Code Assessment Program for MELCOR 1.8.6

Author and Co-author(s)	Tim Haste and Jon Birchley
Institution	Paul Scherrer Institut
Address	5232 Villigen PSI
Tel., E-mail, Internet address	056 310 2764, tim.haste@psi.ch, http://www.psi.ch/
Duration of the Project	March 21, 2006 to February 2, 2009

ABSTRACT

The MELCOR code developed at Sandia National Laboratories for the USNRC is established in Switzerland as the preferred code for analysis of severe accident transients in light water reactors, from initiating event through to potential release of fission products to the environment. One area of international concern is that of air ingress, either into the reactor core during the late stages of an accident initiated at power, into the vessel during shutdown operation with the upper head removed, or to spent fuel in a storage pool or transport cask. This air ingress can lead to accelerated core degradation and enhanced release of fission products, especially the highly radiotoxic ruthenium. Assessment of the code models for oxidation of Zircaloy cladding has shown that the present treatments do not fully represent all the relevant physical processes, and cannot be guaranteed to be conservative under all circumstances. The present contract aims to develop and assess an improved model for Zircaloy/air oxidation, principally for invessel conditions. In the first few weeks of this project, a comprehensive database of available integral and separate-effects tests has been assembled, which will form a basis for this work. The main features of the data and the development/assessment programme are summarised here.

Das Computer Programm MELCOR, entwickelt von den Sandia National Laboratories für das USNRC, ist in der Schweiz etabliert als das bevorzugte Programm zur Berechnung von schweren Unfällen von Leichtwasserreaktoren vom auslösenden Ereignis bis hin zur möglichen Freisetzung von Spaltprodukten in die Umgebung. Ein internationales Anliegen ist die Betrachtung des Lufteinbruchs entweder in den Reaktorkern in der späten Phase des Kernschmelzens bei einem Reaktorunfall, in den Reaktordruckbehälter in der Shutdown Phase bei geöffnetem RDB-Deckel, oder in abgebrannte Brennelemente im Speicherbecken oder Transportbehälter. Ein solcher Lufteinbruch kann zu beschleunigter Brennelementzerstörung und erhöhter Freisetzung von Spaltprodukten führen, speziell von hoch radiotoxischem Ruthenium. Untersuchungen der Rechenmodelle für die Oxidation von Zirkaloy-Hüllrohren haben gezeigt, dass im Moment noch nicht alle relevanten physikalischen Prozesse berücksichtigt werden, und dass deshalb eine konservative Abschätzung noch nicht unter allen Randbedingungen garantiert werden kann. Der vorliegende Vertrag soll helfen, die Entwicklung und Ertüchtigung von verbesserten Zirkaloy-Oxidationsmodellen voranzutreiben, speziell für Konditionen im Reaktordruckbehälter. In den ersten Wochen dieses Projektes wurde eine aussagekräftige Datenbasis von verfügbaren Integral-Tests und Experimenten zur Untersuchungen spezieller Eigenschaften zusammengestellt um als Grundlage für dieses Projekt zu dienen. Die Hauptpunkte der Datenbasis sowie der Entwicklungs- und Ertüchtigungs-Vorhaben sind hier zusammengestellt.

Project Goals

The safety impacts of air ingress on nuclear fuel elements at high temperature have been studied for many years, in situations such as those in-vessel following hot-leg failure in a PWR severe accident with subsequent failure of the lower head [1], in-vessel in shutdown conditions with the lower head removed [1] and with loss of residual heat removal, and in spent fuel ponds after accidental loss of coolant water [2], [3]. The situation is kept under continual review [4], [5], [6]. The presence of air can lead to accelerated oxidation of the Zircaloy cladding compared to that in steam, owing to the faster kinetics, while the 85% higher heat of reaction drives this process further. Air ingress is typically associated with poor heat transfer; the combined effect of these factors can give rise to an increased rate of core degradation. In oxygen-starved conditions nitriding of the metal can occur, the resulting zirconium nitride is highly inflammable and indeed can detonate on re-introduction of oxygen, or steam as can occur during reflood. Furthermore, the exposure of uranium dioxide to air at elevated temperatures can lead to increased release of some fission products, notably the highly-radiotoxic ruthenium [7], [8]. These concerns have given rise to experimental and modelling studies covering major aspects of air ingress accidents; these are still ongoing, notably within the EU 6th Framework SARNET project [9], [10], and the International Source Term Programme (ISTP) [11], [12], in both of which PSI participates.

The MELCOR code [13] has been chosen in Switzerland as the major tool for analysis of severe accidents in light water reactors, from initiating events through to potential release of radionuclide fission products to the environment. This is supported by SCDAP-based codes [14], [15] for more detailed analysis of thermal hydraulics in the vessel and primary circuit, and treatment of core degradation phenomena. Assessment of the code at PSI has shown that while the modelling in MELCOR can capture many aspects of air ingress phenomena, there are important features that are not sufficiently well modelled. One of these is the kinetics of the highly exothermic oxidation of the cladding in air, which is a major driving force towards core degradation and fission product release. The final aim of the current three-year project is to develop and validate an improved Zircaloy/air oxidation model for MELCOR, taking into account the latest data from current experimental programmes, that is consistent with the engineering-level approach adopted in this code. The first stage of the work is collection of the relevant data to be followed by a critical evaluation of the database and identification of the dominant phenomena that need to be treated. In the second and third years, improved models will be developed on the basis of separate-effects data, introduced into MELCOR, and assessed using the results of independent integral experiments, thus completing the study. The end result will lead to improved predictability of core degradation and fission product release under air ingress conditions.

In the first reporting period the aim was to collect available data. The present report summarises the results of this exercise.

Work Carried Out and Results Obtained

Boundary Conditions

This study concentrates on air ingress conditions under in-vessel conditions, both for accidents initiated at-power, and under mid-loop shutdown conditions. A schematic of the conditions is given in Figure 1. The TMI-2 accident [16] showed non-uniform damage to the core; the central regions of the core may melt and slump while the outer regions remain largely intact. Following rupture of the reactor vessel by molten debris, air with maybe an admixture of gases from molten core/concrete interactions may be drawn from the containment into the vessel and react vigorously with these peripheral, largely intact rods. Current studies within the SARNET project [10], using a variety of tools for plant sequence analysis, have shown the possibility of air reaching declad fuel, that could lead to fuel oxidation and increased fission product release rates. Under mid-loop conditions with the upper head removed, air could recirculate over and into the fuel columns; if air enters a channel following development of parallel flow path instabilities and the oxygen is consumed by reaction with metal, a self-sustaining sequence could ensue as more air is drawn in.

The baseline for this study is the major review of Powers et al. [1] that considered accidents from power at shutdown, estimating boundary conditions such as mass flow into the vessel, composition of gases, temperatures, and the impact in terms of temperatures, core degradation and fission product release, notably ruthenium. Previous separate-effects tests were reviewed and analysed in terms of parabolic reaction kinetics, which

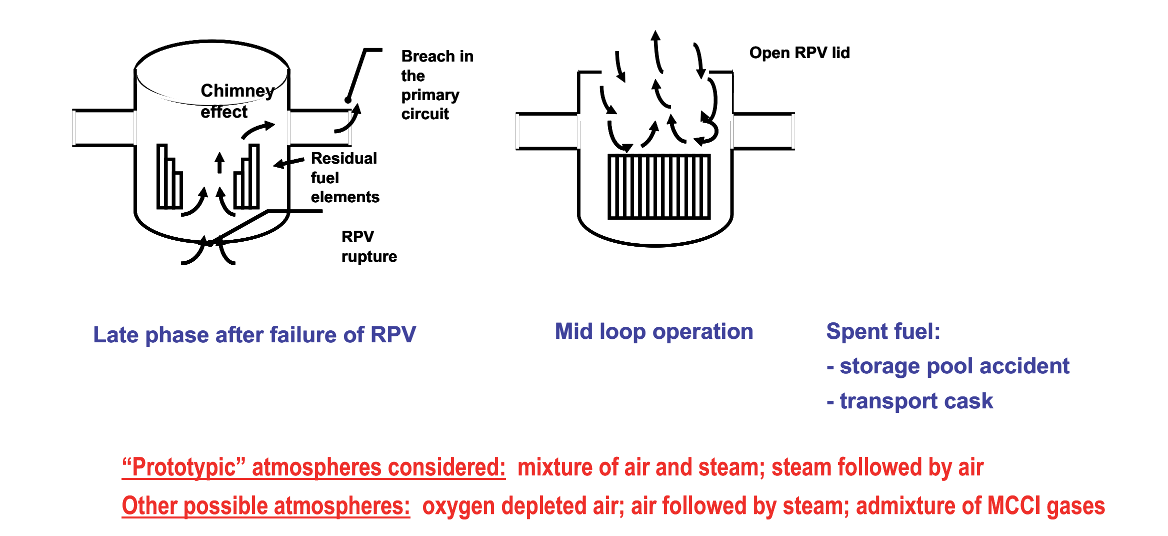

Figure 1: *Schematic for air ingress into the reactor vessel (Powers, NUREG/CR-6148; Steinbrück, ANS2006).*

assumed that the cladding remains intact and imposes and effective barrier to oxygen diffusing to the metal surface underneath; this is a simplification as weakening and breakaway of the oxide film can occur if nitrogen is present, for example see [17], [18], [19]. The review of G Schanz in the EU 4th Framework OPSA project report [20], shows that and post-breakaway oxidation follows linear kinetics. (Note: breakaway oxidation is also observed in steam alone, but only at larger oxide thicknesses; see for example [21].) Correlations were developed using parabolic kinetics, see Figure 2, based on the NUREG work [1] and modified in OPSA project to include data from the CODEX experiments (see below). The NUREG1 correlation consists of an upper and lower branch, joined by a line labelled NUREG2; the upper branch of NUREG1 forms the basis for the current model in MELCOR. It is seen that the NUREG correlation gives oxidation rates higher than typical for steam (Urbanic-Heidrick correlation [22]) at temperatures over 1000° C. A fit to CODEX-AIT data was consistent with the upper branch of NUREG1, while separate-effects tests performed at AEKI [23] in the framework of OPSA were consistent with NUREG2.

The sections below review the major separate-effects experiments performed since 1994 on Western Zr-based cladding materials. The main stimulus has been the need to improve quantitative knowledge of the phenomena so more accurate models can be formulated. Similar separate-effects work has been performed on VVER materials, with similar conclusions, but this is beyond the scope of the present study. Finally, the three integral experiments performed so far in the area are summarised.

Separate-effects Tests

Argonne National Laboratory Tests, USA

Air oxidation tests on Zircaloy-4, Zirlo and M5 cladding were performed at Argonne National Laboratories (ANL) [24], [25], [26] under the sponsorship of the USNRC, under conditions relevant to spent fuel pond accidents, and with an oxide layer representative of conditions of the inventory of spent fuel discharged after medium or high burnup. The oxide layer that forms on the cladding while in the pond was simulated by pre-oxidation in steam at 550° C, to 25-30 µm. Weight

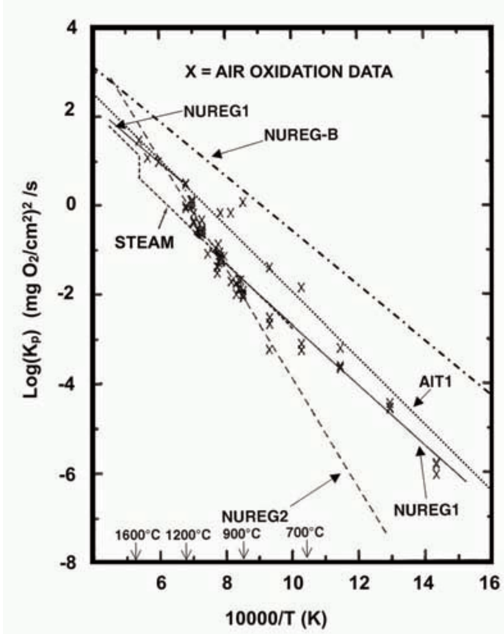

Figure 2: *Comparison of Zircaloy weight gain correlations in air (Powers, NUREG/CR-6148; Shepherd, EUR 19528EN).*

gain and oxide thickness measurements were made on samples oxidised in air in the temperature range 300-900° C, with emphasis on the range 300–600° C. The data showed two kinetic regimes at all temperatures over 400° C. Data were fitted by two parabolic lines to represent kinetics pre- and post-breakaway, for both weight gain and oxide thickness, for each material. The Zircaloy-4 data for 300–900° C showed oxidation rates slightly higher than extrapolation from the NUREG1 formulation and much higher than extrapolation from the NUREG2 or CODEX fits. Results were also obtained for Zirlo and M5, applicable to these advanced cladding materials under high burn-up conditions; the results for Zirlo were similar to those for Zircaloy-4, whereas M5 showed reduced oxidation rates. The further work on the effect of pre-oxidation and pre-hydriding on Zirlo oxidation in air showed that hydrogen concentration in the prototypic range 100–1000 wppm had minimal effect on the oxidation in the range 300–600° C. Full data reports are available [24], [25].

Forschungszentrum Karlsruhe Tests, Germany

A comprehensive test programme of separate-effects experiments has been performed over a number of years at FZK, in three different facilities: the resistance furnace BOX (2 cm long specimens), the inductive facility QUENCH-SR (15 cm specimens), and in a thermal balance facility TG (1 cm specimens). Current tests are performed within the SARNET framework. Most tests are conducted isothermally; temperature ranges are 1073 – 1873 K, thus representative of in-reactor conditions. Atmospheres considered are air, air/steam mixtures, air/nitrogen mixtures, and steam (or oxygen) followed by air (or nitrogen). Specimen temperatures, off-gas composition and mass change (TG) are measured on-line; destructive examinations determine reacted layer thicknesses, formation of nitride phases and weight gain.

The experiments demonstrate strong degradation of the oxide layer due to nitride formation, and that re-oxidation leads to early failure and loss of the barrier effect of the cladding. This is illustrated in Figure 3, for a test at 1200° C, where it is seen that nitriding in pure nitrogen proceeds slowly and parabolically, oxidation in oxygen (or steam) proceeds parabolically too (at least up to reasonable thicknesses) and at a higher rate, while the presence of both nitrogen and oxidation together leads to accelerated formation of oxide and also to increased nitriding compared with exposure in pure nitrogen, with the resulting scale being friable and non-protective. In this last case, there is a resulting rapid transition to linear kinetics. Oxygen if present is consumed in preference to steam. Pre-oxidation in steam prevents air attack as long as the scale is intact. It is noted that re-oxidation of ZrN formed during steam and oxygen starvation phases could cause temperature escalations in la-

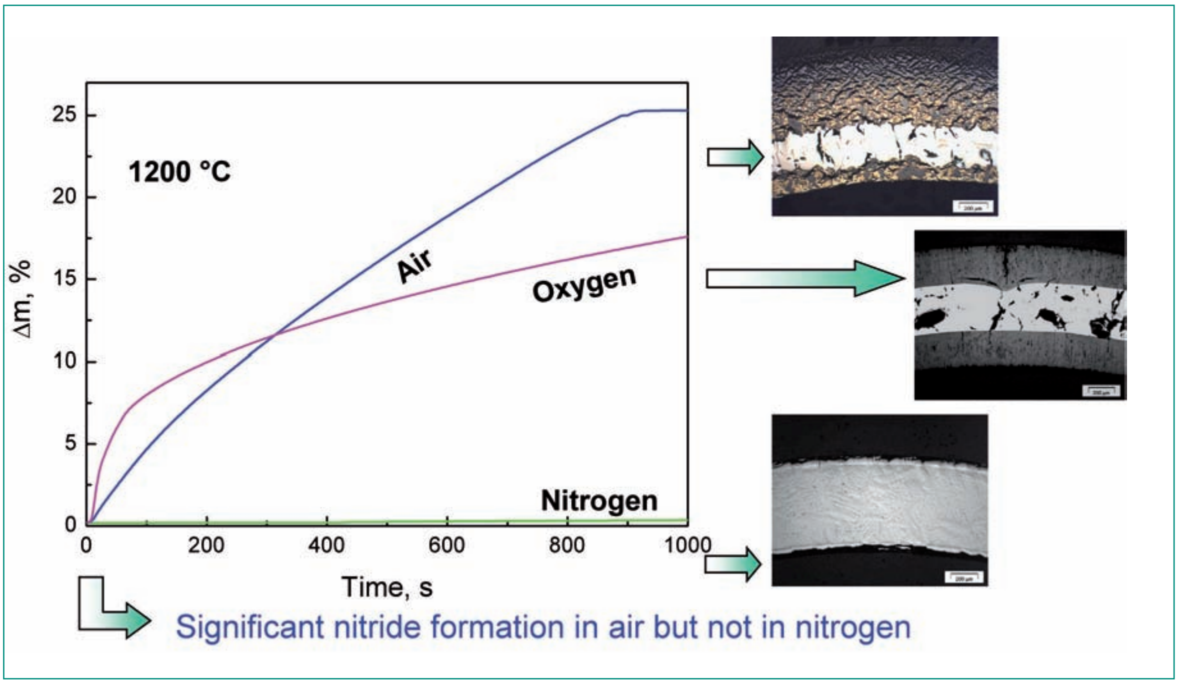

Figure 3: *Illustration of reaction rates of Zircaloy-4 in air, oxygen and nitrogen at 1200° C in FZK separate-effects tests (Steinbrück, ANS2006).*

ter phases. It was concluded that parabolic correlations for oxidation in air may only be applied at high temperatures (>1673 K) and for pre-oxidised cladding (>1373 K). Future tests will be directed towards model development.

IRSN MOZART Tests, France

A major series of air oxidation tests is being performed in the MOZART facility at IRSN Cadarache, France [31], as part of the International Source Term Programme (ISTP), to whose members access to the data and detailed conclusions is restricted. The programme also falls within the SARNET envelope. The facility is based on a thermobalance (ThermoGravimetric Analyser, TGA). Alloys investigated are Zircaloy-4, M5, and Zirlo, in a bare, steam pre-oxidised or pre-hydrided initial state. Double-sided isothermal tests on specimens of length 7, 10 and 20 mm long are performed in the temperature range 300 – 1200 °C, thus being more relevant to spent fuel pond conditions; transient tests with ramp rates 0.5 – 30 K/min are also conducted. A range of flow rates is used, and gas compositions of pure synthetic air, air/steam (to 0.34 bar partial pressure) are considered. Furnace and sample temperatures, and sample mass change, are recorded on-line, while post-test examinations provide information on microstructure.

Results from a typical test are shown in Figure 4. This illustrates the kinetic transition from parabolic to breakaway behaviour, for a test at 800° C, associated with cracking of the dense layer formed during the parabolic phase, and subsequent formation of porous oxide. At lower temperatures, a complex behaviour is often seen; here there is an initial high peak in oxidation rate. Future tests are planned on single-sided oxidation, also with

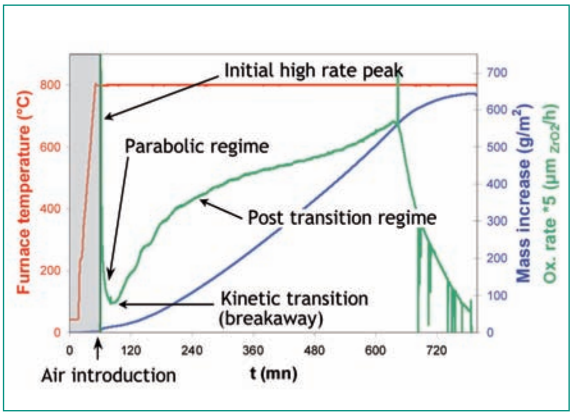

Figure 4: *Illustration of results from MOZART tests on air oxidation of Zircaloy cladding at 800° C, showing transition to breakaway regime (Duriez, priv. comm., 2006)*

extension to shorter duration tests for M5 cladding, and tests with more pre-oxidation on both for Zircaloy-4 and M5.

INR Pitesti Tests, Romania

Experiments are being carried out at INR [32] on Zircaloy-4 cladding of CANDU dimensions, under the auspices of SARNET, looking at oxidation in air, steam and air/steam mixtures. The experimental apparatus and technique, thermobalance (TGA), is very similar to that used in the MOZART programme, while the temperature range is 900-1400 °C, thus relevant to in-reactor air ingress conditions. Data reports are available so far only in Romanian, and have not yet been obtained, however a summary of the work is expected in the SARNET annual review meeting in February 2007.

Integral Tests

CODEX Tests, Hungary

The first integral tests to study air ingress were performed in the CODEX facility [33], [34] at the Atomergia Kutatotintezet (AEKI) institute, Budapest, Hungary, in 1998 and 1999 as part of the Oxidation Phenomena in Severe Accidents (OPSA) shared cost action of the EU 4th Framework programme. Two experiments were performed with electrically heated 9-rod pressurised-water type bundles with a heated length of 0.6m, containing unirradiated annular uranium dioxide fuel pellets. Instrumentation measured the major operational parameters such as power, flow rates, temperatures, water levels and pressures, while aerosol measurements were conducted on the exit gases in the offgas line. In test AIT1, pre-oxidation in an argon/25%oxygen mixture was intended to be performed at 900° C to give and maximum oxide layer thickness of 50 µm; however a temperature excursion took place to 1400° C so the maximum thickness would have been more than expected, possibly up to 1400 µm locally. After the pre-oxidation for 100s, stabilisation in argon at 900° C, air at room temperature was injected and the argon flow stopped, with the power being kept constant. The cold air cooled the lower part of the bundle, but led to a temperature excursion to about 2000° C in the upper part, at which point the test was terminated (after 500s of air injection) by shutting down the power and replacing the air with a high flow of cold argon. Strong oxidation was observed in the upper part of the bundle to a thickness of 520 µm, also with zirconium nitride for-

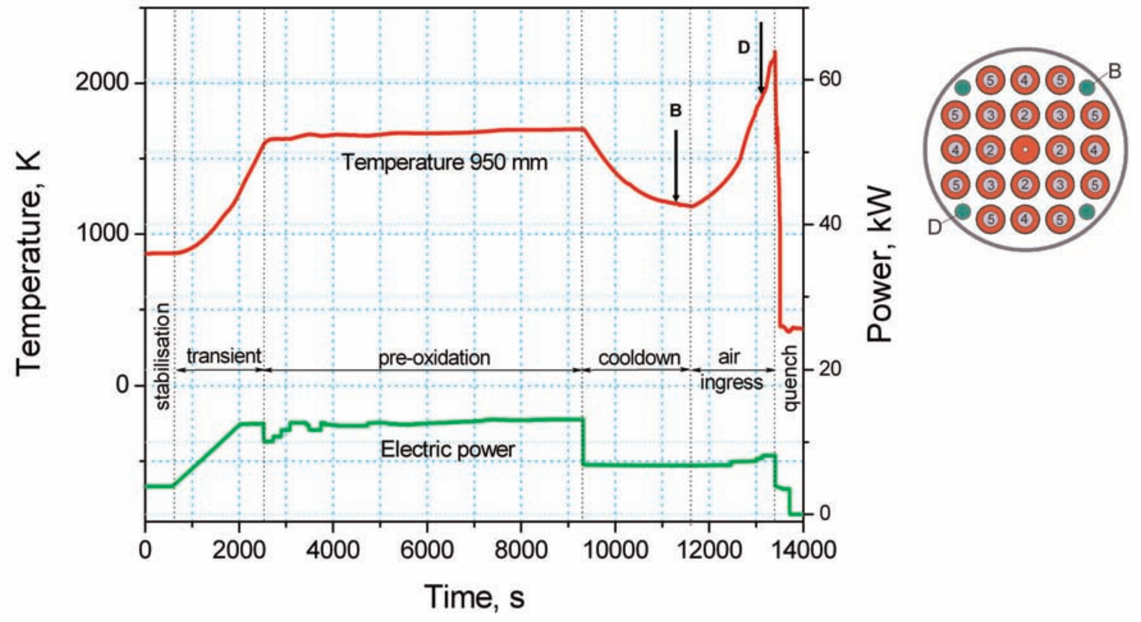

Figure 5: *Conduct of the QUENCH-10 experiment (Sepold, ANS 2006).*

mation up to 290 µm (indicating oxygen starvation in the upper part of the bundle). The original bundle geometry was largely preserved. Aerosol measurements indicated 0.1 to 0.5 µg release uranium release, as the dioxide.

In the second test AIT2, which was intended to lead to more bundle degradation and fuel release, pre-oxidation took place in steam with an accidental small admixture of air, for a total of 3600 s at 820° C and 900° C to give a maximum oxide thickness of 20 – 25 µm. Air at a lower flow rate than in AIT1 was injected starting at about 800° C with a linear power ramp. A temperature excursion was observed in the air injection phase up to a maximum indicated temperature of about 1900 °C; the test was stopped when temperatures reached 1700° C in the middle of the bundle, indicating relocation of molten Zircaloy cladding. Damage to the bundle was more severe than in AIT1, as high temperature conditions were maintained for a longer time, with fragmentation and relocation of debris. Maximum oxide and nitride thicknesses were 30 µm and 170 µm respectively. Much more uranium was released than in AIT1, 50 to 100 µm.

These tests indicated acceleration of oxidation phenomena and core degradation processes compared with those in similar tests in steam. Strong oxidation and nitriding led to mechanical degradation of the cladding. The high temperature at which the oxidation took place resulted in aerosol release, with some of the particles bearing uranium. However no higher oxides, for example the volatile U_{O3}, were detected; this could have been due to the oxygen reacting preferentially with the Zircaloy so none reached the surface of the hot UO_2. While the results were used for some code assessment within OPSA, the results have only a semi-quantitative nature, owing for example to imprecision in the boundary conditions, and therefore further integral testing under well-controlled conditions was necessary. Full data reports are available [35], [36] , along with electronic copies of the data files.

QUENCH Test, Germany

One bundle air ingress test, QUENCH-10 [37], [38], has so far been performed (in July 2004) at the QUENCH facility in the Forschungszentrum Karlsruhe, Germany. The aim was to investigate the effect of air ingress on fuel rods heavily pre-oxidised in steam. It was proposed by AEKI under the EU 5th Framework LACOMERA programme, and supported calculationally by PSI [39] using MELCOR and SCDAP-based codes (modified locally for air oxidation) and IRSN. The facility is mainly designed to investigate the hydrogen source term that results from water injection into a light water reactor (LWR) core, using Zircaloy-clad rods containing zirconia pellets, powered electrically with tungsten heaters of heated length 1m. In PWR tests, a 21-rod bundle is normally used; solid rods on the periphery of the bundle can be withdrawn during the test to check on the oxidation achieved at any stage. The facility is comprehensively instrumented, with measurements of pressure, temperature, gas flow rate etc., and mass spectrometer instrumentation in the offgas line enables detailed measurements of the exit gas composition. In the QUENCH-10 test, aerosol measurements in the offgas stream were made by members of the CODEX team from AEKI.

The test conduct, Figure 5, involved pre-oxidation in argon/steam at 1620-1690 K for 113 min, then a cooling phase lasting 38 min to 1190 K to prepare the conditions for air ingress, and then the steam flow was replaced by air at one-third of the mass flow rate. The duration of this phase was 30 min. Complete consumption of oxygen and partial consumption of nitrogen were observed towards the end of the temperature excursion in this phase. The test was terminated by reducing the electrical power and reflooding with water at room temperature at a high rate, after peak temperatures reached about 2200 K. Cooling was established very quickly; with compete quenching after 100 – 150s. A small amount of hydrogen was measured during the quench phase (but no temperature peak, in contrast to some other tests), and also some nitrogen coming from oxidation of nitride. Corner rods withdrawn indicated oxidation to 514 µm before air ingress and 610 µm before reflooding. Post-test examination revealed severe damage to the bundle and its insulation, Zircaloy-lined, shroud. As in the CODEX tests, the influence of air on bundle degradation was clear, and it was observed also that under oxygen starvation conditions the oxide scale previously formed under steam oxidation is severely attacked in pure nitrogen. Comprehensive data were obtained, available in electronic form, and these with the data report [37] form a good basis for code assessment. A limited post-test analysis has been performed by PSI using MELCOR and SCDAP [40]. Good agreement was observed with the experimental data in the steam phase; however the reference case that used the standard MELCOR correlation for air oxidation, NUREG1, overpredicted the oxidation rate in the air phase. The calculated temperatures are compared with data in Figure 6. The results at 350 mm, where temperatures are too low for oxidation, are in fair agreement and show that the reduced heat transfer in air is well reproduced. However at 550 mm gross overcalculation of the oxidation rate leads to a rapid excursion not seen in the test, to the extent that upper regions are oxygen-starved early. There is similar but less extreme overcalculation at the top of the heated section, 850 – 950 mm. The overcalculation is illustrated again in Figure 7, which shows calculation of the oxygen mass flow leaving the test section compared with the data. Using steam kinetics, the oxidation rate is underpredicted, as expected, while all the air correlations overpredict. The explanation is that the air oxidation models implicitly assume that the oxide layer is only weakly protective against further oxidation throughout. In QUENCH-10 the layer formed in steam was protective for a considerable portion of the air ingress phase. The best fit is obtained by using steam correlation up to 1400 K and a reduced air correlation up to 1500 K, with interpolation between. This agreement should not be used as the basis for a model but demonstrates a «fading memory» effect of an oxi-

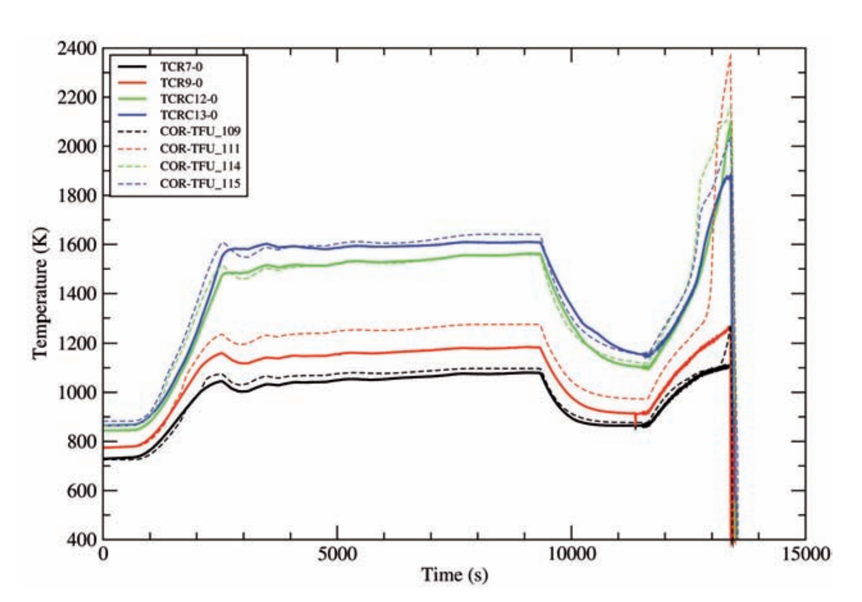

Figure 6: *MELCOR1.8.5 calculation of QUENCH-10 using standard air oxidation model, showing difference between model and data in the air phase.*

Key:
TCR7,9,12,13 = data at 350,550,850,950mm;
COR-TFU_109 to115 = MELCOR calculated temperatures at the corresponding positions
(Birchley and Haste, 10th QUENCH Workshop)

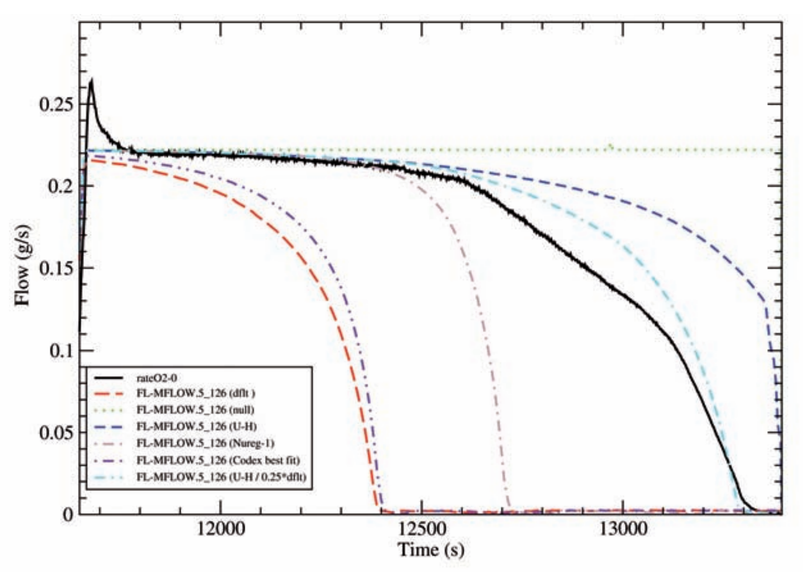

Figure 7: *Sensitivity studies with MELCOR1.8.5 on QUENCH-10 results - effect of oxidation correlation for air.*

Key:
rateO2=data;
dflt=MELCOR default air correlation;
null=no oxidation;
U-H=Urbanic-Heidrick for steam;
Nureg1=air correlation of NUREG/CR-6218, upper branch;
Codex=best fit to CODEX-AIT1 data;
*U-H/0.25*dflt=Urbanic-Heidrick to 1400K, Nureg1*0.25 from 1500K*
(Birchley and Haste, 10th QUENCH Workshop)

de layer previously formed in steam on subsequent oxidation in air. This important effect must be considered in future model development. Separate-effects tests will provide data relevant to such a need.

Other tests

A further air ingress test the QUENCH facility has been discussed; this would be valuable for independent assessment of new models for air oxidation of Zircaloy. An air ingress bundle test FPT5 had formed part of the Phebus FP in-reactor experimental programme [41], but this was removed and the separate-effects tests in ISTP are to some extent a substitute for this. A follow-up Phebus programme on severe accident phenomena is under discussion, with air ingress one of the issues being considered.

National Cooperation

This project does not involve cooperation with other Swiss projects.

International Cooperation

Cooperation with organisations within European countries and Canada generally is performed under the auspices of the SARNET (Severe Accident Research NETwork) Network of Excellence in the EU 6th Framework programme «Nuclear Fission: Safety of Existing Nuclear Installations», under contract number FI6O-CT-2004-509065. This includes access to the data from INR Pitesti, Romania, and Forschungszentrum Karlsruhe, Germany. Access to data from the MOZART programme of separate-effects tests at IRSN Cadarache, France, is obtained through PSI membership of the International Source Term Programme (IRSN-PSI contract dated 28.3.2006). The MELCOR code and early access to the results of USNRC programmes is obtained under the Cooperative Severe Accidents Research Programme Agreement (CSARP) between HSK and USNRC, and close contact is kept with the MELCOR developers at Sandia National Laboratories (SNL) regarding code maintenance, development and use. PSI obtains the SCDAPSIM code, maintenance and user support via a licence agreement with Innovative Software Services (ISS), Idaho Falls, USA. SCDAPSIM is a derivative of SCDAP/RELAP5 formerly supported by the USNRC.

Assessment 2006 and Perspectives for 2007

The project has made a successful start in collecting relevant experimental data from past and present, and has established contact with the current experimental teams so that up-to-date knowledge will continue to be obtained for the duration of the work. The opportunity will be taken to discuss the upcoming test schedules cooperatively with the experimenters, in the SARNET framework, to optimise the chance of obtaining results best suited to modelling needs. The next stage of the work, to be completed by mid-2007, is the production of a detailed review of the data that is intended to draw out the main features of the results obtained, to act as a basis for the model development that is planned for the rest of the year. The project is proceeding according to the timescale foreseen.

References

[1] *D. A. Powers, L. N. Kmetyk and R. C. Schmidt,* A Review of Technical Issues of Air Ingression during Severe Reactor Accidents, USNRC NUREG/CR-6218, SAND94-0731, Sandia National Laboratories, September1994.

[2] *A. S. Benjamin, D. J. McCloskey, D. A. Powers and S. A. Dupree,* Spent Fuel Heatup Following Loss of Water during Storage, NUREG/CR-0649, SAND77-1371, Sandia National Laboratories, March 1979.

[3] *V. L. Sailor, K. R. Perkins and J. R. Weeks,* Severe Accidents in Spent Fuel Pools in Support of Generic Issue 82, USNRC BNL-NUREG-52093, Brookhaven National Laboratories, July 1987.

[4] *D. A. Powers,* Technical Issues Associated with Air Ingression during Core Degradation, 4th Technical Seminar on the Phebus FP Programme, Marseille, March 2000.

[5] *USNRC,* Technical Study of Spent Fuel Pool Accident Risk at Decommissioning Nuclear Power Plants, NUREG-1738, February 2001.

[6] *P. Giordano and C. Séropian,* The Air Ingress Issue for PWRs, 5th Technical Seminar on the Phebus FP Programme, Aix-en-Provence, France, June 2003.

[7] *G. W. Parker, C. J. Barton, G. E. Creek, W. J. Martin and R. A. Lorenz,* Out of Pile Studies of Fission-Product Release from Overheated Reactor Fuels at ORNL, 1955 – 1965, ORNL-3981, Oak Ridge National Laboratory, Oak Ridge, TN, July 1967.

[8] *F. C. Iglesias, C. E. L. Hunt, F. Garisto and D. S. Cox,* Measured Release Kinetics of Ruthenium from Uranium Oxides in Air, Proc. of Int. Seminar on Fission Product Transport Processes during Reactor Accidents, Editor J. T. Rogers, Hemisphere Publishing Corp, Washington DC, 1990.

[9] *J.-C. Micaelli et al.,* SARNET – A European Cooperative Effort on LWR Severe Accident Research, Revue Générale Nucléaire 2006, no. 1, January – February 2006.

[10] *A. Auvinen et al.,* On-going Investigations of Ruthenium Release and Transport, European Review Meeting on Severe Accident Research (ERMSAR-2005), Aix-en-Provence, France, 14 – 16 November 2005.

[11] *B. Clément,* Towards Reducing the Uncertainties on Source Term Evaluations: an IRSN/CEA/EDF R&D Programme, Proc. EUROSAFE Forum, Berlin, 4–5 November 2002.

[12] *B. Clément and R. Zeyen,* The Phebus Fission Product and Source Term International Programmes, Proc. Int. Conf. on Nuclear Energy in New Europe 2005, Bled, Slovenia, September 5 – 8, 2005.

[13] *R. O. Gauntt et al.,* MELCOR Code Manuals – Version 1.8.5, USNRC NUREG/CR 6119 Rev. 2, SAND2000-2417, Sandia National Laboratories, October 2000.

[14] *L. Siefken et al.,* SCDAP/RELAP5/MOD3.2 Code Manual, USNRC NUREG/CR-6150 Rev. 1, INEL-96/0422 Rev. 1, Idaho Falls National Engineering Laboratories, November 1997.

[15] *Innovative Systems Software,* RELAP/SCDAPSIM/MOD3.4 Code Manual, Idaho Falls, USA, 2003.

[16] *J. M. Broughton, P. Kuan, D. A. Petti and E. L. Tolman,* A Scenario of the Three Mile island Unit 2 Accident, Nuclear Technology, vol. 87, pp 34–53, 1989.

[17] *E. B. Evans et al.,* Critical Role of Nitrogen during High Temperature Scaling of Zirconium, Proc. Conf. Met. Soc. AIME, Boston, USA, pp 248–281, 1972.

[18] *C. J. Rosa and W. W. Smeltzer,* The Oxidation of Zirconium in Oxygen/Nitrogen Atmospheres, Z. Metallkunde, vol. 71 pp 470–474, 1980.

[19] *S. Leistikow and H. v. Berg,* Investigation under Nuclear Safety Aspects of Zry-4 Oxidation Kinetics at High Temperatures in Air, 2nd Workshop of German and Polish Research on High Temperature Corrosion of Metals, Jülich, Germany, Proc. Jül. Conf. 76, 114 – 132, December 1987.

[20] *I. Shepherd et al.,* Oxidation Phenomena in Severe Accidents (OPSA) – Final Report, INV-OPSA(99)-P008, EUR 19528 EN, 2000.

[21] *S. Leistikow, G. Schanz, H. v. Berg and A. E. Aly,* Comprehensive Presentation of Extended Zircaloy-4 Steam Oxidation Results 600–1600° C, OECD/NEA-CSNI/IAEA meeting, Risø, Denmark, May 1983, IAEA Summary Report IWGFPT/16, 1983.

[22] *V. H. Urbanic and T. R. Heidrick,* High-Temperature Oxidation of Zircaloy-2 and Zircaloy-4 in Steam, J. Nucl. Mat. Vol. 75, pp251-261, 1978.

[23] *L. Maróti,* How to Conduct the Air Ingress Test, in

I. Shepherd et al., Investigation of Core Degradation (COBE) and Oxidation Phenomena in Severe Accidents (OPSA): Specialist Meeting on Thermocouples, Karlsruhe, 5 March 1997, JRC Ispra Technical Note I.97.78, May 1997.

[24] *K. Natesan and W. K. Soppet,* Air Oxidation Kinetics for Zr-based Alloys, USNRC NUREG/CR-5846, ANL-03/32, June 2004.

[25] *K. Natesan and W. K. Soppet,* Hydrogen Effects on Air Oxidation of Zirlo Alloy, USNRC NUREG/CR-6851, ANL-04/14, October 2004.

[26] *USNRC,* Kinetics of Cladding Oxidation in an Air Environment, Research Information Letter (RIL) 0402, downloaded from ADAMS, http://www.nrc.gov/reading-rm/adams.html, August 2006.

[27] *M. Steinbrück et al.,* Experiments on Air Ingress during Severe Accidents in LWRs, Nucl. Eng. Design, 236, pp 1709–1719, 2006.

[28] *M. Steinbrück,* Prototypical Experiments on Air Oxidation of Zircaloy Cladding, 2006 Annual Meeting of the American Nuclear Society, Reno, Nevada, 4 – 8 June 2006.

[29] *M. Steinbrück, U. Stegmaier and T. Ziegler,* Prototypical Experiments on Air Oxidation of Zircaloy-4 at High Temperature, Forschungszentrum Karlsruhe, FZKA 7257, in publication, 2006.

[30] *M. Steinbrück, U. Stegmaier and N. Vér,* Air Oxidation at High Temperatures: Update, 12th International QUENCH Workshop, Forschungszentrum Karlsruhe, Germany, October 2006.

[31] *Ch. Duriez,* Institut de Radioprotection et de Sûreté Nucléaire (IRSN) Cadarache, private communication to T Haste, PSI, December 2006.

[32] *D. Ohai,* Institute of Nuclear Research (INR) Pitesti, private communication to T Haste, PSI, December 2006.

[33] *Z. Hózer, P. Windberg, I. Nagy, L. Maróti, L. Matus, M. Horváth, A. Pintér, M. Balaskó, A. Czitrovszky and P. Jani,* Interaction of Failed Fuel Rods under Air Ingress Conditions, Nucl. Technology, vol. 141, pp 244 – 256, 2003.

[34] *A. Pintér Czordas et al.,* Investigations of Aerosols Released at High Temperatures from Nuclear Reactor Core Models, J. Nucl. Materials, vol. 282, pp 205 – 215, 2000.

[35] *Z. Hózer, P. Windberg, I. Nagy, L. Maróti, L. Matus, M. Horváth, A. Pintér, A. Czitrovszky and P. Jani,* CODEX-AIT-1 Experiment: Core Degradation Test under Air Ingress, AEKI Budapest, KFKI-2002-02/G, 2002.

[36] *Z. Hózer, P. Windberg, I. Nagy, L. Maróti, L. Matus, M. Horváth, A. Pintér, Á. Griger, M. Balaskó, B. Alföldy, A. Czitrovszky and P. Jani,* CODEX-AIT-2 Experiment: Core Degradation Test with Steam Oxidation and Air Ingress, AEKI Budapest, KFKI-2002-03/G, 2002.

[37] *G. Schanz et al.,* Results of the QUENCH-10 Experiment on Air Ingress, Forschungszentrum Karlsruhe, FZKA 7087, May 2006.

[38] *L. Sepold, Z. Hózer, A. Miassoedov, G. Schanz, U. Stegmaier, L. Steinbock, M. Steinbrück and J. Stuckert,* A QUENCH Experiment Including an Air Ingress Phase Prior to Flooding, 2006 Annual Meeting of the American Nuclear Society, Reno, Nevada, 4 – 8 June 2006.

[39] *Ch. Homann, W. Hering, J. Birchley and T. Haste,* Analytical Support for the Preparation of the Bundle Test QUENCH-10 on Air-Ingress, Forschungszentrum Karlsruhe, FZKA 7086, July 2005.

[40] *J. Birchley and T. Haste,* Post-Test Analysis of QUENCH-10 with SCDAP/RELAP5 and MELCOR, 10th International QUENCH Workshop, Forschungszentrum Karlsruhe, Germany, October 2004.

[41] *B. Clément, Ch. Ktorza, A. V. Jones and K. Trambauer,* The Phebus Fission Products Programme: Experiments and Evaluation, Proc. EUROSAFE Forum, Paris, 18–19 November 2000.

Acknowledgements

The authors gratefully acknowledge M Steinbrück of Forschungszentrum Karlsruhe (FZK), Germany, Ch Duriez of the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) Cadarache, and D Ohai of the Institute of Nuclear Research (INR), Pitesti, Romania, for providing valuable information on the status of their experimental programmes on air ingress, and for giving permission for their illustrations to be used in this report.