OUT OF PILE TEST WITH SIC CLADDING SIMULATING LOCA CONDITIONS

KENICHIRO FURUMOTO, SEIICHI WATANABE, DAIKI SATO

Mitsubishi Nuclear Fuel 622-1, Funaishikawa, Tokai, Ibaraki 319-1197 – Japan

MICHIAKI OKAMOTO, MASAAKI YAMATO

Mitsubishi Heavy Industries 1-1-1, Wadasaki-cho, Hyogo-Ku, Kobe, Hyogo 652-8585 – Japan

SHINICHIRO YAMASHITA, TOKIO FUKAHORI

Japan Atomic Energy Agency 2-4 Shirakata, Tokai, Ibaraki 319-1195 – Japan

ABSTRACT

The tests simulating LOCA conditions were performed using test apparatus for silicon carbide (SiC) cladding sample tubes. As results of the tests, SiC showed higher stability under LOCA conditions compared with conventional Zr-based alloy. This can be an advantage of SiC cladding as well as higher stability under SA conditions.

1. Introduction

SiC fibre reinforced SiC matrix composite (SiC/SiC) is one of the promising material for the accident tolerant fuel (ATF) cladding [1][2]. It is well known that SiC/SiC cladding (SiC cladding) can suppress or mitigate core damage in case of severe accident (SA) such as fuel assemblies melting, hydrogen ignition and steam explosion, thanks to its low oxidation rate and hydrogen generation under elevated temperature [3][4]. On the other hand, performance of SiC cladding under loss of coolant accident (LOCA) condition is not fully known, although fuel rod must be subjected to LOCA conditions before SA. Several previous studies [5][6][7] have reported thermal shock test results that SiC/SiC could keep its shape after water quench from high temperature without significant damage. Taking into account together with low oxidation rate under high temperature steam environment, better resistance against LOCA event can be expected than conventional zirconium (Zr)-based alloy. Such LOCA resistant feature of SiC cladding can enhance safety of LWRs.

In this study, in order to investigate performance of SiC cladding under LOCA conditions, out-of-pile tests were carried out simulating typical LOCA conditions in PWR, such as elevation of temperature and rod internal pressure, loss of coolant, high temperature steam exposure, and thermal shock due to quenching by refilled water supplied from the emergency core cooling system (ECCS).

2. LOCA event in case of conventional Zr-based alloy cladding

A typical behaviour during LOCA event is illustrated in fig 1. Fig 1 shows changes of the cladding temperature with time during LOCA event. At first, the drawdown of coolant level and decrease of system pressure occur. Heat removal by the coolant is lost, but the heat continues to generate due to decay of radioactive fuel and fission products. The cladding temperature begins to increase, and the internal pressure of the fuel rod also increases. As the internal

pressure becomes larger, cladding tube of the fuel rod balloons due to creep deformation. If significant ballooning of many cladding tubes occurs at similar axial position, the blockage of coolant flow can occur. A part of fuel rods can burst, coupled with decrease of mechanical strength of Zr-based alloy in elevated temperature. And radioactive materials can be released from inside to outside of the fuel rod. Subsequently, fuel rods are exposed to steam in high temperature, and their cladding tubes are oxidised in such condition. In particular, burst cladding tubes are significantly embrittled because both of inner and outer surfaces can be oxidised. And then, when coolant water refilled up to the elevation of fuel rods, cladding tubes are quenched. Oxidised and embrittled cladding tubes can fail due to thermal shock during quenching. In order to keep the long-term "coolable geometry" of reactor core after LOCA, most of cladding tubes must be intact after quench. As Japanese current criteria for ECCS in the case of the fuel rod with Zr-based alloy cladding, peak temperature must be lower than 1200°C and equivalent cladding reacted (ECR) must be less than 15% during LOCA to prevent extensive failure of fuel rods.

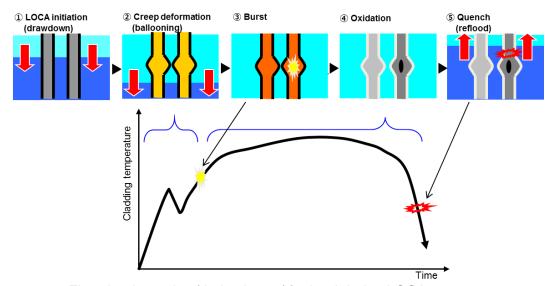


Fig 1 A schematic of behaviour of fuel rod during LOCA

3. Test apparatus

In order to clarify the difference of the performance during LOCA from Zr-based alloy cladding, integral behaviour of SiC cladding has to be investigated by the test simulating the sequence of conditions of LOCA described in fig 1. For this purpose, the LOCA-simulating test apparatus for SiC cladding was fabricated [7]. The basic constitution is the same as the apparatus which has been used for the commercial Zr-based alloy cladding of Mitsubishi PWR fuel rods. Some components of the apparatus were optimized for SiC/SiC. The visual appearance of the test apparatus is shown in fig 2. The sample tube is held inside of an atmosphere-released glass tube. The sample tube is connected to a pressurised gas line, and pressurised by argon gas. The apparatus is equipped with an infrared electric furnace. The sample tube is heated up to 1600°C by the electric furnace. During sample heating, upper and lower gripping positions of the sample tube were cooled by coolant flow to maintain the stability of the bonding between the sample tube and devices. The cooling of gripping position can slightly affect temperature and pressure of inside of the sample tube. A pair of optical windows are equipped to the furnace. Outer diameter of the sample tube can be measured by the optical dimension measurement device through the optical windows. Temperature is monitored by thermocouples attached to the sample tube. A steam line and a water line are connected to inside of the glass tube. Steam during heating up and water to quench the

sample tube can be introduced. The whole test apparatus which comprises above equipment and measurement devices is fixed by the universal test system.

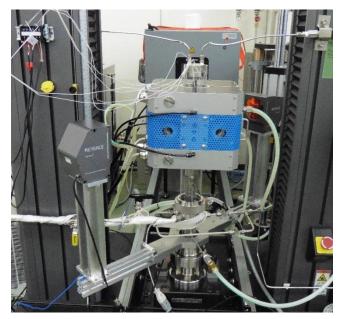


Fig 2 The image of LOCA simulating test apparatus

4. Test conditions and procedures

4.1 Test samples

Test samples were chemical vapour infiltrated (CVI) SiC/SiC tubes. SiC/SiC sample tubes consist of dense monolithic SiC layer manufactured by chemical vapour deposition (CVD) process on the surface to improve gas-tightness. The outer diameter, the thickness, and the length of sample tubes were approximately 10 mm, 1 mm, and 300 mm, respectively.

4.2 Procedures of temperature elevation test

Test conditions of temperature elevation tests and LOCA integral tests are summarized in tab 1. SiC/SiC sample tubes were pressurised with argon gas to 3.5 and 7 MPa. Alumina ceramic was inserted into the sample tubes to reduce the inside free volume. The outer diameter of sample tubes was measured by the optical dimension measurement device at RT before the temperature elevation. Sample tubes were heated in air by the electric furnace at the temperature raising rate of 5°C/second up to 1600°C. After the temperature reached to 1600°C, sample tubes were cooled down by air to RT. Inner pressure and temperature of sample tubes were measured through the test to monitor that burst or leak occurred or not. After cooling down, the outer diameter of sample tubes was measured with optical device to observe creep deformation due to inner pressure. The visual appearance of sample tubes after test was observed.

4.3 Procedures of LOCA integral test

SiC/SiC sample tubes were set to the test apparatus as for the temperature elevation tests described above. In addition, Zr-based Alloy sample tube was tested by using the same test apparatus and procedures for the reference. Sample tubes were pressurised to 1 MPa with argon gas. Sample tubes were heated at a temperature raising rate of 5°C/second in steam flow environment until reaching the predetermined maximum temperature, and then hold for the predetermined duration at that temperature. Tests were performed in various conditions of the predetermined maximum temperature and holding time of (1200°C, 1000 seconds),

(1400°C, 3000 seconds), and (1600°C, 100 seconds), to investigate effect of oxidation. After the duration hold in high temperature was over, the electric furnace was turned-off, and then the sample tubes were immediately cooled down by water flooding from the bottom of the device. The LOCA integral tests were performed without axial loading during water quench. The tests with the axial loading will be performed to investigate interactions between a SiC cladding and a grid spacer during LOCA which are different from conventional fuel design studied previously [8]. And whether sample tubes failed or not due to quench was checked. Water to quench sample tubes was at RT. The visual appearance of sample tubes after tests was observed.

Samples	Environment	Initial internal	Maximum	Holding time	Quench
		pressure	temperature	at max. temp.	
Temperature elevation test					
SiC/SiC tube	Air	3.5 MPa	1600°C	-	-
SiC/SiC tube	Air	7 MPa	1600°C	-	-
LOCA integral test					
SiC/SiC tube	Steam flow	1 MPa	1200° C	1000 seconds	$1200^{\circ}\text{C} \rightarrow \text{RT}$
SiC/SiC tube	Steam flow	1 MPa	1400° C	3000 seconds	1400°C → RT
SiC/SiC tube	Steam flow	1 MPa	1600° C	100 seconds	1600°C → RT
Zr-based Alloy	Steam flow	1 MPa	1200° C	1000 seconds	~700°C → RT

Tab 1 Test conditions of temperature elevation test and LOCA integral test

5. Results and discussion

5.1 Temperature elevation test

Fig 3 shows the temperature and the pressure history for each temperature elevation tests of initial inner pressure of 3.5 and 7 MPa. As shown in fig 3, no burst fracture and no significant pressure loss were observed in both tests up to 1600°C. Hermeticity of the sample tubes at RT did not changed between before and after the test. The inner pressure changed with the change of the sample tube temperature. The degree of the change of inner pressure was slight compared to the temperature change because on this test apparatus, heated volume was only a part of entire volume consisting the inside of the sample tubes and connecting piping. Fig 4 shows the visual appearances before and after the test of initial internal pressure of 7 MPa. A coloration was observed for each sample tubes after the tests. Obvious creep deformation was not observed such as ballooning due to internal pressure. Similar results were obtained in the test carried out with initial internal pressure of 3.5 MPa. The outer diameter measurement showed no significant change before and after the test.

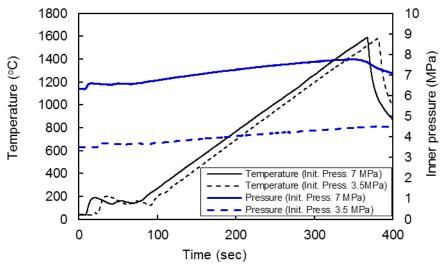


Fig 3 Temperature and inner pressure of SiC/SiC sample tubes during temperature elevation tests

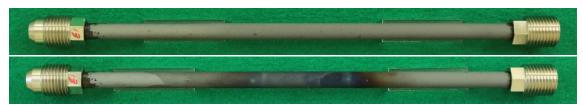


Fig 4 Visual appearance of the SiC/SiC sample tube before (upper) and after (lower) temperature elevation test. (Initial internal pressure of the tube: 7 MPa)

5.2 LOCA integral test

As an example of results of LOCA integral tests, fig 5 shows the history of the temperature and the inner pressure of the SiC/SiC sample tube during LOCA integral test performed under the condition of the maximum temperature of 1600°C and holding time at the maximum temperature of 100 seconds. And fig 6 shows the visual appearances of the sample tube before and after the test. As shown in fig 5 and fig 6, no burst and no ballooning were observed in LOCA integral test as in the temperature elevation tests. And sample tubes after water quench were intact under all of the conditions of (1200°C, 1000 seconds), (1400°C, 3000 seconds), and (1600°C, 100 seconds). Although no fractures were observed, gas-tightness was not maintained after quench. Decrease of the inner pressure and formation of some air bubbles at the surface on the sample tube were observed due to internal gas release from cracks caused by thermal quench during the water filling. Air bubbles were more obvious for sample tubes with higher quenching temperature. LOCA integral test was performed with Zr-based alloy sample tube for comparison. Fig 7 shows the history of the temperature and the inner pressure of the reference Zr-based alloy sample tube. Zr-based alloy sample tube burst during temperature rising, before reaching 1200°C. And Zr-based alloy sample tube failed during quench after holding for 1000sec at the maximum temperature of 1200°C, which is approximately 48%ECR by Backer-Just equation [9]. Zr-based alloy sample tube after the test is shown in fig 8. Significant ballooning and burst due to inner pressure, extensive oxidation and failure were observed as shown in fig 8. The test results are shown in fig 9. Horizontal and vertical axis respectively indicate the maximum temperature and the holding time at the maximum temperature. The current Japanese ECCS criteria on LOCA for Zr-based alloy cladding was together shown in fig 9. The current Japanese ECCS

criteria on LOCA is less than 1200°C of peak cladding temperature (PCT), and less than 15%ECR in fig 8. As clearly shown in fig 9, SiC/SiC sample tubes did not fail even far beyond the conditions corresponding to the current criteria. It can be concluded that SiC cladding presents better capability of maintaining "coolable geometry" than conventional Zr-based alloy cladding during and after high temperature and thermal shock expected in LOCA event. In order to investigate oxidation of SiC surface, scanning electron microscope (SEM) observation were conducted on cross-section near the surface of SiC/SiC sample tubes after high temperature steam exposure in equivalent conditions as LOCA integral tests. Fig 10 shows SEM image of the cross-section of the SiC/SiC sample tube after 1600°C steam exposure for 100 seconds. As shown in the SEM image in fig 10, the formation of oxide layer was not observed. The formation of oxide was similarly not observed on the surface of SiC/SiC sample tubes after steam exposure of equivalent oxidation conditions of the other LOCA integral tests, which are steam exposure at 1200°C for 1000 seconds, and at 1400°C for 3000 seconds. According to the oxidation kinetics of SiC which has been studied and reported in [10], in oxidation conditions of LOCA integral tests carried out in this study, it is estimated that growth of the oxide (SiO₂) due to oxidation of SiC and volatilization by further reaction of SiO₂ should be extremely small. Therefore, it is reasonable that there are no observable changes due to oxidation reaction on the surface of SiC/SiC sample tubes.

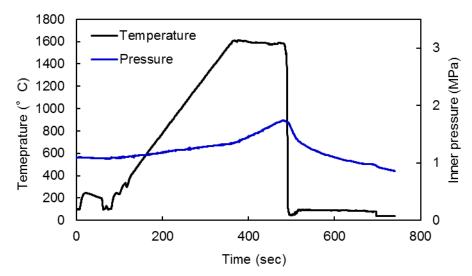


Fig 5 Temperature and inner pressure of the SiC/SiC sample tube during the LOCA integral test.

(The sample tube was hold for 100 seconds at 1600°C in steam, and then quenched)



Fig 6 Visual appearance of the SiC/SiC sample tube before (upper) and after (lower) of the LOCA integral test.

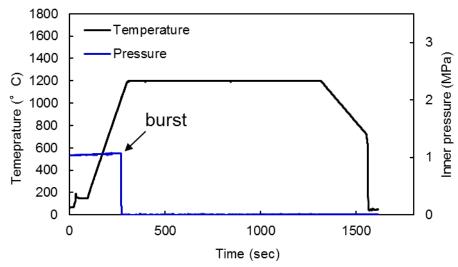


Fig 7 Temperature and inner pressure of the Zr-based alloy sample tube during the LOCA integral test.

(The sample tube was hold for 1000 seconds at 1200°C in steam, and then quenched)



Fig 8 Visual appearance of the Zr-based alloy sample tube after the LOCA integral test.

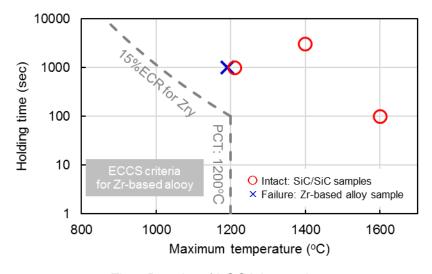


Fig 9 Results of LOCA integral tests

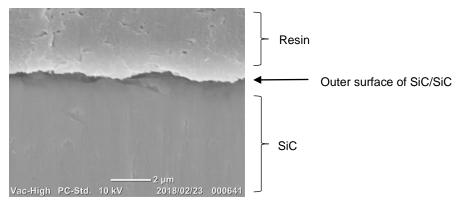


Fig 10 SEM image of the cross-section near the outer surface of the SiC/SiC sample tube after 1600°C steam exposure for 100 seconds

6. Conclusions

Out of pile tests with SiC/SiC sample tubes simulating LOCA were performed by using developed test apparatus to investigate their behaviour during LOCA events. In temperature elevation tests, burst had not been shown on both of SiC/SiC sample tubes pressurised to 3.5 and 7 MPa of 1600°C. During LOCA events, the absence of the cladding burst can significantly reduce the release of radioactive materials. It would be effective to decrease radiation exposure. Furthermore, almost no outer diameter change had been observed on SiC/SiC sample tubes during tests. There should be no concerns of the blockage of coolant water flow by ballooning of cladding due to creep deformation. In LOCA integral tests, although the loss of hermeticity after quench was observed, SiC/SiC sample tubes maintained the shape without significant deformation and damage such as ballooning and burst as Zr-based alloy cladding even in the conditions far beyond the current ECCS criteria in Japan which are 1200°C PCT and 15% ECR. It is suggested that SiC cladding has a better long-term coolability than conventional Zr-based alloy cladding. Performances of SiC cladding under LOCA revealed in this study could be great advantages for the safety evaluation on LOCA and beyond LOCA. Further expansion of the data of the behaviour of SiC/SiC under LOCA condition, modeling for safety analysis will be essential in order to exhibit gaining the margin for each evaluations.

7. Acknowledgement

Results presented in this paper contains a part of the outcomes of the "Development of Technical Basis for Introducing Advanced Fuels Contributing to Safety Improvement of Current Light Water Reactors" which is carried out under the project on Development of Technical Basis for Improving Nuclear Safety by METI of Japan in JFY2017.

8. References

- [1] S. Yamashita, F. Nagase, M. Kurata, T. Nozawa, S. Watanabe, K. Kirimura, K. Kakiuchi, T. Kondo, K. Sakamoto, K. Kusagaya, S. Ukai, A. Kimura, "TECHNICAL BASIS OF ACCIDENT TOLERANT FUEL UPDATED UNDER A JAPANESE R&D PROJECT", Proceedings of 2017 Water Reactor Fuel Performance Meeting, Jeju, Korea, September 10-14 (2017)
- [2] Shannon M. Bragg-Sitton and W. Jon Carmack, "Phased Development of Accident Tolerant Fuel", Proceedings of TopFuel2016, Boise, Idaho, September 11-15 (2016)
- [3] Zeses E. Karoutas, Raymond Schneider, N. Reed LaBarge, Javier Romero, Peng Xu, "WESTINGHOUSE ACCIDENT TOLERANT FUEL PLANT BENEFITS", Proceedings of 2017 Water Reactor Fuel Performance Meeting, Jeju, Korea, September 10-14 (2017)

- [4] Y. Yamakoshi, K. Kirimura, H. Kuramoto, T. Noda, S. Yamashita, T. Fukahori, "Severe Accident Evaluations for Conventional PWR Power Plant with SiC Composite Fuel Cladding", Proceedings of TopFuel2018, Prague, Czech Republic, September 30-October 4 (2018)
- [5] Carlos F. Bacalski, George M. Jacobsen, Christian P. Deck, "Characterization of SiC-SiC Accident Tolerant Fuel Cladding after Stress Application", Proceedings of TopFuel2016, Boise, Idaho, September 11-15 (2016)
- [6] Sang Pill Lee, Jin Kyung Lee, In Soo Son, Dong Su Bae, Akira Kohyama, "Thermal shock properties of 2D-SiC_f/SiC composites", Fusion Engineering and Design, vol. 87, 2012, 1244–1248
- [7] K. Furumoto, S. Watanabe, T. Yamamoto, H. Teshima, S. Yamashita, H. Saito, N. Shirasu, "The applicability of SiC-SiC fuel cladding to conventional PWR power plant", Proceedings of 2017 Water Reactor Fuel Performance Meeting, Jeju, Korea, September 10-14 (2017)
- [8] T. Honma, S. Doi, M. Ozawa, S. Urata, T. Sato, "Thermal Shock Behavior of PWR High-Burnup Fuel Cladding Under Simulated LOCA Conditions", Proceedings of ANS Annual Meeting, Milwaukee, Wisconsin, USA, 2001
- [9] Louis Baker. Jr and Luis C. Just, "Studies of Metal-water Reactions at High Temperatures III, Experimental and Theoretical Studies of The Zirconium-Water Reaction", ANL-6548. May 1962
- [10] Kurt A. Terrani, Bruce A. Pint, Chad M. Parish, Chinthaka M. Silva, Lance L. Snead, Yutai Katoh, "Silicon Carbide Oxidation in Steam up to 2 MPa", Journal of the American Ceramic Society, vol. 97, no. 8, 2014, 2331-2352.