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Experimental Study of Embrittlement of Zr-1%Nb VVER Cladding under LOCA-Relevant Conditions

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ABSTRACT

During 2001-2004, research was performed to develop test data on the embrittlement of niobium-bearing (Zr-1%Nb) cladding of the VVER type under loss-of-coolant accident (LOCA) conditions. Procedures were developed and validated to determine the zero ductility threshold. Pre-test and post-test examinations included weight gain and hydrogen content measurements, preparation of metallographic samples, and examination of samples using optical microscopy, scanning electron microscopy and transmission electron microscopy. Sensitivity of the zero ductility threshold to heating and cooling rates was determined. Oxidation kinetics and ductility threshold were measured for the standard E110 alloy, six variants with different impurities, Zircaloy, and irradiated E110. Oxidation temperatures were varied from 800-1200 C, and mechanical (ring compression) testing temperatures were varied from 20-300 C. It was concluded that (a) the current type of E110 cladding has an optimal microstructure, (b) oxidation and ductility of the oxidized cladding are very sensitive to microchemical composition and surface finish, (c) the use of sponge zirconium for fabrication of cladding tubes provides a significant reduction of the oxidation rate and an increase in the zero ductility threshold, and (d) additional improvement in oxidation and ductility can be achieved by polishing the cladding surface.

FOREWORD

A world-wide trend to substantially increase nuclear fuel burnup to higher levels has led fuel manufacturers in the U.S. and France to develop niobium-bearing cladding alloys that are similar in composition to Russian cladding alloys. These alloys, E-110, E-635, ZIRLO, and M5, all have greatly improved corrosion resistance compared with Zircaloy during normal operation, especially at higher burnup levels. However, in early 2001, it was realized that the Russian alloys and the Western niobium-bearing alloys behaved somewhat differently under conditions of a loss-of-coolant accident (LOCA), during which the cladding is exposed to steam at high temperatures.

At that time, a research program was already underway at Argonne National Laboratory in the U.S. to investigate the effects of high-burnup on cladding behavior under LOCA conditions. Further, a cooperative research effort on fuel behavior was also underway at the Russian Research Center (Kurchatov Institute) with partial sponsorship by the French Institute for Radiological Protection and Nuclear Safety and the U.S. Nuclear Regulatory Commission; additional funding was being provided by the Russian fuel vendor, TVEL. It was then decided to investigate the underlying phenomena that governed cladding behavior of niobium-bearing alloys — particularly the Russian cladding — in this Russian program. By closely coordinating this research with the work underway at Argonne National Laboratory on similar Western alloys, it was hoped that a fuller understanding could be obtained. Coordination between laboratories was further enhanced by including some Zircaloy cladding specimens in the Russian program and including some E110 cladding specimens in the program at Argonne National Laboratory.

After several years of research at both laboratories, the general cause of differences in behavior under LOCA conditions has been isolated to the ore reduction process and the surface finish of the cladding tubing. This understanding is helping to improve licensing criteria that can be applied to new and different cladding alloys. The extensive work performed by the Russian Research Center (Kurchatov Institute) and their collaborating laboratory at the State Research Center (Research Institute of Atomic Reactors) is documented in the following report.

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TABLE OF CONTENTS

Page

1. INTRODUCTION 1.1

2. BACKGROUND 2.1

3. OXIDATION BEHAVIOR AND EMBRITTLEMENT THRESHOLD OF STANDARD E110 CLADDING: PROGRAM, TEST PROCEDURES, DISCUSSION OF TEST RESULTS 3.1

3.1. THE PROGRAM CONCEPT AND TECHNICAL REQUIREMENTS TO EXPERIMENTAL WORKS 3.1

3.2. METHODOLOGICAL ASPECTS OF OXIDATION AND MECHANICAL TESTS 3.6

3.2.1. Oxidation tests 3.6

3.2.2. Mechanical tests 3.11

3.3. DISCUSSION OF TEST RESULTS AND WORKING OUT OF PRELIMINARY CONCLUSIONS 3.26

3.3.1. Reference tests 3.26

3.3.2. The comparative analysis of E110 and Zry-4 oxidation and mechanical behavior 3.29

3.3.3. Determination of sensitivity of the E110 cladding embrittlement to the oxidation type and the characterization of comparative behavior of E110 and M5 claddings 3.48

3.3.4. The evaluation of the E110 oxidation and mechanical behavior as a function of oxidation temperature 3.52

3.3.5. The sensitivity of the behavior of Russian niobium-bearing alloys to the alloying composition 3.60

3.3.6. Interrelation between the zero ductility threshold and the temperature of mechanical tests 3.62

3.3.7. The analysis of representativity of the zero ductility threshold determined due to ring compression tests 3.68

3.3.8. Consideration of the zero ductility threshold of the E110 cladding as a function of the irradiation effect 3.75

3.3.9. The analysis of the E110 oxidation kinetics 3.80

4. OXIDATION BEHAVIOR AND EMBRITTLEMENT THRESHOLD OF THE MODIFIED E110 CLADDING: PROGRAM AND DISCUSSION OF TEST RESULTS 4.1

4.1. MAJOR PROVISIONS OF THE TEST PROGRAM WITH A MODIFIED E110 CLADDING 4.1

4.2. THE ANALYSIS OF EXPERIMENTAL RESULTS OBTAINED AT SURFACE EFFECT STUDIES 4.4

4.3. THE ASSESSMENT OF RELATIONSHIP BETWEEN THE MICROCHEMICAL COMPOSITION AND OXIDATION BEHAVIOR OF NIOBIUM-BEARING ALLOYS 4.9

4.3.1. The analysis of the oxidation and mechanical behavior for the $E110_{G(fr)}$ and $E635_{G(fr)}$ claddings fabricated on the basis of 100% French sponge Zr 4.10

4.3.2. The interpretation of test results with E110$_{G(3fr)}$ and E110$_{G(3ru)}$ claddings 4.11
4.3.3. The sensitivity of the oxidation behavior of the sponge E110 cladding to the oxidation temperature 4.13
4.3.4. The analysis of results obtained in the test with the E635 cladding fabricated using sponge Zr 4.17
4.3.5. The comparative consideration of a microchemical composition of different types of the E110 alloy 4.18
4.4. THE COMPARATIVE ANALYSIS OF THE E110 MATERIAL MICROSTRUCTURE 4.24
4.5. THE OXIDATION KINETICS OF THE SPONGE TYPE E110 CLADDING 4.28

5. SUMMARY 5.1

5.1. MAJOR FINDINGS OF THE PROGRAM FIRST PART 5.1
5.1.1. Methodological aspects of mechanical tests 5.1
5.1.2. Methodological aspects of oxidation tests 5.4
5.1.3. The embrittlement behavior of Zr-1%Nb (E110) cladding 5.5
5.1.4. The embrittlement behavior of Zr-1%Nb (E110) cladding as a function of irradiation effects 5.8
5.1.5. The oxidation kinetics and embrittlement behavior of the E110 cladding according to results of previous investigations in different laboratories 5.9
5.2. MAJOR FINDINGS OF THE PROGRAM SECOND PART 5.11
5.2.1. The concept of special investigations 5.11
5.2.2. Surface effect studies 5.11
5.2.3. Bulk chemistry studies 5.12
5.2.4. Bulk microstructure studies 5.16
5.2.5. Final Remarks 5.17

LIST OF FIGURES

Page

Fig. 3.1. Schematics of the cladding sample 3.7

Fig. 3.2. Oxidation test apparatus 3.8

Fig. 3.3. Types of temperature histories for different combinations of heating and cooling rates 3.9

Fig. 3.4. Development of the data base with test results 3.10

Fig. 3.5. Test machine for ring compression tests of oxidized cladding samples 3.11

Fig. 3.6. The as-measured load-displacement diagram of the E110 oxidized cladding sample 3.12

Fig. 3.7. Procedure for the determination of the sample zero-displacement point 3.13

Fig. 3.8. Determination of effective modulus of elasticity basing on the results of scoping compression tests 3.14

Fig. 3.9. The processing procedure for the load-displacement diagram of the ring compression test of the E110 oxidized cladding 3.15

Fig. 3.10. Relative displacement at failure of the E110 cladding after a double-sided oxidation and S/S combination of heating and cooling rates as a function of the ECR 3.15

Fig. 3.11. The validation of the procedure for the zero-ductility threshold determination 3.16

Fig. 3.12. Sensitivity of a relative displacement at failure to the ring sample length for the ductile sample 3.17

Fig. 3.13. Sensitivity of a relative displacement at failure to the ring sample length for the brittle sample 3.17

Fig. 3.14. Typical E110 load-displacement diagrams for different oxidation 3.18

Fig. 3.15. Demonstration of the state of ductile claddings after ring compression tests 3.19

Fig. 3.16. The data base for the interpretation of load-displacement diagrams for oxidized samples with a high ductility margin 3.20

Fig. 3.17. The load-displacement diagram of Zry-4 cladding sample with a partial ductility margin 3.21

Fig. 3.18. The data base to characterize the mechanical behavior of the cladding sample with a partial residual ductility before the fracture 3.21

Fig. 3.19. The data base to characterize the mechanical behavior of cladding sample with the partial residual ductility at failure 3.22

Fig. 3.20. The data base to characterize the mechanical behavior of the brittle ring sample 3.23

Fig. 3.21. Processing of the load-displacement diagram obtained in the ring tensile test of a simple ring sample manufactured from the oxidized E110 cladding tube 3.24

Fig. 3.22. Three-point bending test apparatus ... 3.25

Fig. 3.23. Schematic for the processing of the load-displacement diagram after three-point bending tests ... 3.25

Fig. 3.24. Results of reference tests ... 3.27

Fig. 3.25. The residual ductility of the E110 cladding vs heating and cooling rates ... 3.28

Fig. 3.26. The zero ductility threshold of the E110 cladding vs heating and cooling rates ... 3.28

Fig. 3.27. Summary of the ring compression test results performed in different laboratories with the Zry-4 oxidized cladding ... 3.30

Fig. 3.28. The comparative test data characterizing the ductility of Zry-4 oxidized cladding vs ECR ... 3.31

Fig. 3.29. The comparative data obtained in different laboratories to characterize the Zry-4 oxidation kinetics ... 3.32

Fig. 3.30. The comparison of E110 and Zry-4 cladding behavior in accordance with RRC KI/RIAR test data ... 3.32

Fig. 3.31. Appearance of the E110 and Zry-4 claddings after the oxidation at 11.3–11.8% ECR and F/F, S/S combinations of heating and cooling rates ... 3.33

Fig. 3.32. The appearance in detail of the E110 oxidized surface vs the ECR ... 3.34

Fig. 3.33. Demonstration of two layers of ZrO_2 oxide on the outer surface of the E110 oxidized standard as-received tube using fractography results ... 3.34

Fig. 3.34. Cross section of the oxide nodule in the E110 cladding ... 3.35

Fig. 3.35. The characterization of the oxidized E110 claddings vs ECR ... 3.35

Fig. 3.36. Visualization of ZrO_2 oxide behavior as a function of the ECR after the double-sided oxidation at 1100 C (F/F combination of heating and cooling rates) of E110 standard as-received tubes ... 3.36

Fig. 3.37. The appearance and microstructure of Zr-Nb oxidized cladding after the operation with the surface boiling (RBMK cladding type) ... 3.37

Fig. 3.38. Angular variations of the E110 standard as-received tube microstructure after a double-sided oxidation at 1100 C and 10% ECR (sample #17) ... 3.37

Fig. 3.39. The microstructure of the Zry-4 cladding (sample #64) oxidized at 11.5% ECR at F/F combination of heating and cooling rates ... 3.38

Fig. 3.40. The microstructure of the E110 cladding after a single-sided oxidation at 1100 C ... 3.39

Fig. 3.41. The comparative data characterizing the ZrO_2 and α-Zr(O) thickness ... 3.39

Fig. 3.42. The appearance of microstructure and characterization of oxygen distribution in the E110 cladding after the double-sided oxidation at 1100 C 3.41
Fig. 3.43. The oxygen distribution in the E110 cladding (1100 C, 8.2% ECR) in accordance with results of SEM examinations 3.42
Fig. 3.44. The comparison of microhardness distributions for the brittle and ductile E110 oxidized cladding 3.43
Fig. 3.45. Demonstration of the morphology of ZrO_2 layers in the E110 oxidized cladding (sample #41-4, 1100 C, 8.2% ECR) 3.44
Fig. 3.46. The niobium distribution in the E110 cladding (1100 C, 8.2% ECR) in accordance with results of SEM examinations 3.46
Fig. 3.47. The SEM micrograph of the E110 oxidized cladding 3.47
Fig. 3.48. The optical microstructure of the E110 oxidized sample with hydrides in the prior β-phase 3.48
Fig. 3.49. The comparison of the E110 appearance and microstructure after single-sided and double-sided oxidation at 1100 C 3.49
Fig. 3.50. Comparative data characterizing the E110 residual ductility as a function of the oxidation type 3.49
Fig. 3.51. The data base characterizing the mechanical behavior of the E110 and M5 claddings after a single-sided oxidation at 1100 C 3.50
Fig. 3.52. The comparison of the E110 and M5 cladding mechanical behavior after the single-sided oxidation at 1100 C in accordance with the ring compression test results at 20 C 3.51
Fig. 3.53. The characterization of the E110 commercial cladding after irradiation and oxidation in the α-phase 3.52
Fig. 3.54. The characterization of the allotropic phase transformation in the E110 and M5 alloys 3.53
Fig. 3.55. Appearances of the E110 cladding after the double-sided oxidation at 800–1100 C and F/F combination of heating and cooling rates 3.54
Fig. 3.56. The microstructure of the E110 cladding after a double-sided oxidation at 800–950 C and F/F combination of heating and cooling rates 3.55
Fig. 3.57. The data base characterizing the residual ductility of the E110 cladding as a function of the ECR and oxidation temperature 3.56
Fig. 3.58. The E110 residual ductility and hydrogen concentration as a function of the ECR after a double-sided oxidation at 1100 C and F/F, F/Q combinations of heating and cooling rates 3.57
Fig. 3.59. The hydrogen content in the E110 cladding as a function of the ECR and temperature after a double-sided oxidation 3.57

Fig. 3.60. The comparison of the E110 microstructure after a double-sided oxidation at different temperatures 3.58

Fig. 3.61. The characterization of the E110 cladding behavior after a double-sided oxidation at 1200 C.. 3.59

Fig. 3.62. The summary of results characterizing the E110K behavior under oxidation and ring compression test conditions 3.61

Fig. 3.63. The characterization of the E635 cladding behavior under oxidation and mechanical test conditions 3.63

Fig. 3.64. Load-displacement diagrams for two Zry-2 samples hydrided (specially) up to 180 ppm and 700 ppm, respectively, as a function of the temperature mechanical tests of the bending type (reprinted from [47]) 3.65

Fig. 3.65. Dependence of the E110 cladding ductility on the ECR (oxidation at 1100 C) and temperature of ring tensile tests 3.65

Fig. 3.66. Residual ductility of two E110 samples oxidized at 10 and 11.7% ECR (1100 C) as a function of temperature ring compression tests 3.66

Fig. 3.67. The sensitivity of the E110 residual ductility (800–1200 C, F/f and F/Q) to the hydrogen concentration at 20 and 135 C 3.67

Fig. 3.68. The data characterizing the sensitivity of the E110 residual ductility at 135 C to the ECR (900–1100 C, F/F and F/Q) 3.68

Fig. 3.69. The comparison of zero ductility thresholds determined from the results of ring tensile and ring compression tests (E110, 1100 C) 3.69

Fig. 3.70. The zero ductility threshold of the E110 cladding determined due to three-point bending tests (1100 C, F/F) 3.69

Fig. 3.71. SEM micrographs for fracture surfaces of the E110 brittle cladding 3.70

Fig. 3.72. High magnification SEM micrographs of fracture surface regions of the E110 brittle cladding 3.71

Fig. 3.73. The SEM micrograph for the fracture surface of the E110 ductile sample 3.71

Fig. 3.74. The maximum load on the E110 oxidized sample as a function of residual ductility 3.72

Fig. 3.75. The comparative data characterizing the E110 residual ductility as a function of the ECR obtained on the processing of test data of different laboratories 3.73

Fig. 3.76. Hydrogen distribution along the width of a special Zry-4 sample 3.74

Fig. 3.77. Demonstration of the end effects on the E110 oxidized cladding samples 3.75

Fig. 3.78. The appearance and microstructure of the E110 irradiated cladding before the tests and after the oxidation tests at 1100 C and F/F combination of heating and cooling rates 3.76

Fig. 3.79. The appearance and microstructure of the E110 oxidized cladding after the slow transient oxidation at 1100 C and standard oxidation at 1200 C 3.77

Fig. 3.80. The residual ductility of the E110 irradiated cladding as a function of the ECR 3.78

Fig. 3.81. Comparative data characterizing the microhardness of the E110 oxidized irradiated claddings 3.78

Fig. 3.82. The zero ductility threshold of the E110 irradiated cladding after different oxidation modes ... 3.79

Fig. 3.83. The E110 hydrogen concentration as a function of irradiation and ECR, residual ductility of the E110 irradiated cladding as a function of hydrogen concentration 3.80

Fig. 3.84. Determination of the rate constant at the oxidation of the E110 unirradiated cladding in the temperature range of 1073–1473 K 3.81

Fig. 3.85. Comparison of the oxidation kinetics for the E110 unirradiated and irradiated claddings 3.82

Fig. 3.86. The comparison of data characterizing the transient test modes with the E110 oxidation kinetics 3.83

Fig. 3.87. The comparative data on the E110 and E635 oxidation kinetics 3.83

Fig. 3.88. The comparison of the E110 and Zry-4 oxidation kinetics 3.84

Fig. 3.89. The E110 oxidation kinetics at 1100 C in accordance with the data obtained in different laboratories 3.84

Fig. 4.1. The relationship between surface scratches and localization of the breakaway oxidation areas .. 4.2

Fig. 4.2. The appearance of the E110 claddings fabricated with the use of two different types of surface finishing before and after oxidation tests at 1100 C 4.5

Fig. 4.3. The microstructure of the E110A and $E110_m$ claddings after the oxidation at 1100 C 4.5

Fig. 4.4. The comparison of residual ductility of the standard E110 as-received tube, E110A and $E110_m$ claddings after the oxidation at 1100 C 4.6

Fig. 4.5. Demonstration of sensitivity of the oxidation and mechanical behavior for the E110 cladding to the cladding surface polishing 4.7

Fig. 4.6. The oxide microstructure on the polished and unpolished parts of the E110 cladding after the oxidation at 1000 C 4.8

Fig. 4.7. Appearances of $E110_{G(fr)}$ samples after the double-sided oxidation at 1100 C 4.10

Fig. 4.8. Results of ring compression tests with $E110_{G(fr)}$ oxidized samples 4.11

Fig. 4.9. Comparative data base characterizing the $E110_{G(3fr)}$ and $E110_{G(3ru)}$ oxidation/mechanical behavior 4.12

Fig. 4.10. The comparison of microstructures for iodide/electrolytic and sponge E110 claddings after the oxidation at 1100 C.......... 4.12

Fig. 4.11. The appearance of the $E110_{low\ Hf}$ cladding after the oxidation at 1100 C and results of mechanical tests.......... 4.13

Fig. 4.12. The appearance and mechanical properties of different E110 claddings after the oxidation at 1000 C.......... 4.15

Fig. 4.13. The comparative test data characterizing the E110 and $E110_{G(3ru)}$ behavior after the oxidation at 900 C.......... 4.16

Fig. 4.14. The characterization of appearance and RT mechanical properties of the $E110_{G(3ru)}$ cladding after the oxidation at 1200 C.......... 4.17

Fig. 4.15. The appearance of the $E635_{G(fr)}$ cladding after the oxidation test at 1100 C and comparative E635 (standard), E110 (standard), $E635_{G(fr)}$ results of ring compression tests.......... 4.18

Fig. 4.16. Comparison of some data on the impurity content in the standard E110 and $E110_{G(fr)}$, $E110_{G(3fr)}$, $E110_{G(3ru)}$ at the beginning of the cladding fabrication.......... 4.21

Fig. 4.17. Low magnification of TEM micrographs for E110, $E110_{G(fr)}$, $E110_{low\ Hf}$ claddings and the M5 cladding [reprinted from 27].......... 4.25

Fig. 4.18. The characterization of the SPP distribution in the α–Zr grains of the E110 and $E110_{G(fr)}$ claddings.......... 4.25

Fig. 4.19. The SPP distribution in the E110 and $E110_{G(fr)}$ claddings.......... 4.26

Fig. 4.20. High magnification of the SPP TEM micrograph in the $E110_{G(fr)}$ cladding.......... 4.27

Fig. 4.21. The oxidation kinetics of different types of Zr-1%Nb claddings at 1100 and 1200 C.......... 4.29

Fig. 4.22. The oxidation kinetics of different types of Zr-1%Nb claddings at 900 and 1000 C.......... 4.30

Fig. 4.23. The characterization of the oxidation rate for the standard E110 (iodide/electrolytic) and E110 claddings manufactured with the use of sponge Zr in the temperature range 900–1200 C.......... 4.31

Fig. 4.24. The difference in the thicknesses of ZrO_2 and α-Zr(O) layers in the E110 cladding of sponge and iodide/electrolytic types at the oxidation at 1000 C.......... 4.32

Fig. 4.25. The difference in the formation of oxide and α-Zr(O) layers in the E110 cladding of sponge type at 1000 and 1100 C.......... 4.32

Fig. 5.1. Outline of the research program.......... 5.1

Fig. 5.2. The schematics of ring compression tests.......... 5.2

Fig. 5.3. The schematic of previous approach to the determination of zero ductility threshold.......... 5.3

Fig. 5.4. The interpretation of ring compression test results on the basis of the load-displacement diagram.......... 5.3

Fig. 5.5. The variability of oxidation tests with different heating and cooling rates 5.4

Fig. 5.6. The appearance of the E110 and Zry-4 claddings (1100 C) as a function of the ECR 5.5

Fig. 5.7. The E110 residual ductility and hydrogen concentration as a function of the ECR after the double-sided oxidation at 1100 C and F/F, F/Q combinations of heating and cooling rates.............. 5.6

Fig. 5.8. Demonstration of the E110 breakaway oxidation effects at 950 C 5.8

Fig. 5.9. The appearance and microstructure of the E110 irradiated claddings as a function of the ECR .. 5.9

Fig. 5.10 The comparison of the E110 oxidation kinetics (1073–1473 K) in accordance with the data of different investigations 5.10

Fig. 5.11. The comparison of the E110 and Zry-4 conservative and as-measured kinetics at 1100 C........ 5.11

Fig. 5.12. The appearance of the E110 polished and unpolished parts after the oxidation at 1000 C......... 5.12

Fig. 5.13. The comparison of the iodide/electrolytic and sponge E110 cladding oxidation behavior at 1100 C 5.13

Fig. 5.14. The comparison of the Zry-4, sponge E110, iodide/electrolytic E110, M5 oxidation kinetics at 1000 C... 5.13

Fig. 5.15. The comparison of the Zry-4, sponge E110, iodide/electrolytic E110 oxidation rates in the temperature range of 900–1200 C 5.14

LIST OF TABLES

Page

Table 2.1. The list of major investigations performed during 1974–1990 to study the fragmentation threshold of unirradiated Zircaloy claddings 2.3

Table 2.2. Results of Russian investigations performed during 1980–2001 to study the oxidation and mechanical behavior of unirradiated Zr-1%Nb (E110) claddings 2.6

Table 2.3. Major results of mechanical tests with the E110 unirradiated oxidized claddings performed in Germany, Hungary and Czech Republic during 1990–2000 .... 2.9

Table 3.1. The list of tasks and technical requirements 3.3

Table 4.1. The E110 surface and bulk effects studies: major provisions of experimental program 4.4

Table 4.2. The specification for the used cladding material 4.9

Table 4.3. The subprogram major tasks for the test with the sponge cladding material and E110 cladding with low Hf content 4.9

Table 4.4. Chemical composition of the E110 alloy (standard) 4.20

Table 4.5. Composition of zirconium alloys used in reactor fuel design 4.21

Table 4.6. The organized results of oxidation and mechanical tests with seven types of the standard E110 and modified E110 .... 4.22

Table 4.7. Chemical composition of the Zirlo cladding tube 4.23

Table 4.8. Zr, Nb content in the matrix, grain boundary and β-Nb precipitates 4.26

Table 4.9. The comparative data characterizing the microstructure of E110, $E110_{G(fr)}$ claddings and the M5 cladding 4.27

LIST OF APPENDICES

Page

Appendix A. Description of Test Apparatus and Test Procedures A-1

Appendix B. Tables with Results of Oxidation and Mechanical Tests: E110, E110A, E110K, E110pol, E635, E110$_{G(fr)}$, E110$_{G(3ru)}$, E110$_{G(3fr)}$, E110$_{low\ Hf}$, E635$_{G(fr)}$, Zry-4 as-Received Tubes and E110 Commercial Irradiated Claddings B-1

Appendix C. Temperature Histories, Appearances and Microstructures of E110 Standard As-received Tubes after a Double-sided Oxidation at 1100 C and S/S, S/F, F/S Combinations of Heating and Cooling Rates C-1

Appendix D. Temperature Histories, Appearances and Microstructures of E110 Standard As-received Tubes after a Double-sided Oxidation at 800, 900, 950, 1000, 1100, 1200 C and F/F (F/Q) Combinations of Heating and Cooling Rates D-1

Appendix E. Appearance and Microstructure of E110 Standard As-received Tubes after a Single-sided Oxidation at 1100 C and F/F Combination of Heating and Cooling Rates E-1

Appendix F. Appearances and Microstructures of E635 Standard As-received Tubes after a Double-sided Oxidation at 1000, 1100 C and F/F Combination of Heating and Cooling Rates F-1

Appendix G. Appearances and Microstructures of Zry-4 As-received Claddings after a Double-sided Oxidation at 1100 C and S/S, F/F Combinations of Heating and Cooling Rates G-1

Appendix H. Appearance and Microstructure of E110, E635 As-received Tubes Manufactured on the Basis of the Sponge Zr, E110$_{low\ Hf}$ As-received Tubes after a Double-sided Oxidation at 900, 1000, 1100, 1200 C and F/F Combination of Heating and Cooling Rates H-1

Appendix I. Appearance and Microstructure of E110 Commercial Irradiated Cladding after a Double-sided Oxidation at 1000, 1100, 1200 C and S/S, S/F, F/F Combinations of Heating and Cooling Rates I-1

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1. INTRODUCTION

To study the oxidation behavior and embrittlement threshold of Zr-1%Nb cladding under loss-of-coolant accident (LOCA) conditions, a research program was developed and implemented by the Russian Research Center "Kurchatov Institute" (RRC KI) in cooperation with the Research Institute of Atomic Reactors (RIAR). The program was performed during the period of 2001–2004 and sponsored by (a) Joint Stock Company "TVEL" (JSC "TVEL", Russia), (b) U.S. Nuclear Regulatory Commission (U.S. NRC, USA), and (c) Institute for the Radiological Protection and Nuclear Safety (IRSN, France).

The incentive to begin this work was directly related the increase of fuel burnup in light-water reactors (LWRs) to 60–70 MW d/kg U and higher. Substantial corrosion is experienced with Zircaloy claddings at fuel burnup higher than 50 MW d/kg U, whereas much less corrosion occurs with Zr-1%Nb cladding during the commercial operation in the Russian type of pressurized-water reactors (VVERs) and with niobium-bearing claddings manufactured from the M5 and Zirlo alloys after operations in the pressurized-water reactors (PWRs). Experimental studies performed with the VVER type of Zr-1%Nb claddings refabricated from commercial fuel rods with burnup up to 60 MW d/kg U have shown that this cladding has a high safety margin under reactivity-initiated accident (RIA) conditions. But the preliminary consideration of safety aspects associated with niobium-bearing claddings under LOCA conditions raised the following issues:

- several investigations performed with Zr-1%Nb cladding of the VVER type in different countries in the 1990s have shown that the niobium-bearing cladding has somewhat different oxidation and embrittlement behavior in comparison with the zircaloy cladding;
- the same general requirements concerning the prevention of the embrittled cladding fragmentation are applied in the LOCA safety analysis of the VVER and PWR reactors, but different approaches are used for this goal.

Taking into account these and other issues, it was decided to perform a special research program including the following main parts of investigations:

- the reassessment of published data concerning the PWR and VVER cladding embrittlement under LOCA conditions;
- the development and validation of test apparatus and test procedures;
- the performance of sensitivity studies and the determination of key factors which must be studied during this program;
- the performance of oxidation, mechanical tests and different pre-test and post-test examinations;
- the analysis and interpretation of obtained results.

The major focus of investigations performed in the frame of this work was concentrated on the characterization of Zr-1%Nb (E110) oxidation and mechanical behavior as a function of such parameters as:

- oxidation conditions (single-sided or double-sided, heating and cooling rates, oxidation temperatures from 800–1200 C, and weight gain);
- mechanical test conditions (ring tensile tests, ring compression tests, three-point bending tests) and test temperature (20–300 C);
- cladding irradiation (as-received and refabricated claddings from the commercial fuel rods with the burnup about 50 MW d/kg U);
- cladding surface conditions (as-received tubes, as-received claddings, polished as-received tubes, ground as-received tubes);
- impurity compositions in the cladding.

In addition to oxidation and mechanical tests with Zr-1%Nb (E110) cladding, several reference tests were performed with the Zircaloy-4 (zirconium-tin) and E635 (zirconium-niobium-tin) claddings. The research program results are presented in this report.

2. BACKGROUND

The prevention of cladding fragmentation in LWR-type reactors under the LOCA conditions is one of the basic principles of the current safety philosophy. The reason for this is related to the following physical phenomena:

- significant increase of the cladding temperature during the LOCA accident caused by the coolant blowdown and degradation of heat transfer;
- high temperature cladding steam oxidation accompanied by cladding embrittlement;
- possible fracture of the embrittled cladding caused by post-LOCA forces during quenching and post-quenching actions.

In accordance with the current world practice, the prevention of the cladding fragmentation under LOCA conditions is assured by special safety criteria. Thus, the main Russian regulatory document contains two special requirements concerning this problem: for the Zr-1%Nb (E110) cladding [1]:

- the peak cladding temperature (PCT) must not exceed 1200 C;
- the local oxidation depth (Equivalent Cladding Reacted layer, ECR) must not exceed 18% of the initial wall thickness.

Similar criteria are contained in the regulatory documents of other countries for the zircaloy cladding (1200 C, 15–17% ECR). It should be noted that the concept for the use of fragmentation criteria of these types was developed by the NRC (USA) with respect to the zircaloy claddings [2]. The motivation for the choice of this approach for the safety fragmentation criteria was reconstructed in the recent paper prepared by H.Chung and G.Hache [3].

The first research to determine the zero ductility threshold of oxidized Zry-4 claddings after the quench cooling was performed by D.O. Hobson at the beginning of 1970s [4, 5]. In accordance with results of a slow compression of Zry-4 oxidized samples, he revealed the relationship between the critical relative thickness of the prior β-phase and zero ductility threshold at 135 C (the saturation temperature during the reflood stage). This relationship was used to develop the following embrittlement criterion [6]:

$$\frac{\xi_T}{W_o} \leq 0.44\,,$$

where ξ_T – the thickness of the oxygen-rich cladding layers (ZrO_2 and α-ZrO);

W_o – the initial thickness of the unoxidized cladding.

Further, it was revealed that the cladding ductility margin at low temperatures (150 C or less) was a function of not only the prior β-phase thickness but also of the oxygen concentration in this layer. The appropriate analysis of Hobson's test has shown that:

- the maximum oxygen concentration in the prior β-phase is a function of the oxygen solubility in the β-phase under high temperature oxidation conditions;
- the zero ductility threshold (at 20 - 150 C) is associated with 0.7% (by weight) oxygen concentration in the prior β-phase. This critical oxygen concentration is achieved very fast if the oxidation temperature is higher than 1204 C (2200 F).

Taking into account these test results, the embrittlement criterion was added with the Peak Cladding Temperature (PCT) criterion. The PCT criterion was estimated as 1204 C [6].

The final evolution of these criteria involved the following:

- the extension of the test data base needed to validate criteria [7, 8, 9];
- the introduction of the reasonable conservative principle into the safety analysis procedure.

As for the conservatism, it was decided to organize the results of different tests into the unified system using the Baker–Just equation allowing to calculate the Zry-4 high temperature kinetics with the conservative margin [10]. This equation was used to determine the oxygen weight uptake with each tested sample. Besides, to improve the physical interpretation of calculated results, the concept of the Equivalent Cladding Reacted (ECR) layer was introduced:

$$ECR = \frac{\delta_e}{\delta_o},$$

where δ_l – oxidation equivalent layer determined using the following condition: the all uptaken oxygen are used for the formation of the stoichiometric zirconium dioxide (ZrO_2);

δ_o – the initial cladding thickness.

The practical implementation of this approach allowed the development of a test data base with the following list of test parameters:

- ECR calculated with the Baker–Just;
- oxidation time and temperature;
- tested cladding sample characterization: intact, failed.

The analysis of this data base presented in Reference 3 shows that:

- at an oxidation temperature less than 1204 C, the brittle fracture of oxidized claddings was not observed under ring compression test conditions (at 135 C and higher) if the ECR calculated with the Baker–Just correlation was less than 17%;
- at an oxidation temperature less than 1600 C, the brittle fracture of oxidation claddings was not observed under thermal-shock during direct quenching conditions if the ECR calculated with the Baker–Just correlation was less than 19%.

Thus, the results of two different types of tests (the comparison of mechanical tests and thermal–shock tests) demonstrated the similarity in the evaluation of the critical oxidized thickness (17–19% ECR) although there was a significant discrepancy in the estimation of the permissible peak cladding temperature. These results led the experts responsible for the development of the proposal on safety criteria to the formulation of their position concerning the choice of the permissible peak temperature [11]. This position may be characterized by the following general provisions:

- the practical application of thermal–shock test results requires such a detailed knowledge of physical processes during the LOCA which cannot be provided;
- to prevent the fragmentation, the oxidized cladding must retain some margin of ductility;
- the choice of 1204 C (2200 F) PCT limit based on results of compression mechanical tests provides the conservative margin in comparison with the thermal–shock test results.

In 1973, this position was used to formulate the NRC criteria [2]: 1204 C, 17% ECR (calculated using the Baker–Just equation). During the period of 1974–1990, experimental investigations with zircaloy claddings were continued to understand the sensitivity of fragmentation threshold to such factors as:

- the cladding ballooning and burst;
- the mechanical interaction of the oxidized cladding in the fuel bundle caused by ballooning and bending;
- the axial mechanical constraint of ballooned cladding by the grid spacers.

A brief description of this cycle of Zry-4 investigations is presented in Table 2.1. The main outcomes of these investigations may be characterized in the following way:

1. The thermal–shock tests performed with the original geometry of the oxidized cladding (without axial constraint or ballooning and burst) or with the original geometry of fuel rod simulators have shown that the current safety criteria (1204 C, 17% ECR) have a margin of about 100% in ECR. The margin in PCT does not exceed 150 C.
2. Thermal–shock studies performed to check the constraining effect of the grids when using deformed cladding (ballooning and burst) have shown that the fragmentation threshold decreased significantly with axial constraint, but the test fragmentation threshold did not exceed the safety criteria.

3. Special impact tests performed with the simulation of potential impact fracture energy (estimated as 0.3 J) have shown that:

- the oxidized cladding with the original geometry had a high margin before fracture;
- the fragmentation threshold of the deformed cladding (after ballooning and burst) was in agreement with the safety criteria. Some claddings were fragmented at values lower than 17% (safety criterion), and the increase in hydrogen content in local parts of the oxidized claddings (local cladding hydriding) was the cause of this effect, but the peak cladding temperature was higher than 1204 C (safety criterion).

4. Additional studies of the hydriding effect performed with compression tests demonstrated that the zero ductility threshold of hydrided cladding corresponded to 700 ppm of hydrogen in the prior β-phase.
5. The axial tensile and ring compression tests confirmed that the zero ductility threshold was reached when the average oxygen concentration in the prior β-phase increased from 0.6% by weight up to 0.8% by weight.

Taking into account results of all these tests and understanding of the fact that the ductility margin of the oxidized cladding is a function of oxygen and hydrogen concentration in the prior β phase, several investigators proposed to change the current safety criteria (1204 C, 17% ECR) into criteria based on the oxygen and hydrogen concentrations in the oxidized claddings. But these suggestions were not apparently implemented due to the fact that:

- the introduction of these criteria must be accompanied by the use of computer codes for the safety analysis which are able to calculate the appropriate parameters with the required accuracy;
- numerous additional experimental programs would be needed to develop and validate the integral criterion based on the oxygen and hydrogen content in the prior β-phase of the oxidized cladding.

In accordance with these considerations, improvement of the safety criteria was postponed for the time being.

Table 2.1. The list of major investigations performed during 1974–1990 to study the fragmentation threshold of unirradiated Zircaloy claddings

<table>
<tr><th>Test type</th><th>Test characterization</th><th>Test results</th></tr>
<tr><td>1. Thermal shock tests performed by H.Chung and T.Kassner, USA, 1980 [12]</td><td>The double-sided oxidation of Zry-4 claddings at 1000–1500 C with slow cooling though phase transition followed by quench type cooling</td><td>The cladding fragmentation threshold was 28% ECR (measured) at 1500 C and 33% ECR at 1200 C</td></tr>
<tr><td rowspan="2">2. Thermal shock tests performed by H.Uetsuka et. al., Japan, 1983 [13]</td><td rowspan="2">The double-sided oxidation of Zry-4 claddings at 950–1350 C with the quench cooling</td><td>The cladding fragmentation was not noted in this temperature range at 17% ECR (as-calculated using Baker-Just correlation)</td></tr>
<tr><td>The cladding fragmentation threshold was higher than 35% ECR (as-calculated using Baker-Just correlation) at 1200 C</td></tr>
<tr><td rowspan="2">3. Thermal shock tests with the cladding axial mechanical constrain performed by H.Uetsuka et. al., Japan, 1983 [13]</td><td rowspan="2">The double-sided oxidation of Zry-4 cladding with the strong fixation of one cladding end at 900–1300 C. The fixation of the second cladding end at the beginning of the quench cooling</td><td>The cladding fragmentation was not observed in this temperature range at 17% ECR (as-calculated using Baker-Just correlation)</td></tr>
<tr><td>The cladding fragmentation threshold was 20% ECR (as-calculated using Baker-Just correlation) at 1200 C</td></tr>
</table>

Test type	Test characterization	Test results
4. The in-pile tests (PHEBUS research reactor) of the tests fuel bundle performed by M.Reocreux and E. Scott de Martinville, France, 1990 [14]	The LOCA-type test (#219) without the quench mode with Zry-4 claddings: the cladding ballooning and burst at high temperature, the cladding oxidation	The cladding fragmentation of one fuel rod occurred at the following parameters: • 16% ECR • 1330 C The reason of the fragmentation was assessed as the mechanical constrain of the temperature-induced cladding replacement
5. Impact tests of oxidized claddings of original geometry performed by H.Chung and T.Kassner, USA, 1980 [12]	The double-sided oxidation of Zry-4 claddings at 1100–1400 C and the cooling rate 5 C/s. The impact tests of oxidized claddings	The failure impact energy was higher than 0.8 J* [15], if the oxidation temperature did not exceed 1315 C and the ECR did not exceed 17% (as-measured using the metallographic method)
6. Impact tests of deformed (after the ballooning and burst) and oxidized claddings performed by H.Chung and T.Kassner, USA, 1980 [12]	Fuel rods with the Zry-4 cladding and fuel pellet simulators were pressurized at the high temperature up to the ballooning and burst. After that, fuel rods were oxidized (under isothermal conditions) and quenched. The impact tests of these fuel rods were performed at the fixed impact energy 0.3 J	A good correlation between the cladding fragmentation threshold and 17% ECR (as-measured with the use of metallographic method), 1204 C was observed for the most tested fuel rods. A special analysis has shown that: • some parts of the inner surface of deformed claddings are characterized by the formation of a thick spalled oxide • it is revealed that stagnant steam/ water conditions are responsible for the initiation of the breakaway oxidation on these parts of the cladding • the high hydrogen uptake up to 2200 ppm was noted in these specific zones • it is observed that the cladding fragmentation threshold sharply decreases at the cladding hydrogen content 700 ppm and higher
7. The compression tests of oxidized and hydrating claddings performed by H.Chung and T.Kassner, USA, 1980 [12]	The mechanical compression tests were performed with fuel rods (Zry-4 cladding, fuel pellet simulators) after the pressurization of the fuel cladding up to the burst and high temperature oxidation	These scoping tests have shown that the residual ductility of the oxidized cladding is a strong function of oxygen and hydrogen content in the prior β-phase
8. The ring compression tests of deformed and oxidized claddings performed by H.Uetsuka et. al., Japan 1981–1982 [16, 17]	Zry-4 claddings were heated, pressurized up to the burst, oxidized and quenched. The compression tests were performed at 100 C with ring samples which were cut off from oxidized claddings	A strong correlation between the hydrogen content in the prior β-phase and cladding residual ductility was developed It was shown that the cladding was fully embrittled at 700 ppm of hydrogen content

* The expert estimations of possible loads in the commercial fuel bundle during late stages of LOCA have shown that the impact energy may achieve 0.3 J

Test type	Test characterization	Test results
9. The tensile mechanical tests of the oxidized performed by A.Sawatzky, UK, 1978 [18]	The Zircaloy claddings were oxidized in the water steam at 1000–1600 c. After that, the tensile tests were performed at room temperature. the test data base was added with the microhardness measurements across the cladding thickness	It was revealed that the oxygen in the prior β-phase was distributed nonuniformely It was shown that the oxidized cladding had some ductility margin if the average oxygen content in the prior β-phase did not exceed 0.6% by weight It was determined that the zero ductility threshold of the oxidized cladding corresponded to 0.8% averaged oxygen content (the appropriate ECR was 16%)
10. The in-pile tests of fuel rods performed in the PBF research reactor, USA, 1982 [19]	The single pressurized fuel rods with the Zry-4 cladding were oxidized at the temperature transient mode	Several fuel rods which failed after the tests at the post-test manipulations had the following parameters: • the fuel rod with the burst at 1100 C oxidized up to 12% ECR (as-measured) at the equivalent temperature 1262 C • three fuel rods oxidized at equivalent temperature 1300 C up to 5–11% ECR (as-measured) • fuel rods oxidized at equivalent temperature up to 7–8% ECR (as measured) were not fragmented

New attempts to resume this activity were made in the middle of 1990s in the context of the increase in fuel burnup up to 60 MW d/kg U and higher in the LWRs. An important aspect of this new stage of investigations was connected with the fact that zircaloy cladding has a tendency towards the breakaway oxidation and cladding hydriding at the high burnup (55 MW d/kg U and higher) under normal operation conditions. These effects result in a decrease of cladding ductility and lead to questions concerning the mechanical behavior of these claddings under accident conditions. The importance of this problem was shown practically in the experiments with the Zry-4 irradiated cladding under reactivity-initiated accident (RIA) conditions [20, 21, 22].

Taking into account the revealed problems, extended investigations were initiated to study the irradiation effects in zircaloy claddings under LOCA conditions [23, 24, 25, 26, 27]. The important direction of these investigations was connected with advanced cladding materials including the niobium-bearing alloys. In this context, it should be noted that a niobium-bearing alloy E110 (Zr-1%Nb) had been used as the VVER cladding material for several decades. Moreover, special investigations performed in 1990s showed that this alloy demonstrated a very high corrosion resistance under normal operation conditions up to 60 MW d/kg U [28].

Further, special investigations devoted to measurements of mechanical properties of E110 irradiated claddings under accident conditions [29, 30, 31, 32] and experimental studies of VVER high burnup fuel rods (50–60 MW d/kg U) under RIA conditions [30, 33, 34, 35] have demonstrated that fuel rods with the Zr-1%Nb (E110) cladding have good prospects in respect to the increase of the fuel burnup in VVERs. But it is obvious that the final analysis of this situation cannot be made without the consideration of experimental results characterizing the ductility margin of E110 claddings after the high temperature oxidation and quenching under LOCA-relevant conditions.

The official history of such investigations with the E110 alloy was initiated in Russia goes back to the beginning of the 1980s. Previous investigations used for the development of the second design limit of fuel rod damage (1200 C, 18% ECR) in the first national regulatory document on LWR safety issues [36] were not described in the open publications. The outline of main Russian research programs performed during the 1990s to develop an experimental data base characterizing the oxidation and mechanical behavior of the E110 cladding is presented in Table 2.2.

Table 2.2. Results of Russian investigations performed during 1980–2001 to study the oxidation and mechanical behavior of unirradiated Zr-1%Nb (E110) claddings

Test type	Test characterization	Test results
1. High temperature oxidation tests performed in VNIINM* and VTE** [37, 38, 39, 40]	The double-sided oxidation of E110 claddings at 700–1500 C under isothermal and transient conditions	The test data base needed to develop the E110 oxidation kinetics correlations was obtained
		The effect of ZrO_2 spallation was revealed in the temperature range 900–1100 C on achieving the critical oxide thickness (~25 μm)
		A significant change of the cladding geometrical sizes was revealed for some transient modes
2. Ring tensile tests of oxidized claddings performed in VNIINM, 1990 [41]	The double-sided oxidation of E110 claddings at 1000–1200 C with the direct current heating and fast air cooling. Ring tensile tests of oxidized samples at 20–1000 C	The zero ductility threshold was associated with the weight gain 450 mg/cm^2 (~6% as-measured ECR) at 20 C
		The reasonable margin of residual ductility in fully brittle samples was revealed at temperatures higher than 500 C
3. Impact tests of oxidized claddings performed in VNIINM, 1990 [41]	The double-sided oxidation of E110 claddings at 1000–1200 C with the direct current heating and cooling rate 10–20 C/s. The impact tests of oxidized claddings at 20 C	The brittle fracture occurred at the weight gain higher than 600 mg/dm^2 (7% ECR)
		The relationship between the failure specific impact energy of the unoxidized sample and brittle oxidized sample (500 mg/dm^2) was assessed as 100 J/cm^3 and 5 J/cm^2, respectively
4. Development of the E110 conservative oxidation kinetics performed in VNIINM, 1990 [42, 43]	The test data base obtained due to investigations stated in item 1 was used	The following correlation was developed: $\Delta m = 920 \exp\left(-\frac{10410}{T}\right)\cdot\sqrt{\tau}$, where Δm–oxygen weight gain (mg/dm^2) T–temperature (K) τ–time (s)
5. High temperature oxidation tests of deformed claddings performed in VNIINM and VTE, 1990–91 [42, 43]	The oxidation tests of pressurized (Ar) cladding samples at 700–850 C. The measurement of the cladding hoop strain in the ballooning area (the variation of the outer diameter relative increment was 0–85 % ($\Delta D/D_o$)	The systematic increase of the oxygen weight gain was observed as a function of $\Delta D/D_o$ increase. The dependence of the weight gain on the cladding surface in the ballooning area was estimated
6. Thermal shock tests of oxidized claddings performed in VNIINM and VTE, 1990–91 [42, 43, 44]	The double-sided oxidation of E110 claddings at 800–1200 C and water quench cooling	The fragmentation did not occur for all claddings with the ECR less than 18% (as-measured)
7. Ring compression tests performed in VNIINM and VTE, 1990 [42, 43]	The double-sided oxidation of E110 and Zry-4 (French and SANDVIK zircaloy) claddings at 800–1200 C and water quench cooling. (Reference tests with slowly cooled samples). Reference tests with the air oxidation of E110 claddings. Ring compression tests of 30 mm oxidized claddings at 20 C	Ring compression tests showed that: • a sharp decrease of the E110 ductility occurred after the achievement of some criterial ECR • this criterial ECR was a function of the oxidation temperature • the worst studied temperature was 1000 C • the best studied temperature was 800 C • the range of the E110 zero ductility threshold may be estimated as 3–4% (as-measured)

* All-Russian Research Institute of Inorganic Materials

** All-Russian Heat Engineering Institute

Test type	Test characterization	Test results
		• the Zry-4 oxidized cladding had the monotonic character of ductility reduction at the ECR increase. The zero ductility threshold of Zry-4 cladding was not higher than 15% ECR (as-measured)
		The visual observations of oxidized samples allowed to reveal that: • at ECRs close to the criterial value (3–4%) the white spalled oxide appeared on the E110 cladding surface • Zry-4 oxidized samples were covered with the black bright oxide
		The comparative analysis of results of hydrogen content measurements showed that E110 claddings unlike Zry-4 had the tendency towards the high hydrogen absorption at 1000–1100 C. Reference tests with air oxidation confirmed that the E110 residual ductility increased significantly in this case (without the cladding hydriding effect)
		The comparative data characterizing the sensitivity threshold to the cooling rate showed that this effect was insignificant
		The metallographic studies of the E110 oxidized cladding allowed to note that: • the following difference in the E110 and Zry-4 α-Zr(O) phase morphology was revealed: – the α-Zr(O) phase in the Zry-4 cladding consisted of equiaxed grains; – the α-Zr(O) phase in the E110 cladding consisted of thin plates • taking into account the difference in the phase transition temperatures for E110 and Zry-4 alloys, the E110 β-phase must dissolve more oxygen than the Zry-4 one to provide the α-Zr(O) phase initiation condition. This effect led to the difference in the α-Zr(O) thickness in E110 and Zry-4 claddings
8. Thermal shock tests of VVER fuel rod simulators performed in VNIINM and RIAR*, 1998–2001 [45, 46, 47, 48]	The oxidation of unpressurized fuel rod simulators with the E110 cladding at 900–1200 C, after that water quenching of oxidized simulators. Two types of simulators are used: • with Al_2O_3 fuel pellets and radiant heating of the fuel rod • with UO_2 fuel pellets and W-heaters installed inside the fuel stack One end of the fuel rod was open for the steam penetration	Both types of VVER fuel rod simulators were not fragmented during and after thermal shock tests in the following range of test parameters: • 900–1200 C • as-calculated ECR (using the E110 conservative correlation) less or equal to 18%
		The different margin for the safety fragmentation threshold (18% ECR) was demonstrated as a function of the oxidation temperature and simulator type

* State Research Center "Research Institute of Atomic Reactors"

Test type	Test characterization	Test results
9. Impact tests of oxidized VVER fuel rod simulators performed in VNIINM, 1998–2001 [45–48]	The E110 oxidized cladding after investigations performed as stated in the item 8 were tested in accordance with the following requirements	The reference tests with the unoxidized cladding shown that the impact-toughness fracture was of 64–89 J/cm^2
	• claddings removed from the first type of fuel rod simulators were used • the circamferel notch 1–1.5 mm deep and 0.5 mm wide was made on each 100 mm oxidized cladding • the impact tests were performed at 20 C	Impact tests with the oxidized cladding demonstrated that: • the impact-toughness fracture was reduced down to 2.5 J/cm^2 at 5% ECR (as-calculated using the E110 conservative correlation) • the impact-toughness fracture at 10–15% ECR (as-calculated) was about 1 J/cm^2 • the tough type of the cladding fracture was observed up to the 5% ECR • the brittle fracture occurred at 7% ECR
10. Compression tests of E110 oxidized claddings performed in VNIINM, 1998–2001 [45–48]	The double-sided oxidation of E110 claddings at 800–1200 C with water quenching. The compression tests of oxidized samples 30, 50 mm long at 20–900 C	Test data in general confirmed the results of previous tests: • the oxidation at 800 C was much better than the oxidation at 1000 C
	The compression tests (at 20 C) of oxidized claddings (18% as-calculated ECR) taken from type two simulators tested in accordance with requirements given in the item 8	• the relative displacement at failure was 4% at the following combinations of as-calculated ECR and the oxidation temperature: – 800 C: 10% ECR – 1000 C: 5% ECR • no new effects were revealed in the tests of claddings taken from the fuel rod simulators
		The following effects of the mechanical test temperature were noted: • the ductility of E110 oxidized at 200 C occurred if the ECR was less than 5% • at 18% ECR, the temperature effect in the range 20–500 C was insignificant

It should be noted that intensive studies with the E110 irradiated cladding were started in the middle of 1990s in addition to the test program presented in Table 2.2. The first results of investigations performed with the E110 irradiated claddings refabricated from VVER high burnup fuel rods (50 MW d/kg U) under LOCA conditions are presented in References 46–48.

The analysis of the whole scope of obtained results allowed to conclude that:

1. Numerous thermal–shock tests performed with different E110 samples (unirradiated and irradiated oxidized claddings and fuel rod simulators) showed that the E110 fragmentation threshold was higher than 18% ECR (calculated with the VNIINM conservative correlation) in the temperature range 800–1200 C.
2. The effect of breakaway oxidation accompanied by the hydrogen uptake was revealed in E110 claddings oxidized at temperatures higher than 800 C and relatively low ECRs.
3. Mechanical tests (tensile, compression, impact) demonstrated that a sharp decrease in residual ductility of the E110 oxidized cladding occurred in the measured ECR* range 4–7%.

* The measured ECR should not be compared with the safety criterion (18%) because (as it was noted earlier) the calculated ECR was used for the safety and analysis. That ECR was calculated using the conservative oxidation kinetics.

During 1990s several research programs devoted to the high temperature oxidation behavior of the E110 cladding were initiated in Germany, Hungary and the Czech Republic. Major findings of these test programs obtained by 2001 are presented in Table 2.3.

Table 2.3. Major results of mechanical tests with the E110 unirradiated oxidized claddings performed in Germany, Hungary and Czech Republic during 1990–2000

Test type	Test characterization	Test results
1. Ring compression tests performed by J.Böhmert in Germany (NC Rossendorf), 1992 [49, 50]	The double-sided oxidation of E110 and Zry-4 (SANDVIK) in water steam at 800–1100 C with the water quench cooling. Ring compression tests at 20 C	Results of mechanical tests showed that: • the ductility of the E110 oxidized cladding (850–1100 C) decreased sharply down to the zero ductility threshold in 2(3)-4(5,6)% range of the as-measured ECR • the ductility of E110 claddings oxidized at 800 C was high up to the 8% ECR (the maximum value was achieved under this test conditions) • the Zry-4 ductility decreased monotonically as a function of ECR, relative displacement at failure was 8% at 18.5% ECR (as-measured)
		The visual observations showed that: • the white porous spalled oxide covered the E110 cladding sample at the relatively low ECRs • the black bright oxide covered the Zry samples
		The comparative measurements of hydrogen content in the oxidized claddings demonstrated that E110 had the tendency towards the high hydrogen absorption in contrast to the Zry-4 cladding (especially at 1000 C). So, the maximum hydrogen content at the ECR of about 18% (as-measured) was: • 2050 ppm in E110 • 130 ppm in Zry-4
		The microhardness measurements allowed to reveal that the microhardness (oxygen concentration) in the prior β-phase was significantly higher in the E110 cladding at the following test parameters: • temperature oxidation: 900–1100 C • oxidation time: 30 min
2. Thermal shock tests performed in Hungary [51]	The oxidation of 50 mm fuel rods simulators (E110 cladding, Al_2O_3 pellet) at 1000–1250 C with the water quench cooling	The cladding fragmentation did not occur if the ECR (calculated using the VNIINM conservative correlation) was less than 30%
3. Ring compression tests performed in Hungary [51, 52]	The double-sided oxidation of E110 and Zry-4 samples at 900–1200 C. Ring compression tests at 20 C	Visual observations showed that: • the black bright oxide covered the Zry-4 samples in most cases • the white spalled oxide covered E110 samples at relatively low ECRs
		The mechanical tests allowed to reveal that: • the Zry-4 ductility decreased monotonically at the ECR increase • the E110 ductility decreased sharply in the ECR range (as-measured) 1.6–5%

Test type	Test characterization	Test results
		The hydrogen content measurement showed that the Zry-4 hydrogen uptake was very low, the E110 hydrogen content achieved 600–800 ppm at the 5% ECR in the temperature range 900–1100 C. The E110 hydrogen absorption rate was somewhat slowed down at 1200 C
4. Thermal shock tests performed in Czech Republic [53, 54]	The double-sided oxidation of 30 mm E110 cladding samples at 800–1200 C with the water quench cooling	The E110 cladding fragmentation threshold was observed at ECRs higher than 30% (as-measured)
5. Ring compression tests performed in Czech Republic [53, 54]	The double-sided oxidation (argon and steam mixture) of 30 mm E110 cladding samples at 800–1200 C with the water quench cooling. Ring compression tests at 20 C	The visual observations showed that: • the E110 steam oxidation at temperatures higher than 800 C led to the formation of the light color oxide and flaking-off effect (but without nodular corrosion effects) • the E110 heating in the argon atmosphere up to the isothermal oxidation temperature led to the formation of the black lustrous oxide on the E110 surface
		The ring compression tests and hydrogen content measurements allowed to reveal the following: • the zero ductility threshold of the E110 cladding was associated with 5% ECR (as-measured) • besides, the zero ductility condition was accompanied by the hydrogen content 500–700 ppm in the prior β-phase • the further ECR increase led to the oxide spallation and the increase of hydrogen content up to 2000 ppm

The experimental data organized in Table 2.3 confirmed that:

- the tested E110 claddings had a tendency towards breakaway oxidation and the hydrogen uptake at temperatures 850–1200 C;
- a sharp decrease in E110 ductility occurred at measured ECRs of 4–6%;
- fragmentation of E110 oxidized cladding was not observed at a calculated ECR less than 18% (1200 C) in accordance with the thermal–shock tests.

Analysis of results of experimental investigations with the E110 cladding performed in Russia and abroad lead to the following observations:

1. All investigations presented in Table 2.2 and Table 2.3 were performed with as-received E110 tubes manufactured during 1980s. But numerous improvements and changes were made during the 1990s in procedures for producing Zr ingots and fabricating E110 cladding.
2. The first published results characterizing the oxidation and mechanical behavior of other Zr-1%Nb cladding (French M5 alloy) showed that earlier breakaway oxidation and high hydrogen uptake were not observed in these tests with niobium-bearing cladding [55].
3. Procedures of many oxidation tests (including the oxidation history, heating and cooling rates, temperature and weight gain measurements, steam flow rate value, etc.) were not validated and documented along with the published data. The nature of cladding material used in investigations performed outside of Russia was not documented also. Procedures of mechanical tests, parameters of cladding samples and processing of obtained results were quite different in some cases and often unknown in other cases.

4. Taking into account that niobium-bearing alloys are of a high priority with respect to the increase of LWR fuel burnup, an understanding of physical phenomena defining the mechanical behavior of these claddings under accident conditions is an important international research task.

These considerations have led to the decision to perform the special research program concerning the experimental study of embrittlement of Zr-1%Nb (E110) cladding under LOCA–relevant conditions.

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