# MODELING OF QUENCH-16 EXPERIMENT WITH MAAP4 SEVERE ACCIDENT CODE

### E. BEUZET

EDF R&D 1 Avenue du Général de Gaulle, 92140 Clamart, France emilie.beuzet@edf.fr

#### M. STEINBRÜCK, J. STUCKERT

Karlsruhe Institute of Technology Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany martin.steinbrueck@kit.edu, juri.stuckert@kit.edu

# ABSTRACT

In a nuclear power plant, a potential risk in some low probable situations of severe accidents is air ingress into the vessel, due to a chimney effect from core heat. Air is a highly oxidizing atmosphere that can enhance core oxidation and degradation intensified by nitride formation. In addition, air may have an effect on the release of fission products, especially that of ruthenium which is of particular interest due to its high radio-toxicity and ability to form highly volatile oxides. Because of an oxygen affinity decrease from Zircaloy claddings to fuel matrix and ruthenium inclusions, claddings are firstly oxidized by air. It is consequently fundamental to establish the phenomena governing their oxidation by air, and their simulation, as a prerequisite about the fission products release and source term issues in case of an air ingress scenario.

The QUENCH-16 experiment was conducted at KIT in the frame of the European LACOMECO programme in order to extend the database on Zircaloy cladding oxidation by air. Following test phases were applied to represent a realistic air ingress scenario: bundle heatup, steam oxidation due to water boiling, air oxidation due to air ingress and quenching. In order to enhance phenomenological understanding, the main goal of this test was to achieve a limited pre-oxidation of the cladding by steam and an extended oxygen starvation during the air phase. The first analyses from experimenters show that the main objectives were attained.

The QUENCH-16 experiment was simulated with the EDF version of MAAP4 code in which cladding oxidation by air is treated using weight gain correlations from the literature. Previous studies on QUENCH-10 and PARAMETER-SF4 experiments pointed up the NUREG correlation as the best current compromise solution. The different experimental parameters needed for the simulation of QUENCH-16 were defined in collaboration with the experimenters and enhanced the precision of the modeling. The simulation gives encouraging results and underlines ways to improve the modeling.

#### 1. Introduction

In some low probable situations, severe accidents in PWRs can lead to core degradation and relocation of molten material from the core into the lower head. Heat exchange between the molten debris and the vessel may lead to the rupture of the vessel. Air may be drawn from the containment into the vessel, governed by a chimney effect due to residual heat in core.

Air creates an oxidizing atmosphere inside assemblies still in core. Cladding oxidation by air induces a higher exothermic reaction than in steam [1], which can enhance core degradation and fission products release (notably ruthenium) from the fuel, their transport through the primary circuit, their release in the containment and possibly in the environment.

A number of previous air ingress bundle experiments on claddings have been performed under a range of configurations and oxidising conditions, namely AIT-1, AIT-2 [2], QUENCH-10 [3] and PARAMETER SF4 [4]. The accumulated data have demonstrated that cladding air oxidation is a remarkably complicated phenomenon governed by numerous processes whose role can depend critically on the oxidising conditions, the preceding oxidation history and the details of the cladding material specification. Hence, extensive separate-effects tests have been performed recently for better understanding of the mechanisms of air oxidation of zirconium alloys and extraction of corresponding data mainly at IRSN [5, 6] and KIT [7, 8]

The QUENCH-16 test was performed in the frame of the EC-sponsored LACOMECO programme [9]. The proposal included a target scenario characterised by: 1) a long period of oxygen starvation; 2) reflood initiated at temperatures well below the melting point of the

cladding. Pre-test calculations with different severe accident codes have been conducted to help define the experimental conditions [10,11].

The present work gives an overview of the QUENCH-16 experiment and its modeling with the MAAP4 code, which is dedicated to the NPPs severe accidents simulation [12].

### 2. Results of QUENCH-16 experiment

The QUENCH test facility is described in detail in [3]. Fig. 1 shows a schematic of the test section with the 21-rod simulator bundle and the various inlets for steam, air and water. The bundle composed of 20 rods heated by tungsten heaters (prolonged with electrodes of molybdenum/copper in upper and lower bundle parts) and one central unheated rod, which allows to check the rod integrity without support of rigid heater. An electrical power supply is connected to the electrodes with cables and a special slide contacts. The electrical resistance of heaters/electrodes depends strongly on the test temperature (factor 3 between room conditions and time point of peak test temperatures), whereas the resistance of permanently cold cables and contacts fluctuates slightly around 5 m $\Omega$ /rod. The off-gas was analysed by a mass spectrometer.

Fig. 2 illustrates the various phases of the test. In the pre-oxidation phase and following slow cooling phase the claddings were oxidised in superheated steam (3.3 g/s) during about 7300 s. It led to a maximal 130  $\mu$ m ZrO<sub>2</sub> scale measured at withdrawn corner rod (at 7249 s). In the subsequent air ingress phase, which lasted 4035 s, the steam flow was replaced by 0.2 g/s of air at about 7300 s. The change in flow conditions had the immediate effect of reducing the heat transfer so that the temperatures began to rise again. After some time measurements demonstrated gradually an increasing consumption of oxygen, accompanied by acceleration of the temperature increase at some locations. The faster increase was most pronounced at the mid elevations of the bundle. Oxygen was completely consumed at about 3200 s after beginning of air ingress. Shortly before that time, partial consumption of the nitrogen was first observed, indicating local oxygen starvation which promoted the onset of nitriding. Following this, the temperature continued to increase until water injection was initiated when the maximum observed temperature was ca. 1873 K. Thus there was a period of about 835 s complete oxygen consumption and hence starvation in at least part of the bundle. The total uptakes of oxygen and nitrogen were about 58 and 29 g, respectively. The generally limited rate of temperature increase was the result of a rather low air flow rate, probably prototypic of reactor conditions. A residual steam flow rate was measured during the oxygen starvation, characterised by a small hydrogen production. This is the subject of ongoing investigation.

Reflood was initiated by injecting 50 g/s of water. Almost immediately after the start of reflood there was a temperature excursion in the mid to upper regions of the bundle (500 to 1400 mm), leading to maximum measured temperatures of about 2420 K. Cooling was established at the hottest location ca. 70 s after the start of injection, but was delayed further at other locations. Reflood progressed rather slowly, perhaps due to the high temperatures and partial degradation, and final quench was achieved after about 500 s. In line with the temperature escalations, a significant quantity of hydrogen was generated during the reflood (128 g). There are also indications of nitrogen release during the quench phase (24 g from 29 g consumed during oxygen starvation period).



Fig. 1. Containment and test section.

Fig. 2. QUENCH-16 test conduct.

Post-test examinations of cross sections between 300 and 500 mm reveal frozen partially oxidised melt (Fig. 3), relocated from upper elevations 500 - 800 mm, which could have been the main source of hydrogen during reflood. At elevations 800 - 900 mm only local melt was observed between pellet and outer oxide layer. No melt was formed at elevations above 900 mm.

According to metallographic investigations at a corner rod, withdrawn at the end of the air ingress phase, intensive nitride formation was observed at elevations 300 - 900 mm. Posttest investigations of cross sections reveal many residual nitride traces at elevations 350 - 550 mm. The upper oxide scales above nitrides have a porous structure due to re-oxidation of nitrides during reflood. At elevations above 550 mm only some nitride traces at the boundary between inner dense and spalled outer porous oxide scales were observed (Fig. 4).



Fig. 3. Bundle cross section at 430 mm: frozen melt relocated from upper elevations



Fig. 4. Cladding structure elevation 550 mm: <u>nitrides</u> between inner dense and outer porous oxide layers

#### 3. Modeling of QUENCH-16 experiment with MAAP4

#### 3.1. Modeling parameters

The QUENCH-16 experiment is meshed in MAAP using 3 radial rings of respectively 5 (with the central unheated rod), 8 and 8 rods and 58 axial meshes with 48 for the heated core part, 5 for the lower and upper plenums (Fig. 5). The shroud, the thermal insulation and the cooling circuit are also modeled. The initial conditions, the geometric and transient data, notably the different flow rates and the electrical power injections, are used as inputs.



Fig. 5. Mesh of QUENCH-16 experiment in MAAP4

As described in chapter 2, the bundle is firstly pre-oxidized under a steam atmosphere: this phase experimentally reproduces the accident phase during which the core is uncovered and the residual power leads to evaporation of residual water. The steam thus produced causes an exothermic oxidation reaction between cladding metal and steam resulting in hydrogen generation and possible temperature escalation. The interest in simulating this phase is justified by the need to have good initial conditions for the air ingress phase. To simulate this phase, the parabolic weight gain correlations of Cathcart and Urbanic are used, which are recommended for Zry-4 cladding oxidation in steam under severe accidents conditions [13,14]. The contact resistance to simulate the loss of power in the zones where the electrodes are connected to the power supply is put at 4.5 m $\Omega$  for the steam phase, near to the value described in [15].

The bundle is then oxidized in air atmosphere: this phase experimentally reproduces the air ingress consequently to the vessel failure. This will create a cladding oxidation, more exothermic than oxidation in steam. To simulate this phase, the parabolic weight gain correlations of NUREG [1] are used, which are considered as the currently best compromise for air oxidation modeling in MAAP despite its parabolic nature [17]. The contact resistance is put at 6.5 m $\Omega$  for the air phase, as it was previously assumed in [16] that this resistance can be increased during the transient.

#### 3.2. Main results of modeling

The overall QUENCH-16 transient was simulated with the MAAP4 code. The measured and calculated thermal behaviors are shown on Fig. 6.



Fig. 6. Temperatures evolution of the bundle for the experiment (in dashed lines) and for the simulation (in solid lines) at 250mm, 850mm and 1250mm of height (in red, green and black,

#### respectively)

During the steam pre-oxidation phase, one can see a good agreement between the experiment and the modeling for the overall bundle: the thermal behavior is well reproduced (Fig. 6). This observation is confirmed by the hydrogen production figures (Fig. 7). About 14g of hydrogen are produced during the experiment and about 13g for the calculation. The initial conditions to switch to the air phase are thus globally reached.

During air oxidation phase, the temperature calculation (Fig. 6) is in a good agreement with the experiment for the upper part of the bundle. On the other side, the temperature escalation that occurred in the middle part of the bundle, and to a lesser extent in the lower part, is under-estimated. Besides, oxygen starvation (Fig. 8) is predicted at 10360 s for the calculation against 10200 s for the experiment, respectively equivalent to an oxygen consumption of about 49 g vs 58 g for the experiment. The under-estimation of the temperature in the hot zone, which is directly linked to the under-estimation of the overall air phase and the exothermic nitriding phenomenon is not taken into account in the model when oxygen starvation happened. It is necessary to note that the oxidation due to the residual steam flow rate during the air phase is well reproduced by the calculation: 2.5 g of hydrogen is produced with MAAP4 against 1.3 g during the experiment.

As explained in chapter 2, the bottom reflood caused a significant temperature escalation with a high hydrogen production and a consistent nitrogen release (24g of the nitrogen consumed was released). The reasons were: 1) the intensive re-oxidation of the nitrides, which perhaps have acted as a trigger due to its exothermicity 2) intensive cladding metal oxidation by steam penetrated through the porous nitride layer and 3) the following oxidation of the formed melt. In addition to the under-estimation of the temperature increase during the air ingress phase, these phenomena are not simulated in MAAP, so that the calculation gives a too fast cooling without temperature excursion (Fig. 6).



Fig. 7: Hydrogen production for the experiment (in dashed lines) and for the simulation (in solid lines)



Fig. 8: Calculated oxygen flow rates during the air phase

# 4. Conclusions and perspectives

The conduct of the QUENCH-16 experiment, as a collaboration between modeling and experimental teams, is a success. The experimental conditions have been met as previously defined, namely low cladding pre-oxidation and a period of oxygen starvation during the air phase. The QUENCH-16 experiment constitutes a case for modeling nitrides formation and its impact on cladding degradation.

The simulation of the QUENCH-16 experiment with MAAP4 is satisfying for the steam phase. The simulation of the air phase shows the need of model improvements for the oxidationnitriding kinetics under oxygen starvation conditions in order to reproduce properly the thermal behavior of the bundle. Furthermore, MAAP showed no temperature escalation and increased hydrogen production, which were observed during the reflood: this can be directly related to the not modeled 1) intensive steam oxidation of nitrides and subjacent cladding metal as well and 2) oxidation of melt developed due to temperature escalation.

#### Acknowledgements

The KIT work is sponsored by the HGF Programme NUKLEAR and was performed in the framework of the European LACOMECO Project.

#### References

[1] D.A. Powers, L.N. Kmetyk, R.C. Schmidt: "A Review of The Technical Issues of Air Ingression During Severe Reactor Accidents", Technical report NUREG/CR-6218, Sandia National Laboratories, 1994.

[2] Z. Hózer, P. Windberg, I. Nagy, L. Maróti, L. Matus, M. Horváth, A. Pintér, M. Balaskó, A. Czitrovszky, P. Jani: "Interaction of failed fuel rods under air ingress conditions", Nuclear Technology, Volume 141, 2003, Page 244,

[3] G. Schanz, M. Heck, Z. Hozer, L. Matus, I. Nagy, L. Sepold, U. Stegmaier, M. Steinbrück, H. Steiner, J. Stuckert, P. Windberg: "Results of the QUENCH-10 experiment on air ingress". Wissenschaftliche Berichte, FZKA-7087, SAM-LACOMERA-D09, Karlsruhe, Mai 2006. http://bibliothek.fzk.de/zb/berichte/FZKA7087.pdf

[4] A. Kiselev, D. Ignatiev, V. Konstantinov, D. Soldatkin, V. Nalivaev, V. Semishkin: "Main results and conclusions of the VVER fuel assemblies tests under severe accident conditions in the large-scale PARAMETER test facility". 16th International QUENCH Workshop, Karlsruhe, 16-18 November, 2010, ISBN 978-3-923704-74-3.

[5] C. Duriez, T. Dupont, B. Schmets, F. Enoch: "Zircaloy-4 and M5<sup>®</sup> high temperature oxidation and

nitriding in air", Journal of Nuclear Materials, Volume 380, 2008, Pages 30–45. [6] C. Duriez, M. Steinbrück, D. Ohai, T. Meleg, J. Birchley, T. Haste: "Separate-effect tests on zirconium cladding degradation in air ingress situations", Nuclear Engineering and Design, Volume 239, 2009, Pages 244-253.

[7] M. Steinbrück: "Prototypical experiments relating to air oxidation of Zircaloy-4 at high temperatures", Journal of Nuclear Materials, Volume 392, 2009, Pages 531-544.

[8] M. Steinbrück, M. Böttcher: "Air oxidation of Zircaloy-4, M5® and ZIRLO™ cladding alloys at high temperatures" Journal of Nuclear Materials, Volume 414, 2011, Pages 276-285.

[9] A. Miassoedov: "LACOMECO project within the 7th EU FWP large scale experiments on core degradation, melt retention and containment behaviour", Proceedings of the 15<sup>th</sup> International QUENCH Workshop, Karlsruhe, November 3-5, 2009. CD-ROM, ISBN 978-3-923704-71-2.

[10] J. Stuckert, Z. Hózer, J. Moch, C. Rössger, U. Stegmaier, M. Steinbrück: "Experimental results of the QUENCH-16 bundle tests on air ingress, performed within the framework of the LACOMECO project", 17th International QUENCH Workshop, Karlsruhe, 22-24 November, 2011. ISBN 978-3-923704-77-4.

[11] J. Birchley, L. Fernandez Moguel, C. Bals, E. Beuzet, Z. Hozer, J. Stuckert: WP5.1: "Conduct and analytical support to air ingress experiment" Conference ERMSAR 2012, Cologne, Germany, 21-23 March 2012.

[12] F.Rahn: "Technical Foundation of Reactor Safety - Knowledge Base for Resolving Severe Accident Issues", Technical report 1020497, 2010, EPRI.

[13] R.E. Pawel, J.V. Cathcart: "The Kinetics of Oxidation of Zircaloy-4 in Steam at High Temperatures", Journal of Electrochemical Society, Volume 126, 1979, Pages 1105-1111.

[14] V.H. Urbanic, T.R. Heidrick: "High Temperature Oxidation of Zircaloy-2 and Zircaloy-4 in Steam", Journal of Nuclear Materials, Volume 75, 1978, Pages 251-261.

[15] J. Stuckert, M. Große, U. Stegmaier, M. Steinbrück: "Results of Severe Fuel Damage Experiment QUENCH-14 with Advanced Rod Cladding M5<sup>®</sup>, Scientific report KIT 7549, 2010. http://digbib.ubka.uni-karlsruhe.de/volltexte/1000018823

[16] T. Hollands, C. Bals, H. Austregesilo, W. Luther: "Post-test calculations of PARAMETER-SF4 using ATHLET-CD", 16<sup>th</sup> International QUENCH Workshop, Karlsruhe, 16-18 November, 2010. ISBN 978-3-923704-74-3.

[17] E. Beuzet, J-S. Lamy, A. Bretault and E. Simoni: "Modelling of Zry-4 cladding oxidation by air, under severe accident conditions using the MAAP4 code", Nuclear Engineering and Design, Volume 241, 2011, Pages 1217-1224.