, regular deterioration behaviors are likely to be detected provided that the temperature range does not significantly exceed the thermal limit for the material and the evaluation environment is not unstable.

Therefore, in this evaluation, which was conducted under a saturated steam environment of 200 to 300°C, the deterioration had developed according to the chemical-reaction theory, and was not likely to cause sudden and abnormal deterioration. Therefore, lifetime estimation was confirmed to be possible with this deterioration.

3-2) Leakage test using small flanges

The results from leakage tests using small flanges showed that no leakage occurred from the specimen which had been exposed to the saturated steam environment at 280°C and 300°C for 168 hours.

After the evaluation had been completed, the small flanges were dismantled to check the appearance of the gaskets. Figure9 shows a photograph of a specimen which was exposed to 280°C for 168 hours; no abnormality was found in the appearance. Similarly, regarding a specimen which was exposed to 300°C for 168 hours, cracks were confirmed on part of the gasket in the direction of the outer circumference; however, no cracks or damages, which affect leakage, were confirmed in the direction of inner or outer circumference. Regarding the abovementioned cracks, like the internal cracks in the compression-set tests, rapid expansion of compressed steam due to rapid decompression appears to have caused the cracks.

Therefore, at 168 hours after the initiation of exposure

Figure9 Specimen after Test No. 7-1 leakage tests

under saturated steam environments at 280°C and 300°C, no significant defect of sealing material was confirmed. Therefore, the sealing performance appears to have been maintained.

4. Summary

The results of the compression-set tests and leakage tests using small flanges under high-temperature saturated steam environments were as follows.

- The mechanism of deterioration of our improved EPDM material (No. H3070) appeared to be the same at temperatures ranging between 200 and 300°C; the possibility of sudden abnormal deterioration developing due to temperature changes is low.
- The results of the leakage tests using small flanges showed no leakage when the flanges had been exposed to saturated steam at 300°C for 168 hours.
- These results indicate that the improved EPDM material can be used in a stable manner under saturated steam environments at temperatures up to 300°C.

5. Conclusion

In this evaluation, the improved EPDM material was confirmed to maintain its sealing performance even under the very severe environment of 300°C, provided the environment was limited to that used in this evaluation. This finding highlighted the importance of continuing with development and verification based on unconventional, free-thinking without bias, as well as listening to users' requests. We will continue to strive hard to produce such reports in the future.

Note that this report is an edited version of a paper presented at the autumn academic lecture session of the Atomic Energy Society of Japan in 2017.

Lastly, we sincerely thank the staff of Chubu Electric Power Co., Inc. for their assistance in publishing this report.

6. References

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