Aerosol and Iodine Issues, and Hydrogen Mitigation under Accidental Conditions in Water-cooled Reactors

Thermal–hydraulics, Hydrogen, Aerosols and Iodine (THAI–2) Project

Final Report

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NUCLEAR ENERGY AGENCY
COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

Aerosol and Iodine Issues and Hydrogen Mitigation under Accidental Conditions in Water Cooled Reactors

Thermal-hydraulics, Hydrogen, Aerosols and Iodine-2 (THAI2) Project
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The OECD member countries are: Australia, Austria, Belgium, Canada, Chile, the Czech Republic, Denmark, Estonia, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Israel, Italy, Japan, Korea, Latvia, Luxembourg, Mexico, the Netherlands, New Zealand, Norway, Poland, Portugal, the Slovak Republic, Slovenia, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The European Commission takes part in the work of the OECD.

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NUCLEAR ENERGY AGENCY

The OECD Nuclear Energy Agency (NEA) was established on 1 February 1958. Current NEA membership consists of 31 countries: Australia, Austria, Belgium, Canada, the Czech Republic, Denmark, Finland, France, Germany, Greece, Hungary, Iceland, Ireland, Italy, Japan, Korea Luxembourg, Mexico, the Netherlands, Norway, Poland, Portugal, Russia, the Slovak Republic, Slovenia, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States. The European Commission also takes part in the work of the Agency.

The mission of the NEA is:

– to assist its member countries in maintaining and further developing, through international co-operation, the scientific, technological and legal bases required for a safe, environmentally friendly and economical use of nuclear energy for peaceful purposes;

– to provide authoritative assessments and to forge common understandings on key issues, as input to government decisions on nuclear energy policy and to broader OECD policy analyses in areas such as energy and sustainable development.

Specific areas of competence of the NEA include the safety and regulation of nuclear activities, radioactive waste management, radiological protection, nuclear science, economic and technical analyses of the nuclear fuel cycle, nuclear law and liability, and public information. The NEA Data Bank provides nuclear data and computer program services for participating countries.

THE COMMITTEE ON THE SAFETY OF NUCLEAR INSTALLATIONS

The NEA Committee on the Safety of Nuclear Installations (CSNI) is an international committee made up of senior scientists and engineers with broad responsibilities for safety technology and research programmes, as well as representatives from regulatory authorities. It was created in 1973 to develop and co-ordinate the activities of the NEA concerning the technical aspects of the design, construction and operation of nuclear installations insofar as they affect the safety of such installations.

The committee's purpose is to foster international co-operation in nuclear safety among NEA member countries. The main tasks of the CSNI are to exchange technical information and to promote collaboration between research, development, engineering and regulatory organisations; to review operating experience and the state of knowledge on selected topics of nuclear safety technology and safety assessment; to initiate and conduct programmes to overcome discrepancies, develop improvements and reach consensus on technical issues; and to promote the co-ordination of work that serves to maintain competence in nuclear safety matters, including the establishment of joint undertakings.

The priority of the committee is on the safety of nuclear installations and the design and construction of new reactors and installations. For advanced reactor designs, the committee provides a forum for improving safety-related knowledge and a vehicle for joint research.

In implementing its programme, the CSNI establishes co-operative mechanisms with the NEA's Committee on Nuclear Regulatory Activities (CNRA), which is responsible for the Agency's programme concerning the regulation, licensing and inspection of nuclear installations with regard to safety. It also co-operates with the other NEA Standing Technical Committees as well as with key international organisations such as the International Atomic Energy Agency (IAEA) on matters of common interest.

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EXECUTIVE SUMMARY

Background

Progress in nuclear reactor containment safety research has led to a deeper understanding of phenomena and processes that may occur during a severe accident. However, a number of safety-relevant gaps in this understanding exist due to limitations in the experimental databases. Such gaps are − among others − related to combustible hydrogen and to the behaviour of fission products, in particular iodine and aerosols.

In the case of hydrogen, uncertainties arise mainly in the behaviour of deflagration flames, e.g. in the presence of water spray, and in the performance of Passive Autocatalytic Recombiners (PAR) under oxygen starvation conditions including PAR-induced ignition. The relevance to reactor safety is the destructive potential of deflagrations with the peak pressures and high temperatures involved.

In the case of airborne fission products, interaction processes of molecular iodine with reactive and non-reactive aerosol particles have not yet been investigated under accident-typical conditions to assess their effect on a potential source term. Another important challenge related to iodine behaviour was to quantify the release of gaseous iodine from a flashing jet, representing a PWR Design Basis Accident scenario (steam-generator-tube rupture during reactor shutdown sequence).

At an international level, large efforts have been undertaken for the evaluation and further development of Lumped Parameter (LP) and Computational Fluid Dynamics (CFD) codes for the analysis and simulation of severe accidents. However, the validity of these LP or CFD codes has to be assessed against technical and large-scale experiments representative for severe accident conditions. The need for such experiments in the frame of an international programme is twofold; first, to enhance the confidence in the performance of passive mitigation systems during severe accident scenarios and second, to establish a common database accessible by a large research community to support further development and validation of LP and CFD codes.

Phase 2 of the NEA Thermal-hydraulics, Hydrogen, Aerosols and Iodine (THAI) project was started in August 2011 under the auspices of the OECD Nuclear Energy Agency (NEA) to fill the above mentioned knowledge gaps by delivering suitable data for evaluation and simulation of hydrogen and fission product interactions. The THAI-2 project was supported by the signatories – safety organisations and research laboratories from eleven countries, listed in Table 0-1 - and received contributions from the partners listed in Table 0-2.

Table 0-1: THAI-2 signatories with their national origin

Signatories	Country
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH	Germany
Atomic Energy of Canada Ltd (AECL)	Canada
Nuclear Research Institute of the Czech Republic	Czech Republic
Valtion Teknologian Tutkimuskeskus (VTT) and Säteilyturvakeskus (STUK)	Finland
Institut de Radioprotection et de Sûreté Nucléaire (IRSN)	France
Nuclear Safety Research Institute (NUBIKI) and Paks nuclear power plant	Hungary
Japan Atomic Energy Agency (JAEA)	Japan
Korea Atomic Energy Research Institute	Korea
Ministry of Infrastructure and the Environment	Netherlands
Swedish Radiation Safety Authority (SSM)	Sweden
Office for Nuclear Regulation (ONR), agency of the health and Safety Executive (HSE)	United Kingdom

Table 0-2: German partners in the THAI-2 Project

Project partners
Becker Technologies GmbH (Operating Agent)
AREVA GmbH Erlangen
Siempelkamp NIS Ingenieurgesellschaft mbH in Alzenau
RWTH Universität Aachen
Forschungszentrum Jülich
RUB Ruhr-Universität Bochum
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH

Objective of the work

The overall objective of the THAI-2 project is to address open questions concerning the behaviour of hydrogen, iodine and aerosols in the containment of water-cooled reactors during design-basis or severe accident typical conditions. The understanding of the processes taking place during such events is essential for evaluating the challenge posed on containment integrity (hydrogen) and for evaluating the amount of airborne radioactivity (iodine and aerosols), which may be present inside a containment during such severe accidents with core damage. The project is to generate valuable data for evaluating the hydrogen deflagration behaviour during spray operation and its effective removal by PARs. Concerning fission products, the project will focus on the interaction of molecular iodine with non-reactive and reactive aerosol particles. Another important issue related to iodine behaviour is to quantify the release of gaseous iodine from a flashing jet, representing a PWR Design Basis Accident scenario (steam generator tube rupture during reactor shutdown sequence). An extensive analytical effort will accompany the experimental program consisting of code calculations to support test design, for pre-test assessments, result evaluation and extrapolation to reactor conditions.

The project contributes to the validation and further development of advanced LP- and CFD codes used for reactor applications by e. g. providing experimental data for code benchmark exercises. Furthermore, the project is aimed at maintaining competence and expertise in the field of reactor safety in the NEA member countries and promotes international co-operation between its member countries.

Work performed

The tests have been performed in the technical-scale THAI containment test facility (a 60 m³ stainless steel vessel 9.2 m high and 3.2 m in diameter) with exchangeable internals for multi-compartment investigations. The vessel is designed for a maximum overpressure of 1.4 MPa at 180°C and can withstand moderate hydrogen deflagrations. The facility is operated by Becker Technologies GmbH at Eschborn, Germany, under the sponsorship of the German Federal Ministry for Economic Affairs and Energy.

The facility is equipped with the necessary supply systems for steam, compressed air, nitrogen, oxygen, light gases (helium and hydrogen), aerosols and gaseous iodine, the latter also radioactive tracered. The vessel walls can be heated and/or cooled to establish the desired thermal hydraulic conditions.

An advanced instrumentation and data acquisition system is installed to monitor the relevant physical quantities with high spatial and time-related resolution.

Within the THAI-2 Project, the following test series have been performed:

- Hydrogen Deflagration Tests with Spray (HD)

- Hydrogen Passive Autocatalytic Recombiner Tests (HR)

- Interaction of Molecular Iodine with Aerosols

- Release of gaseous Iodine from a flashing jet

- Laboratory scale experiments on the Interaction of Iodine and Aerosols

In addition to the experimental work, an analytical workgroup was established in the frame of the THAI-2 Project, aimed at the evaluation of the test results for further development and validation of the predictive capabilities of advanced LP codes and CFD codes currently in use in the reactor safety field. For this purpose, experimental data of various tests has been selected for blind and open post-test calculations in the frame of benchmark exercises.

Results and their significance

Hydrogen deflagration tests with spray

With the hydrogen deflagration experiments performed, additional data for an improved understanding of hydrogen combustion phenomena in combination with the use of spray systems are obtained. For the HD-tests, by systematic variation of the test parameters, e.g. initial gas temperature, steam content, spray water temperature, spray water droplet size, spray duration before ignition, and burn direction, their influence on pressure build-up, temperature development, flame front propagation and completeness of combustion was quantified. For all tests with upward burn direction, spray operation resulted in lower pressures of the combustion compared to the reference tests without spray performed in the previous THAI project. In case of downward burn direction, the spray induced turbulence increased flame speed, resulting in a slightly higher peak pressure of the combustion compared to the reference test without spray. The data of selected tests have been used for open post-test calculations for the validation of combustion models.

Hydrogen passive autocatalytic recombiner tests

The HR tests are performed in a similar manner as the previous HR tests conducted in the frame of THAI project, while the objective was to test the PAR performance and ignition potential in oxygen-lean atmosphere. The PARs used for the tests are from AREVA and NIS. Three additional tests have been performed with NIS-PAR, which have been sponsored by the PAR vendor Siempelkamp NIS Ingenieurgesellschaft mbH in Alzenau, Germany. The PAR behaviour under different initial

conditions with ambient/superheated/saturated steam atmosphere and elevated initial pressure and temperature in the test vessel has been investigated. The data obtained from the HR experiments for onset of hydrogen recombination, hydrogen recombination rate, hydrogen depletion efficiency and ignition potential under oxygen-lean conditions provide additional insight on PAR behaviour under accident-typical thermal hydraulic conditions.

The AREVA-PAR started operation at very low oxygen concentrations (O_2 concentration at PAR inlet < 0.5 vol %) far below an oxygen surplus ratio of $\Phi = 1$. Recombination rate increases with increasing pressure (for the same hydrogen and oxygen concentration at the PAR inlet). The hydrogen depletion efficiency is almost independent of the steam content and increases by increasing oxygen surplus ratio. It remains constant once an oxygen surplus ratio of $\Phi = 2$ has been exceeded.

Ignition was initiated at PAR outlet and occurred for an oxygen starvation ratio as low as $\phi = 1.3$ and 18 vol % steam concentration, and a dilution ratio δ ($\delta = c_{O2}/(c_{O2} + c_{N2})$) at PAR inlet of $\delta = 0.08$, which also characterises the oxygen starvation conditions. For a steam concentration of 30 vol % and a similar dilution ratio δ, no ignition was observed.

The experimental data of AREVA PAR test HR-35 has been used for blind and open post-test code calculations in the frame of a benchmark exercise.

The NIS-PAR started operation at very low oxygen concentrations (O_2 concentration at PAR inlet < 0.5 vol %). Recombination rate increases with increasing pressure (for the same hydrogen and oxygen concentration at the PAR inlet). The hydrogen depletion efficiency is almost independent of the steam content and increases by increasing oxygen surplus ratio. It remains constant once an oxygen surplus ratio of $\Phi = 2.7$ has been exceeded.

Ignition behaviour of the NIS-PAR was investigated with vessel atmosphere initially filled with air (no oxygen starvation conditions). Under the investigated test conditions, ignition was observed for a hydrogen concentration of 4.1 vol % (no steam), but was not observed for steam contents of 40 vol % and above.

Interaction of molecular iodine with aerosols

The interaction between molecular iodine and aerosols (non-reactive or reactive) is one of the issues lacking experimental data for modelling and code validation purposes. The tests demonstrated that the removal of gaseous molecular iodine from the vessel atmosphere by interaction with the reactive silver aerosol (test Iod-26) was twenty-five times faster as compared to the test with the inert SnO_2 aerosol (test Iod-25). Tests Iod-25 and Iod-26 can be considered to provide the bounding cases for the removal of gaseous molecular iodine from atmospheres by interaction with aerosol, the reactive Ag aerosol providing the maximum and the inert SnO_2 the minimum removal rates under the given boundary conditions of the tests. Real containment aerosol can be expected to show removal effects between these bounding cases. This data can be used for the implementation of additional iodine depletion models in existing containment codes.

Release of gaseous iodine from a flashing jet

The test is designed to deliver experimental data on release of gaseous iodine from a flashing jet under high pressure and high temperature thermal hydraulic conditions. The design of the experiment is oriented towards PWR design-basis accident "steam generator tube rupture during reactor shut-down", but the data may be applicable also to other scenarios with flashing of primary circuit water. The results of the measurements in the THAI vessel (gas space and sump) and in the pressure vessel during the iodine flashing test Iod-29 consistently show that there was no gaseous iodine released from the flashing jet under the investigated test conditions. The iodine mass balance was found to be closed. The observation that no gaseous iodine existed in the THAI vessel is consistent with measurements on iodine speciation in the water of the pressure vessel, sampled during heat-up just before and just after

the flashing process. Although molecular iodine was injected before heat-up, only the iodide form was found, and iodate as a product of iodine hydrolysis was not detected above the detection limit of 5.4 % with respect to the sum of all iodine species. This indicates that the injected molecular iodine had quickly dissolved and hydrolysed, with intermediate hydrolysis species such as HOI and I_2OH^- reacting quantitatively with the steel wall of the primary vessel or metal ion impurities in the aqueous phase during heat-up to produce the non-volatile iodide form.

Laboratory scale experiments on the interaction of iodine and aerosols

In parallel to tests Iod-25 and Iod-26, a laboratory-scale test programme was run to support the design and procedure of THAI-tests Iod-25 and Iod-26, and to study a number of chemical parameters that could influence the iodine/aerosol interaction such as temperature, aerosol material and gaseous iodine concentration. The laboratory tests on I_2 / Ag interaction within the current THAI-2 project are qualitatively consistent with Iod-26 results, especially with respect to the similar and efficient iodine loading on the Ag surface. They provide additional data for parameter variations like relative humidity, iodine concentration in the gas phase, particle mass distribution, and for the behaviour of other aerosol material like CsI and Fe.

Analytical activities

The experimental programme of the THAI-2 project has been strongly supported by accompanying analytical activities performed by partners of the Analytical Working Group (AWG). The outcomes of these analytical activities are reported in the respective reports and publications: Blind pre-test calculations have been performed for Iod-25 and Iod-26 by GRS and for Iod-29 by IRSN. In the frame of a benchmark exercise, blind pre-test and open post-test calculations for test HR-35 have been performed, the results of which are documented in a comparison report. For all of the 10 HR-tests, post-test calculations have been performed by partners of the AWG.

By the use of the results of experiments Iod-25 and Iod-26, the respective models have been further validated by JAEA for modelling severe accident scenarios in PWR. Using the results of test Iod-29, IRSN performed post-test calculations. Accompanying design calculation and post-test analysis of the Iod-29 test indicate lack of validation of existing iodine chemical models in the aqueous phase for the investigated Design Basis Accident (DBA) test conditions.

The use of the experimental results obtained in the frame of the present experimental program has helped considerably for achieving the goals of the THAI-2 project set-forward during its realisation, namely to improve confidence in available LP- and CFD-tools for containment analysis. The analytical activities performed by the project partners provided very complete analysis of the experimental data produced by carrying out pre- and post- test calculations of the test series performed. The enhanced tools validated against experimental data will provide a more reliable approach for the analysis of the containment response during accidents and the planning of accident management measures.

Conclusions and recommendations

The NEA THAI-2 project as a continuation of the THAI project has widely extended the knowledge and data base of severe accident related issues. In case of PAR investigations, an almost complete picture of its behaviour has been obtained which allows to assess this hydrogen mitigation measure under a large spectrum of accident conditions and to implement appropriate models in accident analyses codes. The tests about hydrogen deflagration behaviour during water spray operation provide data for a sound safety-related assessment of spray systems when used during severe accidents. The tests about molecular iodine release and interaction with aerosols are necessary for the quantification of a potential accident source term.

The database developed in the frame of the THAI-2 project has been and will be extensively used for improvement, further development and validation of CFD and LP codes applied in reactor safety

analysis thus, increase confidence in the available accident analysis tools. The accompanying analytical activities of the project partners provided intensive analyses of the experimental data by pre- and post-test calculations performed.

Remaining open questions shall be further addressed in upcoming projects. Issues proposed are related to investigations on PAR performance under counter-current flow conditions and to hydrogen deflagration experiments in a two-compartment system under natural convection flow regimes. Additionally, in the light of the Fukushima Daiichi accident, experimental investigations are proposed to study fission product (aerosols and gaseous iodine) re-entrainment from a water pool at elevated temperature (as a consequence of continuous water heating or venting induced boiling (depressurisation). Another proposed experiment is related to the "delayed source term" and shall investigate the resuspension of aerosols as well as iodine deposits from steel/painted surfaces due to hydrogen combustion.

For this purpose, an THAI-3 project was started in early 2016.

1. INTRODUCTION

Progress in nuclear reactor containment safety research has led to a deeper understanding of phenomena and processes that may occur during a severe accident. However, a number of safety relevant gaps in this understanding exist due to limitations in the experimental data bases. Such gaps are related to e. g. hydrogen combustion under specific conditions and to the behaviour of fission products, in particular iodine and aerosols interactions.

In the case of hydrogen, uncertainties show up in the behaviour of hydrogen deflagration in the presence of an operating spray system and in the performance of a Passive Autocatalytic Recombiner (PAR), i.e. start-up behaviour, hydrogen depletion efficiency, recombination rate and ignition potential in an oxygen-lean atmosphere. The relevance for reactor safety is the destructive potential of fast deflagrations.

In the case of fission products, interaction processes of molecular iodine with non-reactive (SnO_2) and reactive (Ag) aerosol was subject of experiments. Another issue was to quantify the release of gaseous iodine from a flashing jet, representing a PWR steam generator tube rupture under reactor shutdown conditions.

In order to develop a high level of confidence that nuclear reactor containment systems and components establish an exceptional level of safety, it is of utmost importance to understand the aforementioned phenomena and processes which may occur during an accident. At international level, large efforts have been undertaken for the evaluation and further development of Lumped Parameter (LP) and Computational Fluid Dynamics (CFD) codes for the analysis and simulation of severe accident conditions. However, the validity of these LP or CFD codes can be better assessed against technical and large scale experiments reasonably representative of dominant accident sequences or conditions. Additionally, as more and more passive safety systems are being installed in operating reactors and planned for future reactors, the aforesaid issues need to be coupled with operation of passive safety systems in phenomena-oriented experiments. The need to carry out such coupled-phenomena experiments in the frame of an international programme is twofold: First, to enhance confidence in the performance of passive mitigation systems during severe accident scenarios and second, to establish a common database accessible by a large research community to support further development and validation of LP and CFD codes.

The THAI-2 project was started in August 2011 under the auspices of the Nuclear Energy Agency to fill knowledge gaps and to deliver suitable data for evaluation and simulation of the hydrogen deflagration behaviour during spray operation, PAR operation under low oxygen concentration, iodine interaction with reactive and non-reactive aerosol particles, and gaseous iodine release from a flashing jet. The tests are performed in the technical-scale THAI test facility (a 60 m³ stainless steel vessel 9.2 m high and 3.2 m in diameter) with exchangeable internals for multi-compartment investigations. The vessel is designed for a maximum overpressure of 1.4 MPa at 180 °C and can withstand moderate hydrogen deflagrations.

The THAI-2 project is supported by signatories, i.e. safety organisations and research laboratories from 11 countries, Table 0-1, and received contributions from the German partners listed in Table 0-2.

Table 0-1:   THAI-2 signatories with their national origin

Signatories	Country
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH	Germany
Atomic Energy of Canada Ltd (AECL)	Canada
Nuclear Research Institute of the Czech Republic	Czech Republic
Valtion Teknologian Tutkimuskeskus (VTT) and Säteilyturvakeskus (STUK)	Finland
Institut de Radioprotection et de Sûreté Nucléaire (IRSN)	France
Nuclear Safety Research Institute (NUBIKI) and Paks nuclear power plant	Hungary
Japan Atomic Energy Agency (JAEA)	Japan
Korea Atomic Energy Research Institute	Korea
Ministry of Infrastructure and the Environment	Netherlands
Swedish Radiation Safety Authority (SSM)	Sweden
Office for Nuclear Regulation (ONR), agency of the health and Safety Executive (HSE)	United Kingdom

Table 0-2:   German Partners in the THAI-2 Project

Project partners
Becker Technologies GmbH (Operating Agent)
AREVA GmbH Erlangen
Siempelkamp NIS Ingenieurgesellschaft mbH in Alzenau
RWTH Universität Aachen
Forschungszentrum Jülich
RUB Ruhr-Universität Bochum
Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH

The operating agent was supported by AREVA's Radiochemical Laboratory, Erlangen, Germany, to operate the iodine instrumentation, to perform chemical analyses, to perform the iodine specific laboratory-scale tests, and to evaluate and assess the iodine results.

PAR tests HR-40 to HR-42 were sponsored by the PAR vendor Siempelkamp NIS Ingenieurgesellschaft mbH in Alzenau, Germany.

Table 0-3 provides the details of the test matrix designed to provide better insight on the hydrogen and fission products related issues. In addition to the experimental work, an accompanying analytical workgroup was established in the frame of the THAI-2 project, aimed at performing design calculations, the evaluation of the test results for further development and validation of the predictive capabilities of advanced LP- and CFD-codes currently applied in reactor safety.

Table 0-3:   Test matrix developed for the THAI-2 project

Test series/ Test no.	Test series/test designation	No. of tests	Objectives
1	Hydrogen Deflagration (HD) Tests with spray	7	- Influence of spray operation on H_2 combustion behaviour for different gas- and spray-water temperatures, different hydrogen and steam content, and for upward and downward burn direction, - Influence of spray water droplet size - Influence of cold spray on steam condensation and the thermal hydraulic conditions
2	Hydrogen Recombiner (HR) Tests	10	- PAR performance in oxygen-lean atmosphere - onset of recombination under different initial conditions - hydrogen recombination rate - hydrogen depletion efficiency - ignition potential - different PAR types
3	Interaction of molecular Iodine with Aerosols	2	- Interaction of I_2 and non-reactive SnO_2 and their depletion from the vessel atmosphere - Interaction of I_2 and reactive Ag and their depletion from the vessel atmosphere
4	Release of gaseous Iodine from a flashing jet	1	- Quantification of gaseous iodine release under thermal hydraulic conditions (test design oriented towards PWR steam generator tube rupture scenarios during reactor shutdown (DBA scenario)
5	Laboratory scale experiments on the Interaction of Iodine and Aerosols	25	- Investigation of the Interaction of I_2 with various reactive and non-reactive particles for a wide range of chemical and thermal conditions

The experimental data obtained from the different test series demonstrated the capability of the THAI facility in producing high-quality, high-resolution data on the issues mentioned in Table 0-3. The tests provide a better physical understanding and data for the validation and further development of analytical tools related to:

- A deeper insight into the phenomenology of hydrogen deflagration during spray operation. The tests provide a comprehensive database for further development and validation of combustion models implemented in containment system codes. Test conditions and parameters were defined considering severe accident thermal hydraulic conditions. For most of the tests, initial conditions were set to 1.5 bar, 10 vol % hydrogen concentration and 25 vol % steam concentration in pre-mixed gas atmosphere. Test parameters varied were burn direction (upward or downward), spray duration before ignition, spray water temperature, droplet size distribution and gas temperature.

- The behaviour of two commercial PAR units (AREVA and NIS) in initially air-filled as well as in oxygen-lean atmospheres. The test series evaluates the influence of initial thermal-hydraulic conditions (gas temperature, pressure) and gas composition (H_2 concentration, steam concentration, O_2/N_2 ratio) on PAR performance and PAR induced ignition behaviour. The data obtained significantly improve the existing knowledge about the influence of low oxygen concentration on PAR performance like onset of recombination, H_2 recombination rate and H_2 depletion efficiency, effect of initial pressure and temperature on PAR performance, effect of steam content in a superheated or saturated condition on PAR

performance, and about the steam inertisation effect on PAR induced ignition for different O_2/N_2 ratios.

- The interaction of gaseous molecular iodine with a non-reactive aerosol (SnO_2) and its physisorption on the particle surfaces as well as the interaction with reactive aerosol (Ag) and its chemisorption on the particle surfaces in dry air during an intermediate time period of typically several hours.

- The quantification of gaseous iodine release from a flashing jet, simulating a PWR DBA scenario: steam generator tube rupture under reactor shutdown conditions.

- The determination of chemical parameters like time dependent adsorption and desorption of the iodine-aerosol interaction for various aerosols, e. g. Ag, CsI, Fe, SnO_2 by laboratory-scale tests to provide data for modelling and to support design and procedure of the large-scale THAI tests.

For most of the experiments design calculations have been performed by the Operating Agent for the detailed determination of test conditions, test parameters and test procedures and for the detailed design and configuration of the instrumentation and other experimental equipment.

Numerous analytical activities with respect to the tests are documented in the respective reports and publications: Blind pre-test calculations have been performed for Iod-25 and Iod-26 by GRS [Web 2014], and for Iod-29 by IRSN [Can 2013]. In the frame of a benchmark exercise, blind pre-test and open post-test calculations for test HR-35 have been performed, the results of which are documented in a comparison report [Fre 2013]. For all of the 10 HR-tests, post-test calculations have been performed by partners of the Analytical Working Group (AWG).

By the use of the results of experiments Iod-25 and Iod-26, the respective models have been further validated by JAEA [Ish 2014] for modelling severe accident scenarios in PWR. Using the results of test Iod-29, IRSN performed post-test calculations [Can 2014].

The use of the experimental results obtained in the frame of the present experimental program has helped considerably for achieving the goals of the THAI-2 project set-forward during its realisation, namely to improve confidence in available LP- and CFD-tools for containment analysis. The analytical activities performed by the project partners provided very complete analysis of the experimental data produced by carrying out pre- and post- test calculations of the test series performed. The enhanced tools validated against experimental data will provide a more reliable approach for the analysis of the containment response during accidents and the planning of accident management measures.

The data of all the performed experiments have been processed, qualified and stored on a server accessible to signatory partners. Detailed experimental results and analysis to the user of the data is provided in Quick Look Reports (QLR) and Technical Reports. The Management Board of the THAI-2 project has agreed that these Quick Look Reports and Technical Reports will be disclosed for non-signatories after 31 July 2017 and can be obtained after this date upon request from the NEA. These reports are compiled in Appendix A of this report.

2. GENERAL OBJECTIVES OF THE THAI-2 PROJECT

The overall objective of the THAI-2 project is to address open questions concerning the behaviour of hydrogen, iodine and aerosols in the containment of water cooled reactors during design-basis or severe accident typical conditions. The understanding of the processes taking place during such events is essential for evaluating the challenge posed on containment integrity (hydrogen) and for evaluating the amount of airborne radioactivity (iodine and aerosols), which may be present inside a containment during such severe accidents with core damage. The project is to generate valuable data for evaluating the hydrogen deflagration behaviour during spray operation and its effective removal by PARs. Concerning fission products, the project will focus on the interaction of molecular iodine with non-reactive and reactive aerosol particles. Another important issue related to iodine behaviour is to quantify the release of gaseous iodine from a flashing jet, representing a PWR Design Basis Accident scenario (steam generator tube rupture during reactor shutdown sequence). An extensive analytical effort will accompany the experimental program consisting of code calculations to support test design, for pre-test assessments, result evaluation and extrapolation to reactor conditions.

The project contributes to the validation and further development of advanced LP- and CFD codes used for reactor applications by e. g. providing experimental data for code benchmark exercises. Furthermore, the project is aimed at maintaining competence and expertise in the field of reactor safety in the NEA member countries and promotes international co-operation between its member countries.

3. EXPERIMENTAL FACILITY

In this section, the basic configuration, including the basic instrumentation of the test facility is described.

3.1 Basic configuration

The tests have been performed in the THAI (Thermal-hydraulics, Hydrogen, Aerosols, and Iodine) containment test facility which is operated by Becker Technologies GmbH at Eschborn, Germany, under the sponsorship of the German Federal Ministry for Economic Affairs and Energy. Main component of the facility is a cylindrical stainless steel vessel of 9.2 m height and 3.2 m diameter with a total volume of 60 m³, Fig. 0-1. The maximum admissible overpressure is 14 bar at 180 °C.
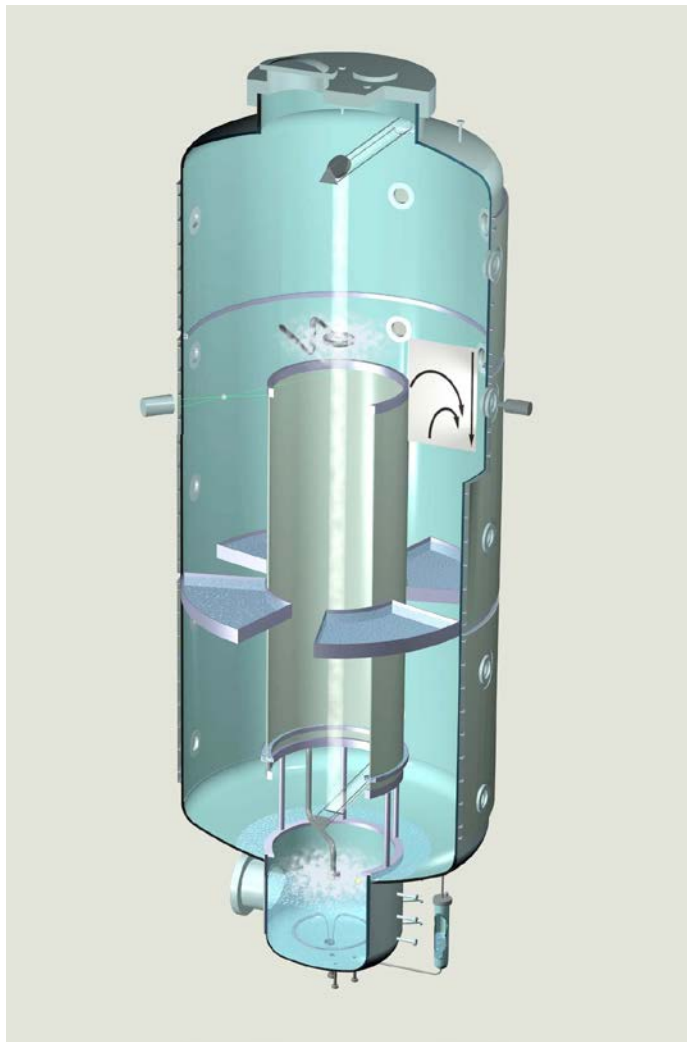
Fig. 0-1: THAI Test Vessel

3.1.1 Supply systems

The vessel is equipped with supply systems for steam, gases (hydrogen, helium, air, nitrogen, oxygen), aerosols (soluble, non-soluble, droplets), and iodine as described briefly in the following.

Water supply

For the HD-tests with spray a water flow rate of 1 kg/s was established. The overpressure in the water line was maintained constant by use of a compressed-air operated separate pressure vessel with a sufficient large amount of water to ensure a constant water flow rate during spray operation and an instantaneous water flow after opening of the quick-action valve.

Steam supply

Steam is provided by a local 110-kW steam generator. The test-specific steam concentration is adjusted by steam injection until the required steam partial pressure is achieved, i.e. by pressure measurement.

Gas supply

Hydrogen gas is taken from gas bottles, its pressure is reduced to a constant level of e.g. 5.5 bar overpressure; its mass flow (e.g. 12 Nm³/h) is adjusted by a needle valve, measured by a float-type (rotameter) gauge, and fed into the THAI vessel via a ring-shaped perforated line close to vessel bottom.

Oxygen is also taken from gas bottles and injected into the vessel at test specific locations. Dry oil-free air is taken from the compressed air supply system and additionally filtered. The flow rate is controlled by a flow meter and a motor valve.

Nitrogen, which is used in the HR-tests for purging the THAI vessel to achieve an almost oxygen-free atmosphere, is taken from a gas bottle battery.

Aerosol injection

Non-soluble aerosol (SnO_2 and Ag) is produced by means of a fluidized bed reactor. The fluidized bed reactor is filled with SnO_2- or Ag-powder which is then fed with air as carrier gas via a pipe into the vessel. By defined opening and closing times of the air supply together with variations in the air flow rate the amount of injected aerosol is controlled.

Iodine injection

For the injection of iodine (labelled with I-123) into the vessel atmosphere of the I_2/aerosol tests, the iodine reactor - a small glass flask placed inside a steel container - was filled with solid I_2 and I-123. The flask was heated above the melting point of the I_2 to allow for a homogeneous labelling. Thereafter, the flask was mechanically destroyed and the labelled I_2 vapour was injected into the THAI vessel with a pre-heated carrier gas (synthetic air) at a rate of 20 to 50 l/min over a time period of 10 to 15 min. Iodine was injected at H = 8.4 m in the vessel centreline against a baffle plate.

3.1.2 Instrumentation

The standard instrumentation encompasses thermal hydraulic and gas composition instrumentation. It is supplemented by special instrumentation depending on the specific test design and test objectives.

Feed mass flow measurements

Steam mass flow as well as compressed air mass flow is measured by a float-type gauge (rotameter).

The mass flow of the gases hydrogen, helium, nitrogen and oxygen (taken from gas bottles) is measured by float-type gauges. For test data evaluation, the integral of the gas mass flow measured with this device is compared with the injected gas mass determined from the start and the end pressure of the gas bottles and compared with the pressure increase in the vessel.

Gas concentration measurement

Fifteen gas sampling lines continuously take gas samples from the PAR and from the vessel atmosphere at different locations. The sampled gas is fed to a 15-channel gas concentration analyser system which determines the hydrogen concentration in dry air. In case of experiments with steam, steam is removed by coolers and condensate traps upstream of the analysers. Up to six of the 15 lines for hydrogen concentration measurement are additionally equipped with oxygen sensors. Concentration of hydrogen and oxygen is then determined in dry air. For those experiments for which the steam in the vessel atmosphere was at saturation state, the real ("wet") concentrations of hydrogen and oxygen in the vessel atmosphere could be determined by use of the steam table for the partial pressure of steam at the temperature measured in the vessel.

In some of the PAR tests, a mass spectrometer was installed to measure gas concentrations. However, the measurements proved not to be reliable and therefore were excluded from data analysis.

Temperature measurement ("slow thermocouples")

Up to 80 calibrated 1.5-mm thermocouples are installed in the THAI vessel for temperature measurement in the vessel atmosphere with an accuracy of $\pm$ 0.3 K for a temperature range 0 to 130 °C.

In addition, another 200 thermocouples are installed to measure the vessel wall temperature.

Temperature measurement ("Fast thermocouples")

To monitor the PAR catalyst temperature, flame front propagation and flame temperature during hydrogen combustion, "fast" sheathed thermocouples (outer diameter 0.5 mm) were used. As the arrival of the flame front at the individual thermocouple causes a steep temperature signal rise, the time points of flame front arrival can be determined. with an uncertainty smaller than 0.01 s.

Pressure measurement

The initial vessel pressure and the pressure increase due to gas injections are determined by two absolute-pressure transducers. These pressure transducers are backed-up by the readings of two high-precision manometers (accuracy ±4 mbar or ±6 mbar). For highly transient pressure measurement (e.g. during HD-test series or HR-tests with ignition), two fast pressure transducers are installed.

Flow velocity measurement

In order to determine the H_2 recombination rate of a PAR, H_2 concentration at PAR inlet, at PAR outlet and the volume flow through the PAR is to be measured. The latter is achieved by a turbine flow meter installed in the inlet flow channel. The vane wheel transducer applied has a lower threshold value for vane wheel rotation of approx. 0.25 m/s

Aerosol measurements

In order to determine aerosol concentration and particle size distribution, different methods have been established.

Measurement of the aerosol mass concentration is performed with single filters. The filters are weighed before and after the experiment. From gas sample volume flow, the sampling time and mass of the retained particles, a mean aerosol concentration can be calculated for the sampling period.

A number of filter samplings were performed in parallel, to measure the I-123 loading and thus to measure the mass concentration of iodine in aerosol form. This is referred to as aerosol single filter measurements and the results are provided with the iodine results.

Furthermore, low-pressure cascade impactors are available to measure the particle size distribution. The impactors separate 10 particle size classes, the mass of each particle class collected during the sampling period is determined by weighing.

An additional aerosol concentration measurement is performed by laser light extinction measurement, which provides a continuous, in-line and non-intrusive measurement. Reference for this system is the concentration measurement of the single filter stations.

Iodine measurements

The measurement of iodine is mainly based upon the use of detectors for radioactive I-123 as a tracer, measuring samples from gas scrubbers and Maypack filters, or online monitoring of deposition coupons. It is particularly relevant for test Iod-29, for which no I-123 tracer was used, that chemical analysis of iodine and its speciation using e.g. inductively coupled plasma mass spectrometry (ICP-MS) at AREVA's Radiochemical Laboratory at Erlangen is also involved.

For the monitoring of iodine labelled with radiotracer I-123, two lead-shielded NaI scintillation detectors are available for discontinuously measuring all samples of an iodine test in THAI in defined and calibrated geometries. At the usual measuring time of 1 minute, the acquired counts are usually sufficiently high to achieve a statistical error of a radiation measurement reasonably low and any scattering of the resulting iodine concentrations mainly can be attributed to the uncertainties of the sampling techniques.

Measurement of gas-borne iodine concentrations is based upon the gas scrubber (GS) technique with six gas scrubbers. To avoid long gas sampling lines often associated with significant iodine losses, these gas scrubbers are installed inside the vessel at various positions. Absorber solution is fed into and removed from the gas scrubbers from outside the test vessel repeatedly during a test, so that concentration evolutions with time can be determined. After sampling (scrubbing) for several minutes at a gas flow rate 0.5–1 l/min, the absorber is recovered, cooled down and measured by I-123-γ-counting and by UV-VIS spectroscopy. Sampling times vary between 2 min and 30 min, with increasing tendency in the course of a test to account for the depletion of gas-borne iodine by deposition onto surfaces and for the short half-live of I-123 (13.2 h).

Two Maypack filter sampling stations are available to discriminate gas-borne iodine in molecular iodine (I2), gaseous organic iodide, and iodine in aerosol form. Each Maypack device consists in general of one quartz filter to retain aerosols, two filters to retain gaseous I2, and two charcoal filters (KI-impregnated) to retain organic iodides. This enumeration defines at the same time the flow direction. The I2 filter material consists of KI-coated silica grains, available as commercial product DSM-11 by Süd-Chemie AG.

In test Iod-29, a predefined amount of molecular iodine was dissolved in the pressure vessel water. Samplings from aqueous phases and the THAI vessel gas phase were measured with the standard instrumentation. Samples were analysed chemically for iodine at AREVA's Radiochemical Laboratory, Erlangen, Germany, using mainly ICP-MS. These analyses also included the determination of phosphor, which was used in the phosphate form to establish the desired pH-value of the water, and at the same time also considered as a tracer for quantifying the efficiency of water droplet trapping by the pre-filters of the gas scrubbers.

4. PERFORMED EXPERIMENTS

In this chapter, the tests of the experimental program with their specific objectives, their specified and measured test parameters and their most important results are compiled.

4.1 HD test series

The objective of the hydrogen deflagration experiments was to provide additional data for an improved understanding of hydrogen combustion phenomena (since hydrogen deflagrations cannot be ruled out completely even by operating PAR) and for the further development and validation of combustion models incorporated in advanced LP- and CFD codes. More details can be found in the respective Technical Report [Gup 2014c].

Experiments have been conducted under conditions typical for severe accidents, including the operation of a spray system during combustion in a relatively large test facility, which allows investigating the effect of spray induced flow on upward and downward flame propagation, which is not feasible in small geometries. In large geometries, deflagrations may proceed faster due to e. g. increased turbulence generation and therefore may produce higher loads for the vessel and its internals.

Table 0-1 shows the test conditions of the HD-tests. For comparison, the conditions of the respective tests without spray operation, performed in the framework of the earlier THAI project have been added [Kan 2010a].

After the initial conditions had been established, the test phase started with the onset of water flow for the spray (t = 0 s). The test procedure applied was as follows:

- Injection of hydrogen from gas bottles into the test vessel until the specified H_2 concentration was reached.

- Isolation of H_2 concentration measurement system to exclude flame propagation inside the sampling lines.

- Activate spray operation.

- Ignition of the mixture by an arc igniter. Ignition is initiated 2 s after starting the spray operation (estimated time for droplet arrival time in the sump), except for test HD-35 with 60 s of spray operation prior to ignition.

- Open the gas sampling valves again for measuring the residual H_2 concentration in the vessel.

The results of the HD-tests with spray are discussed and compared to reference tests without spray performed in the previous THAI project [Kan 2010a].

Apart from the influence of initial gas temperature and initial steam content on peak pressure reduction, spray operation adds up a beneficial effect for pressure reduction. It is therefore not surprising that the combination of elevated initial gas temperature, high steam content and spray operation produces the most significant peak pressure reduction. Operation strategy for a containment spray system may vary depending on reactor design and severe accident management guidelines. However, temperature and steam content in the containment depend on the scenario of a severe

accident and cannot be influenced directly by operator action, but develop to a "benign" situation with respect to combustion peak pressure.

An increased spray water temperature and larger water droplets produce slightly higher peak pressures than the reference tests with cold spray water and smaller water droplets.

Peak temperatures are clearly lower for the tests with low spray water temperature and either low initial temperature or steam (HD-30 and HD-31). Tests with 90 °C spray water temperature (HD-32.1) and larger droplets (HD-34) are in the same range as the reference tests without spray. Peak temperatures are observed in the vessel centreline for the tests without spray, but outside the spray cone for the tests with spray. Temperatures measured inside the spray cone (lower part of the vessel) showed much lower temperatures.

All tests with spray and upward combustion exhibit lower pressure peak, and a higher flame speed immediately after ignition in the lower part of the vessel compared to the reference tests without spray, see Fig. 0-1. An increase in flame speed is observed only for the vessel zones outside the spray cone close to the vessel wall, where the flame travels preferably and where upward directed gas velocities are induced by the spray operation.

$P = 1.5$ bar, $T_{gas} = 20$ ºC, $C_{H2} = 10$ vol. %, $T_{water} = 20$ °C

$C_{steam} = 0$ vol. %

$P = 1.5$ bar, $T_{gas} = 90$ ºC, $C_{H2} = 10$ vol. %, $T_{water} = 20$ °C

$C_{steam} = 25$ vol. %
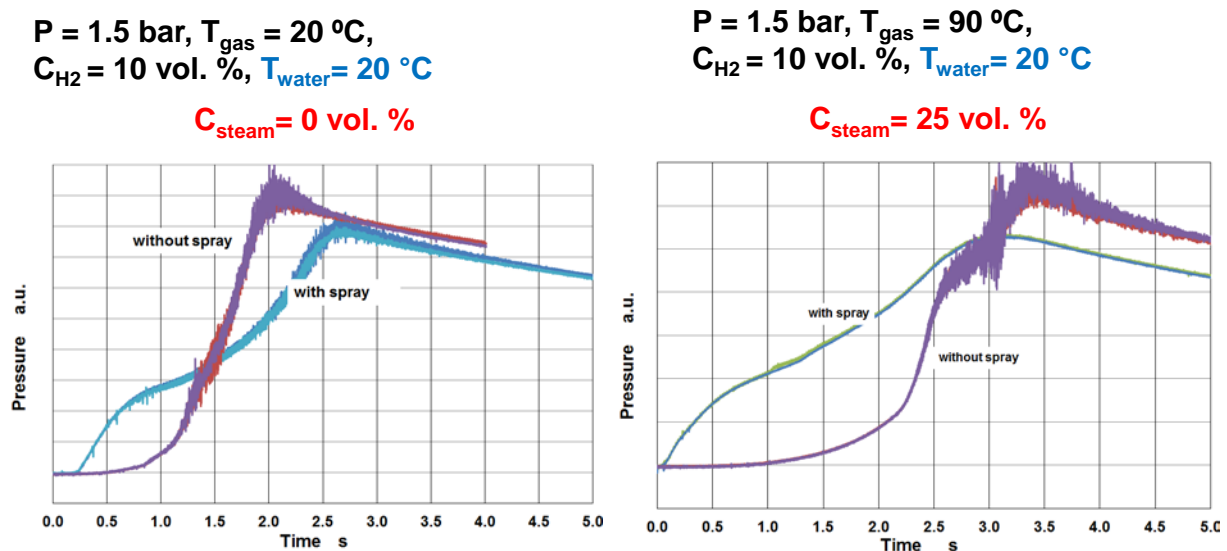
Fig. 0-1: Effect of spray operation on peak pressure

For test HD-31 with vessel atmosphere at 90 °C and a steam content of 25 vol %, this steam content together with droplet vaporisation is sufficient to suppress combustion completely in the spray cone zone down to 1 m below the spray nozzle, a zone of high water concentration. Further below, the flame is suppressed in terms of low flame speed, but combustion takes place, Fig. 0-2. For tests HD-32, HD-32.1, HD-33 and HD-35, the region of no combustion is slightly smaller.
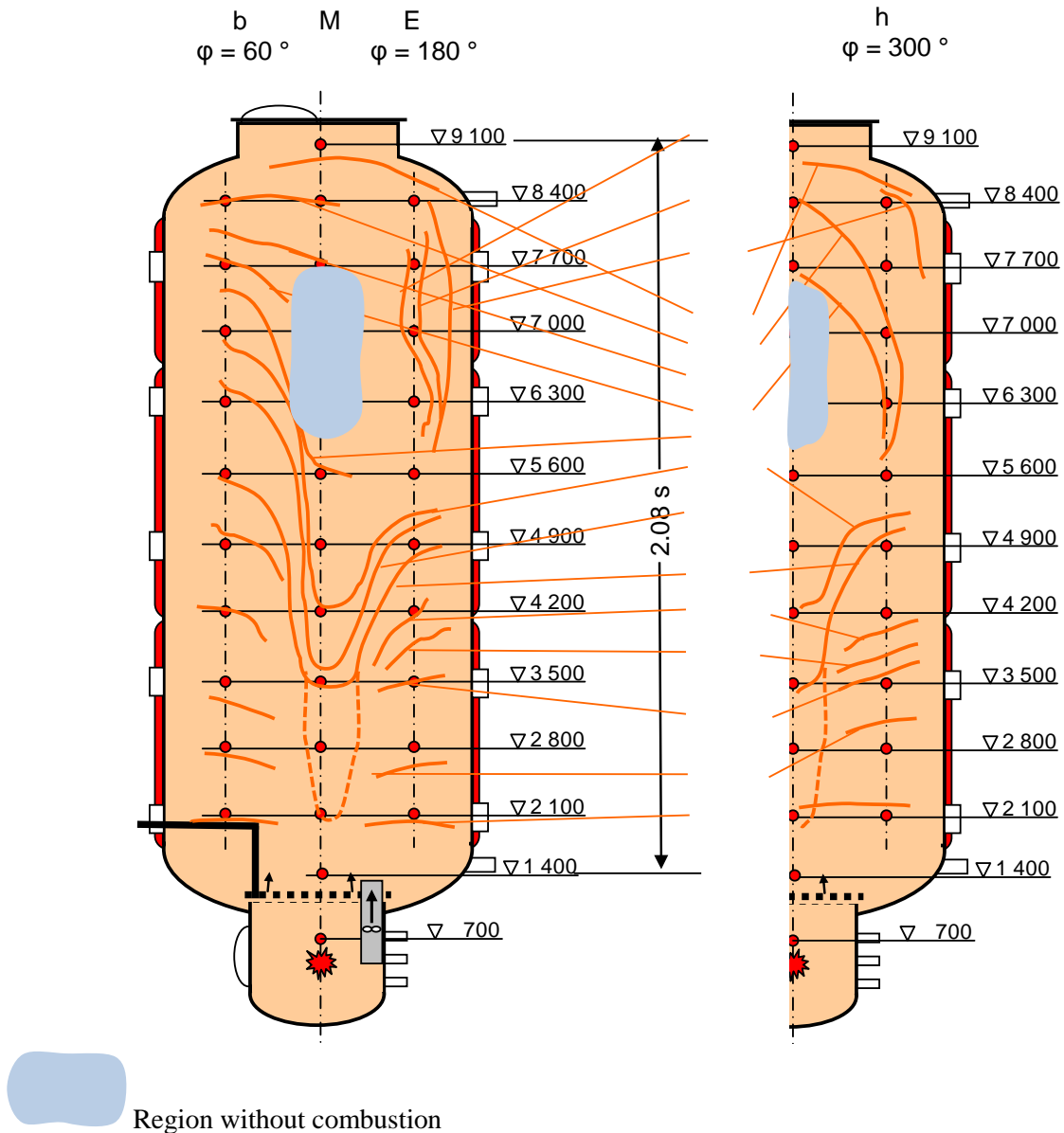
Fig. 0-2: HD-31: Flame front propagation as isochrones

For test HD-30 (cold vessel atmosphere, no steam, cold spray water) and for test HD-34 with the larger spray droplets (different spray nozzle), a no-combustion region could not be detected with the installed instrumentation. This allows concluding that the combustion suppression depends on the spray droplet size distribution and requires a marked initial steam content.

An even more pronounced influence of the spray induced flow field provides the comparison of the test HD-35 (with spray) and HD-23 (without spray) with downward burn direction Test HD-35 was the only test of the HD-spray-test series for which spray operation increased peak pressure and peak temperature compared to the reference test without spray. The reason is, that in case of downward burn direction flame speed becomes extremely high due to the superimposition of the downward travelling flame and the downward directed flow produced by the spray. Additional to the downward directed flow, the spray droplets induce turbulent fluctuations, which promote flame acceleration additionally.

4.2 HR test series

The objective of the Hydrogen Recombiner (HR) tests was to provide additional data about the behaviour of passive autocatalytic recombiners under severe accident typical conditions. More details can be found in the respective Technical Reports [Gup 2014a], [Gup 2014b], [Gup 2014d] and [Gup 2014e]. The experiments aimed at the operation behaviour of commercial PAR types provided by the PAR vendors, AREVA GmbH in Offenbach, Germany and Siempelkamp NIS Ingenieurgesellschaft mbH in Alzenau, Germany, with focus on

- onset of recombination,

- recombination rate and hydrogen depletion efficiency,

- conditions for ignition by a PAR,

under oxygen lean conditions for most of the experiments. The data will be used as a basis for modelling the PAR behaviour and PAR-initiated deflagrations in advanced LP- and CFD-codes.

The initial conditions of the HR experiments HR-33 to HR-42 are compiled in Table 0-2.

Test group HR-33 to HR-35 was dedicated to onset and performance behaviour, test group HR-36 to HR-37 to the ignition behaviour under oxygen lean conditions (AREVA-PAR).

Test group HR-38 to HR-39 was aimed at the onset and performance behaviour under oxygen lean conditions, test group HR-40 to HR-42 to the PAR performance and ignition behaviour under different thermal-hydraulic conditions (NIS-PAR).

AREVA-PAR

By comparing the results of tests HR-33 to HR-35 with the previous AREVA PAR tests conducted in the THAI test facility [Kan 2010b], it has been demonstrated that the onset of hydrogen recombination is much faster and occurs at lower concentrations, if the catalyst surface is exposed initially to hydrogen molecules (no oxygen present) than in the tests with the catalyst surface being exposed initially to an air atmosphere (oxygen molecules).

A comparison of the hydrogen depletion efficiencies of tests HR-33 to HR-35 with the HR-9 and HR-12 tests conducted in the previous THAI project is depicted in Fig. 0-3. It reveals, that the hydrogen depletion efficiency increases almost linearly for an oxygen surplus ratio up to about $\Phi = 2$ in the oxygen starvation regime, regardless of the test conditions. For an oxygen surplus ratio $\Phi > 2$, hydrogen depletion efficiency γ does not increase further and stabilises within a range of $50 - 70\,\%$.
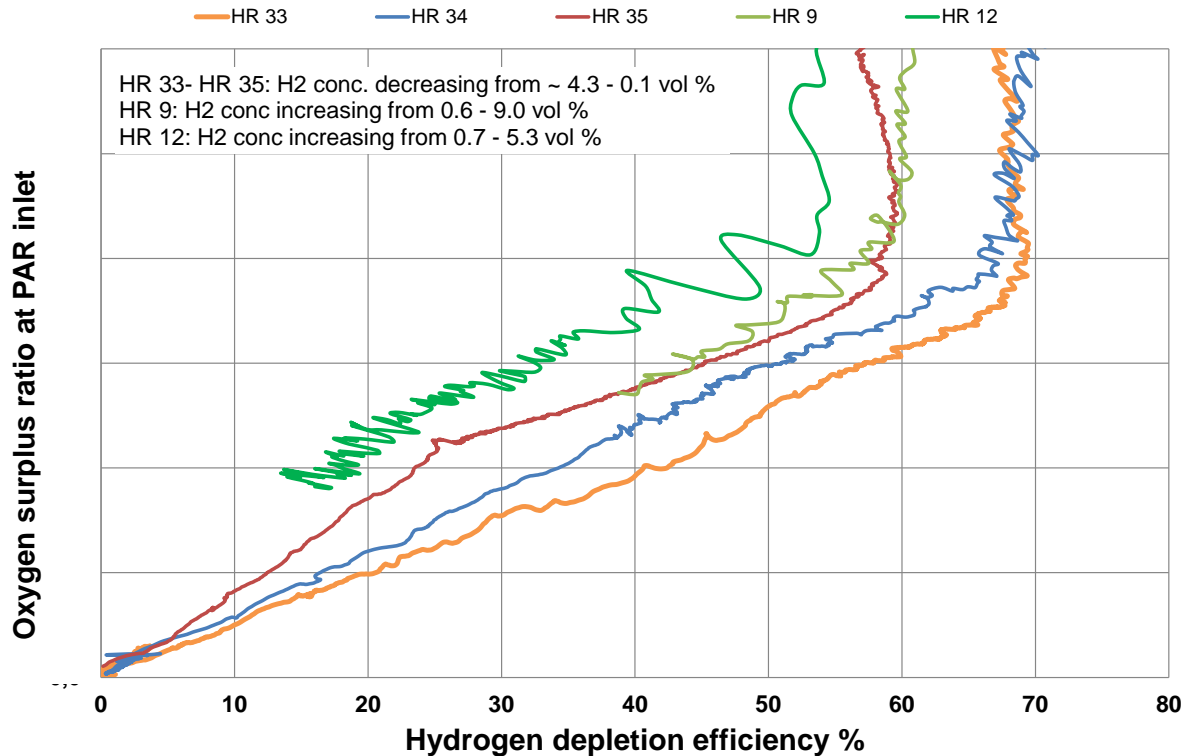
Fig. 0-3: Effect of O₂ surplus ratio on hydrogen depletion efficiency

Recombination rates have been determined both for H_2 controlled and O_2 controlled test phases. The measurement-based recombination rates are to be compared to those calculated with the empirical correlation provided by the PAR vendor AREVA. For test HR-33 the agreement is fair for low oxygen surplus ratios up to $\Phi = 1.5$ with an overestimation of the recombination rate, then for ongoing H_2 depletion the recombination rate is underestimated.

Similar results are found for tests HR-34 and HR-35 with the deviation between measured and calculated recombination rate becoming larger for high steam contents. For all three tests, the recombination rate is underestimated for oxygen surplus ratios of 2 or higher. This underestimation can be regarded as a conservative feature of the AREVA correlation.

The initial thermal hydraulic conditions of test HR-36 and test HR-37 (ignition tests) differed in steam content in order to evaluate the steam concentration threshold for inertisation for a given dilution ratio of $\delta = 0.10$ (defined as $\delta = c_{O2} /c_{(N2+O2)}$), and an initial H_2 concentration of 10 vol %. The vessel gas volume was ignited for test HR-36, but not for test HR-37.

For test HR-36 at the point of time of ignition and for test HR-37 at the point of time of maximum catalyst surface temperatures, gas compositions at PAR inlet and above catalyst within the PAR were almost the same for both tests and very close to the flammability limit. The main difference was the gas composition measured above the PAR in the vessel, which is clearly in the flammability range for test HR-36, but on the edge of the flammability limit for test HR-37. Considering the higher steam content for test HR-37, gas composition might have been even slightly outside the flammability range, Fig. 0-4.

The results of tests HR-36 and HR-37 are in accordance with an analysis performed by IRSN that for a dilution ratio $\delta = 0.10$, which indicated that an O_2-H_2-N_2-H_2O mixture becomes inert once the steam concentration exceeds 30 vol % [Mey 2012].

By combining the results of the test HR-36 and other HR tests on ignition behaviour conducted in the previous THAI project [Kan 2010b], the impact of dilution ratio δ on inertisation of an O_2-H_2-N_2-H_2O mixture is clearly demonstrated: The HR-36 test data indicate that to ignite a gas mixture, PAR

does not necessarily need to operate at its full capacity with O_2 surplus ratio $\Phi \geq 2 - 3$ required for an optimal performance. At the time of ignition in the HR-36 test, the PAR was operating with a H_2 depletion efficiency below 40 % and Φ was much lower than 2. Therefore, Φ is not a useful criterion for ignition by PAR as ignition can occur for a large range of Φ.

The effect of variations in gas composition may also have an impact on PAR ignition threshold during an accident progression, depending on the time and scenario of an accident. Therefore, it is desirable to develop a set of conditions which can be used to assess the PAR ignition potential under a broad range of accident typical conditions. Based on former and present PAR experimental data in THAI, the following set of conditions can be regarded as necessary prerequisites for ignition by a PAR:

- Hydrogen concentration at PAR inlet $\geq$ PAR ignition threshold H_2 concentration under O_2 surplus conditions (6 vol % H_2 /0 % steam and 8 vol % H_2 /45 vol % steam),

- O_2 concentration at PAR inlet $\geq$ 6 vol % and 1.1 times PAR ignition threshold H_2 concentration under O_2 surplus conditions,

- Steam concentration < inertisation limit corresponding to the given dilution ratio and minimum ignition threshold H_2 concentration, e.g. < 25 vol % for $\delta = 0.1$,
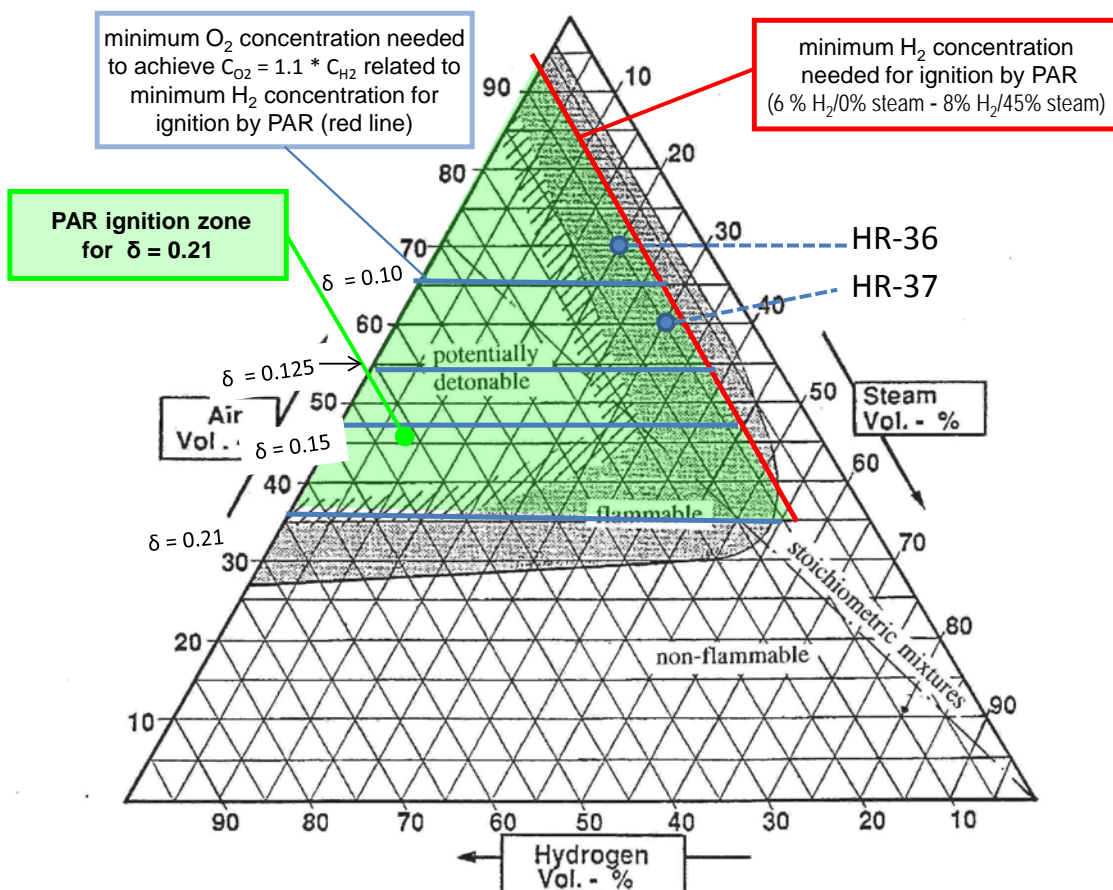

Fig. 0-4: Flammability limit of the H_2-O_2-H_2O system for different dilution ratios

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NIS-PAR

By comparing the results of tests HR-38 and HR-39 with other NIS PAR tests conducted in the THAI facility; [Kan 2009], [Gup 2014e], it has been found that the onset of hydrogen recombination is much faster and occurs at lower concentrations, if the catalyst surface is exposed initially to hydrogen molecules (no oxygen present) as in the tests HR-38 and HR-39 than in the tests with the catalyst surface being exposed initially to air.

Test results indicate that the hydrogen depletion efficiency increases almost linearly for an oxygen surplus ratio up to about $\Phi = 2$ (oxygen starvation regime). Above an oxygen surplus ratio of $\Phi = 2$, further increase of hydrogen depletion efficiency becomes very small and remains between 40-60 %. No marked influence of different initial test conditions like pressure, temperature and steam content on hydrogen depletion efficiency could be identified, Fig. 0-5.
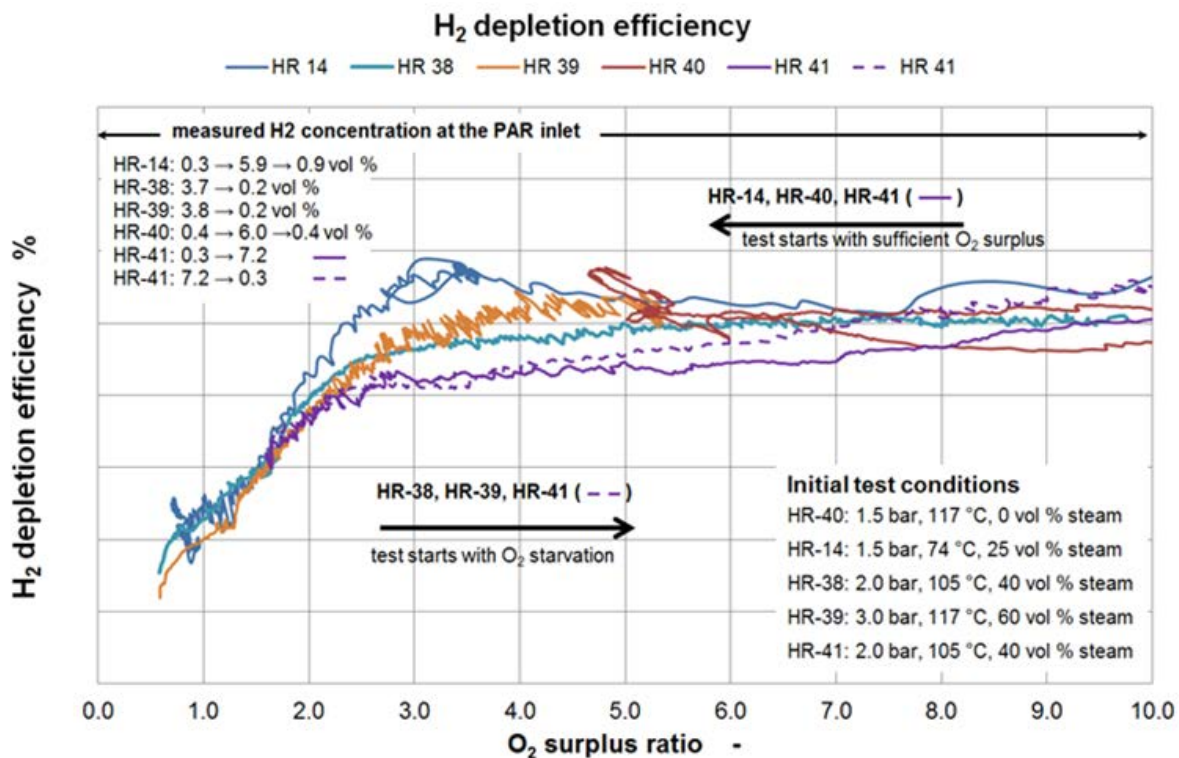
Fig. 0-5: Hydrogen depletion efficiency vs. oxygen surplus ratio for various HR-tests

Previous PAR tests which started with an air-filled atmosphere, indicated that as long as sufficient oxygen is available, recombination rate increases almost linearly with H_2 concentration at the PAR inlet. Then, the quotient of H_2 recombination rate divided by H_2 concentration at PAR inlet should be approximately constant. For tests HR-38 and HR-39, initial air atmosphere has been replaced by an almost inert atmosphere. After adjusting the desired H_2 -and steam concentrations, PAR operation started after O_2 injection. For a given initial H_2 concentration, H_2 recombination rate was found to increase almost linearly with O_2 concentration at the PAR inlet in an O_2 lean atmosphere.

The critical surplus ratio at which transition from O_2 surplus to O_2 starvation condition occurs was determined to be 2.75 for the NIS PAR based on the HR-14 test [Kan 2009]. This critical O_2 surplus value has also been confirmed by the NIS PAR tests HR-40 to HR-42, which had been conducted with different initial gas composition and thermal-hydraulic conditions.

For test HR-40, the first combustion event was observed for a H_2 concentration of 3.7 vol % at PAR inlet and of 4.3 vol % in the inner cylinder, and catalyst surface temperature of 542 °C. Though the catalyst surface temperature might have been sufficiently high to ignite the mixture, the H_2 concentration at PAR inlet was too low (below the lower flammability limit) for an ignition within the PAR.

The measured data indicate that the ignition occurred within the inner cylinder with the highest H_2 concentration. Most probable source of ignition was the expulsion of particles produced within the catalyst cartridges and transported to the vessel atmosphere after passing through H_2 lean atmosphere inside the PAR. As observed in-situ, these particles start to glow while floating in the vessel atmosphere (which means that they consist of catalytic material) and thus demonstrate their ability to act as a potential ignition source after entering into a region of flammable gas mixture. These "glow worms" remain airborne for an extended period of time and can be transported in the lower vessel plenum with the PAR induced convection loop in the vessel.

On continuation of the hydrogen release, measured test data detected a second ignition event in the vessel atmosphere. Also for the second ignition event which occurred in the upper part of the vessel, the "glow worms" have to be assumed as ignition sources. The gas concentrations at PAR inlet and in the vessel prior to the second ignition event were clearly in the flammable range and the catalyst surface temperature was high enough to ignite. However, the maximum H_2 concentration measured at PAR outlet was 2.7 vol % only (the H_2 concentration above the catalyst is about the same) which is too low to propagate a stable flame within the PAR from the catalyst cartridge to the outlet (even considering the wider flammability limits for a gas of 350 °C), and hence, no ignition source existed at PAR outlet.

In case of HR-41 test with an initial steam content of 39.7 vol % (superheated), for the first H_2 release (Phase 1 of the test), onset of recombination was observed at about 1 vol % H_2 at PAR inlet. H_2 release yielded catalyst temperatures up to 574 °C and a H_2 concentration at PAR inlet up to 5 vol %. The oxygen surplus ratio remained above $\Phi = 4$ during the entire release period of Phase 1. In Phase 2, H_2 release duration was longer and the maximum H_2 concentrations were higher (7.3 vol % at PAR inlet and 9 vol % in the vessel volume). The maximum catalyst temperature yielded 670 °C. The recombination rate and the H_2 depletion efficiency measured were in the same range as for the comparable tests of the preceding HR-test series (HR-14 to HR-16) with NIS-PAR.

For the test HR-42, with an initial steam content of 62 vol % (saturated) for the first H_2 release (Phase 1 of the test), onset of recombination was observed at about 1.2 vol % H_2 at PAR inlet. H_2 release yielded catalyst temperatures up to 550 °C and a H_2 concentration at PAR inlet up to 5 vol %. The oxygen surplus ratio decreased to $\Phi = 2.6$ during the release period of Phase 1. In Phase 2, H_2 release duration was longer and the maximum H_2 concentrations were higher (9.5 vol % at PAR inlet and 10 vol % in the vessel volume). The maximum catalyst temperature yielded 730 °C. The recombination rate and the H_2 depletion efficiency measured was in the same range as for the comparable tests of the preceding HR-test series (HR-14 to HR-16) with NIS-PAR.

The effect of O_2 surplus ratio on H_2 recombination rate and catalyst surface temperatures was clearly visible. As soon as the O_2 surplus ratio falls below 2, both H_2 recombination rate and catalyst surface temperatures decrease drastically.

For test HR-41 (initial steam concentration 39.7 vol %, superheated) and HR-42 (initial steam concentration 62 vol %, saturated) no ignition event was observed. PAR catalyst temperatures were high enough for ignition for both tests and the mixture in the vessel bulk was flammable, but an ignition source (glowing particles) did not exist or could not provide the minimum required ignition energy for the steam rich and oxygen lean mixtures (late in Phase 1 and Phase 2 of the experiments, when H_2 concentrations and catalyst temperatures were high).

For previous NIS-PAR tests performed In the THAI facility [Kan 2009], bulk ignition with (very moderate) pressure increase was observed only for tests with initial steam concentrations up to 25 vol % and (relatively low) H_2 concentrations up to 7.2 vol % at PAR inlet and up to 5.8 vol % in the vessel bulk. Oxygen surplus ratios measured at that point of time were 3.7 and 4, respectively. For tests HR-41 and HR-42 with initial steam concentrations of 40 vol % and 62 vol %, respectively, no ignition event was observed, though the oxygen surplus ratios were in the same range, except in the late phase of test HR-42 for which it was < 2.

Numerical analysis of Test HR-35

For test HR-35, a blind and open benchmark exercise was performed, [Fre 2013].

Results of blind simulations

Five institutions participated in the blind simulation exercise and a total of six simulation results were submitted for evaluation. Thereby lumped parameter codes (MELCOR, COCOSYS and GOTHIC) have been used as well as CFD codes (GASFLOW and CFX). The internals of the PAR have either been resolved within the nodalisations or modelled by specific code modules, or by black box type models. The physics of the PAR were modelled using REKO-DIREKT or a model based on it, the AREVA correlation, similar types of correlations or self-developed correlation functions. Thereby the simulation benchmark addresses a large variety of different approaches which are used by today to analyse the containment conditions during accident scenarios where hydrogen has been released.

Within the coupled CFX simulation by participants from FZJ and IRSN the recombination was well predicted. In the GASFLOW simulations from KIT both type of correlations (AREVA and the fitted correlation) overpredicted recombination of the PAR device. The MELCOR simulations done by the participant from UJV which also applied the AREVA correlation was successful in predicting the natural convection inside the PAR, but overestimated the recombination rate as well. Nevertheless the interaction between the PAR induced natural convection and the gas vessel atmosphere is predicted in good quality.

The COCOSYS simulation by GRS delivered good results for the PAR internal behaviour, especially the dependency of the recombination rate on the aspirated hydrogen and oxygen amount is captured very well. The GOTHIC simulation of AECL showed a successful modelling for the first minutes of the start-up of the recombiner, but underestimated the recombination efficiency by the time where sufficient oxygen becomes available. The reduced amount of recombination in this simulation results into a smaller heat release, a decrease of the natural convection and as a consequence to a slower aspiration of hydrogen and oxygen.

The main features of the simulated recombination and its effect on the surrounding atmosphere revealed significant differences among the codes and the models applied. At least for models which rely on the AREVA correlation there is still a substantial need for improvement under low oxygen conditions and probably on the correlation itself as well.

Results of open simulation

Three institutions participated in the open simulation exercise and a total of four simulation results were submitted for evaluation. Thereby GOTHIC has been used as well as CFD codes GASFLOW and CFX. From the two GOTHIC simulations only the one without continuous steam injection was selected in order to be comparable with the other two model approaches.

None of the changes which have been introduced between blind and open simulation phase resulted into a noticeable difference in the prediction of the hydrogen and oxygen mixing after the individual gas injection.

In terms of the recombination rate all three models are able to reproduce the experimental data with acceptable quality whereas the results from GASFLOW improved most between blind and open simulations. By changing the modelling constants in the fitting correlation the GOTHIC model was able to improve the modelled reaction rate of the PAR which then ended up in a good prediction of the peak recombination value, compared to its blind simulation counterpart.

4.3 Tests Iod-25 and Iod-26

The interaction between molecular iodine and aerosols (non-reactive or reactive) is one of the issues lacking experimental data for code validation purposes, and for this reason no models have been incorporated in severe accident containment codes to model quantitatively the gaseous iodine

depletion by adsorption on aerosol particles. Therefore, it is necessary to investigate this interaction in large scale experiments with well controlled boundary conditions. The experimental data will contribute to quantify the extent of gaseous iodine depletion by aerosols as compared to depletion by the large surface areas of decontamination paint in the containment. By this, uncertainties related to source term quantification in the severe accident containment codes are reduced.

Initial conditions and test parameters as specified and as achieved are compiled in Table 0-3. More details can be found in the respective Technical Reports [Fun 2014b] and [Fun 2014c].

The test procedure for tests Iod-25 and Iod-26 consisted of a preconditioning phase and three main test phases. The specified thermal hydraulic conditions in the test vessel were established in the preconditioning phase prior to the three test phases. Iodine labeled with I-123 tracer (I-123 activity: 1.1E9 Bq) was released into the THAI vessel at the beginning of Phase 1. At the beginning of Phase 2, either SnO_2-aerosol (test Iod-25) or Ag-aerosol (test Iod-26) was released into the THAI-vessel.

The three test phases were as follows:

Phase 1: Iodine interaction with surfaces, equilibrium gas-walls (reference condition)

Phase 2: Iodine/aerosol interaction during rapid aerosol release

Phase 3: Iodine/aerosol interaction during constant aerosol concentration

Test Iod-25

The evolution of the iodine concentration during the three test phases is shown in Fig. 0-6. The figure provides the iodine concentration for the gaseous and the aerosol-bound iodine (the aerosol-bound iodine measured during the I_2 phase is due to a limited chemical adsorption of gaseous iodine on the aerosol filter and to I_2 adsorbed on background aerosol in the vessel atmosphere). The gaseous I_2 concentration is significantly higher than the aerosol-bound iodine, even during the aerosol phases with high SnO_2 concentration. The concentrations of gaseous I_2 and aerosol-bound I_2 differ by more than one order of magnitude.

THAI Iod-25: Maypack filters: Gaseous and aerosol-borne iodine

THAI Iod-25: Interaction of I_2 with SnO_2 aerosol particles
Iodine concentrations in the gas phase measured by gas scrubbers
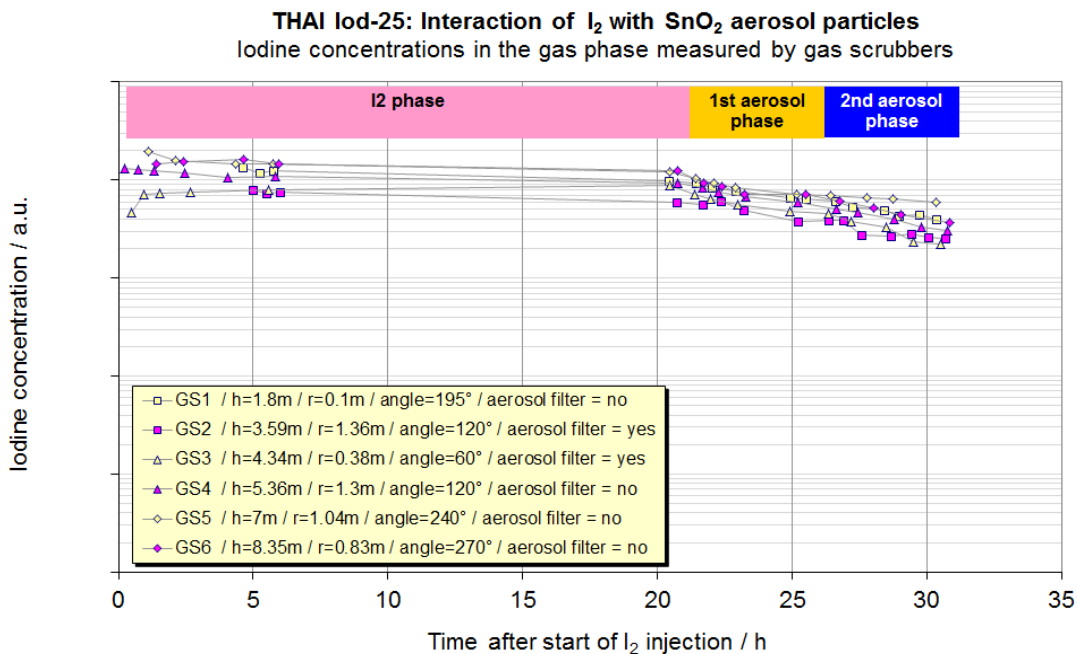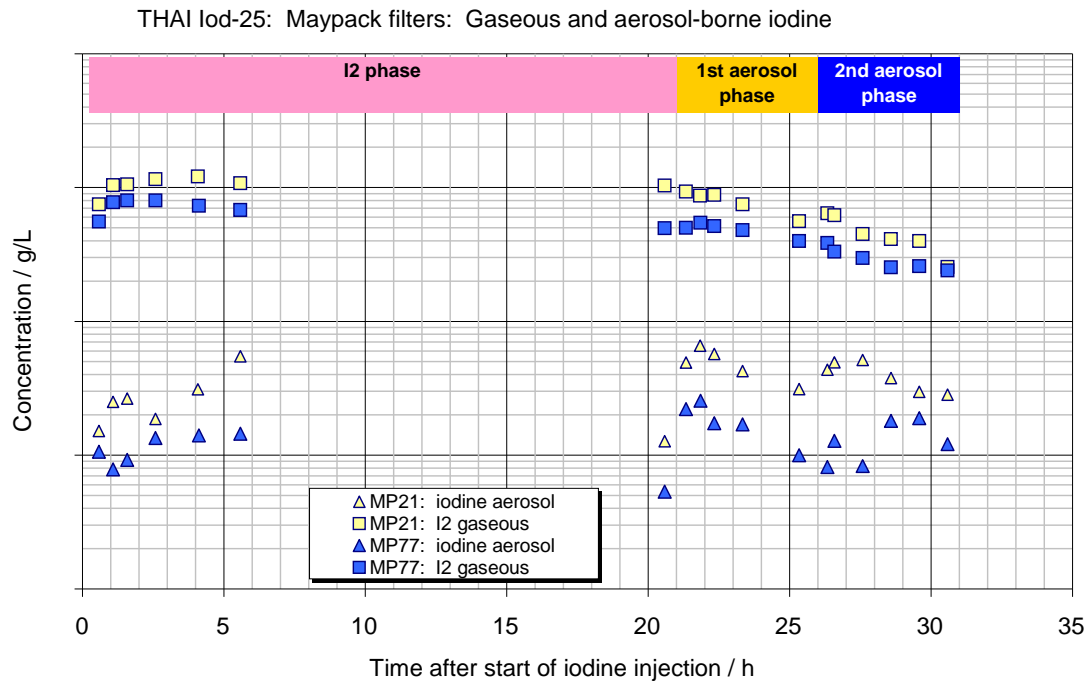
Fig. 0-6: Iod-25: Concentrations of gaseous and aerosol-bound iodine as measured with the Maypack filters (upper) and decrease of gas-borne iodine by physisorption of I_2 onto settling SnO_2 particles measured with gas scrubbers (lower)

During phase 1, the concentration of the injected atmospheric iodine decreases slowly due to adsorption onto steel surfaces. During the aerosol phases, the decrease of the atmospheric iodine is accelerated due to adsorption on the SnO_2 particles which are depositing faster than the process of I_2 adsorption onto steel, thus removing iodine faster from the atmosphere.

During phase 2, the iodine concentrations decrease by a factor of about 2. In the same time window, the measured aerosol concentration decreases by a factor of about 5. The clearly slower iodine decrease during this phase indicates that the removal of iodine from the vessel atmosphere by settling SnO_2 particles with physisorbed iodine is weak.

From the measurements, a deposition velocity for the iodine adsorption on SnO_2 was calculated from the two aerosol test phases, assuming a total SnO_2 particle surface of 118 m^2 and no I_2-resuspension. A different approach to derive a rate constant for the depletion of gaseous I_2 by adsorption on both the steel surface (100 m^2) and the SnO_2 particles (118 m^2), assuming also the same reactivity of both materials to I_2, yields a similar value. Given the uncertainties of the assumptions, both estimates of the I_2 / SnO_2 reaction rate are agreeing reasonably.

Test Iod-26

The iodine speciation measurements with Maypack filters in test Iod-26 clearly show that the air-borne iodine is pre-dominantly aerosol-bound during the aerosol phases, Fig. 0-7. In contrast, iodine was pre-dominantly gaseous in Iod-25. The measurements revealed, that the conversion of gaseous I_2 into aerosol-bound iodine was already observed in the first samplings taken only a few minutes after the first Ag-aerosol injection. This means that a fast and efficient chemisorption took place.

The depletion of I_2 from the vessel atmosphere is by a factor of 25 faster for the Ag-aerosol as compared to the SnO_2 aerosol. For the reactive Ag, aerosol settling dominates the overall iodine depletion, whereas for the inert SnO_2 the overall iodine depletion is dominated by the weak / reversible physisorption of gaseous I_2 onto aerosol particles and steel surfaces. This factor of 25 is specific to tests Iod-25 / Iod-26, and does not account for the different particle surface areas in the two tests.

THAI Iod-26: Interaction of I$_2$ with Ag aerosol particles
Iodine concentrations in the gas phase measured by gas scrubbers
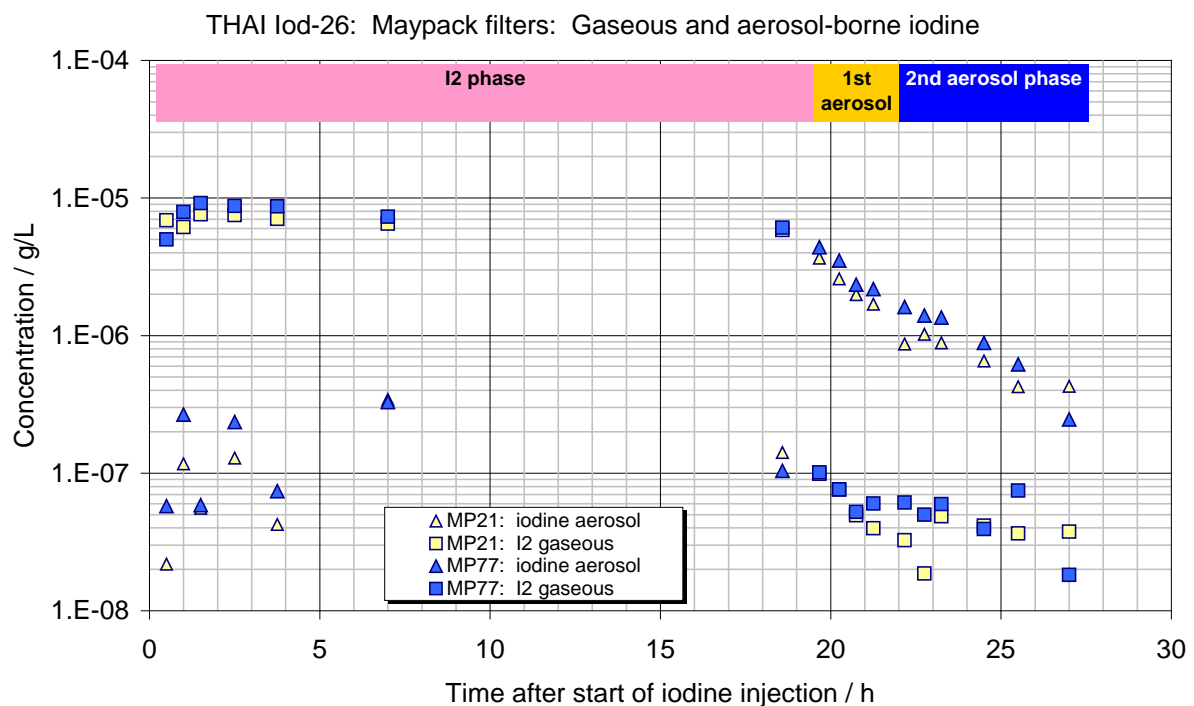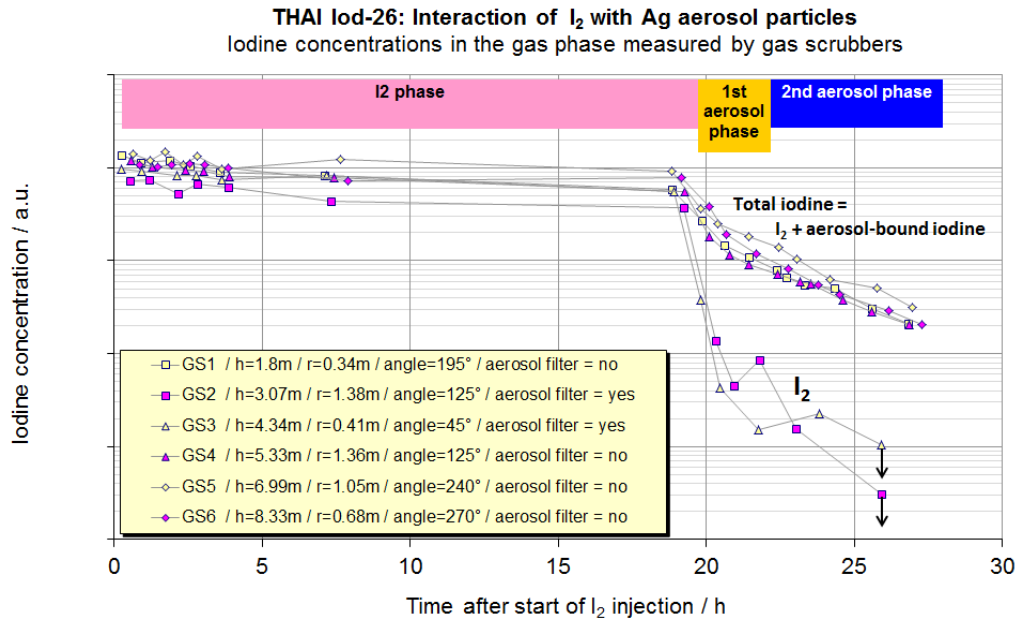
Fig. 0-7: Iod-26: Iodine species concentrations measured with Maypack filters (upper) and gas scrubbers (lower): Decrease of gas-borne iodine by chemisorption of I$_2$ onto settling Ag particles.

In Iod-26, I$_2$ adsorption onto reactive Ag particles leads to formation of silver iodide molecules at the surface. This is a strong bond of iodine to the surface, called chemisorption. Iodine desorption only takes place upon additional impacts such as temperature increase or radiation dose, i.e. chemisorption is practically "non-reversible". The interaction of I$_2$ with "inert", non-reactive SnO$_2$ is much weaker, there is no chemical reaction but only weak adsorption/desorption of I$_2$ molecules until an equilibrium between gaseous I$_2$ and particle-adsorbed I$_2$ is reached.

A first-order rate constant for I$_2$ deposition onto Ag particles has been determined from the initial decrease of the I$_2$ concentrations in test Iod-26 as measured by gas scrubbers which had been provided especially for the measurement of gaseous iodine. Assuming a constant Ag surface area of 75 m^2 in the 60 m^3 vessel, the rate constant determined is similar to typical I$_2$ mass transfer rate constants as known from experiments with containment-typical atmospheres, i.e. the I$_2$ /Ag reaction in Iod-26 was limited by mass transfer. The value is roughly three orders of magnitude faster than the rate determined from the SnO$_2$ test, and this is consistent with the expectation for reactive Ag and inert SnO$_2$ particles.

4.4 Test Iod-29

Test Iod-29 was designed to deliver experimental data on release of gaseous iodine from a flashing jet under high pressure and high temperature thermal hydraulic conditions. More details can be found in the respective Technical Report [Fun 2014a].

Present models for iodine chemistry in nuclear reactor systems are focused on containment conditions, where pressure and temperature are substantially lower than in the reactor cooling circuits. The constitutive model relations (like e.g. the dependency of the iodine partitioning coefficient upon temperature) are validated for containment-typical ranges. The applicability of these relations to the high pressure/high temperature regime is not well known. The proportions of chemical forms of iodine dissolved in water at high temperature may be different from those at lower temperature. There is a general lack of validation for iodine chemistry and transport models under temperatures typical for the primary system thermal hydraulics.

The Iod-29 experiment is oriented towards PWR steam generator tube rupture scenarios, but the data may be applicable also to other scenarios with flashing of primary circuit water. The proposed

experiment shall improve the state of knowledge in this area and provide data for the further improvement of the existing models.

The test started with the flashing, i.e. a release of pressurized water from a pressure vessel (PV) via a droplet separator (efficiency 99.95 %) into the THAI vessel, which involves partial evaporation of the depressurized water. The test conditions as achieved at start and end time of the flashing are given in the table below. More details can be found in the respective Technical Report [Fun 2014a].

Measurements	At the start time of flashing	At the end time of flashing (t = 3.03 min)
Absolute pressure in the THAI vessel	1.497 bar	2.848 bar
Average gas temperature in the THAI vessel	100.28 °C	122.89 °C
Absolute pressure in the PV (incl. hydrostatic pressure)	40.2 bar	38.6 bar
Water temperature in the PV	247.0 °C	245.6 °C

During the flashing period of 3.03 min, about 178 kg of water was discharged from the PV corresponding to a mean mass flow rate of 0.98 kg/s. Out of this, about 41 kg of steam was generated. Based on the generated steam amount, a flashing evaporation rate of approximately 23 % is estimated.

The main result of the iodine measurement in Iod-29 is that no gaseous iodine was measured in the THAI vessel atmosphere after flashing. A single data point which could possibly be interpreted as being a finite concentration of gaseous iodine has to be attributed to an artefact which is caused by cross-contamination during the washing procedure for this specific gas scrubber.

Liquid samples of the pressure vessel water during heat-up, just before and just after flashing were analysed chemically for total iodine amount and iodine speciation. Although 19 g of iodine were injected before heat-up in the molecular form, only the iodide form was found already during heat-up, and iodate as a product of iodine hydrolysis was not detected above the detection limit of 5.4 % with respect to the sum of all iodine species. This indicates that the injected molecular iodine had quickly dissolved and hydrolysed, with intermediate hydrolysis species such as HOI and I_2OH^- reacting quantitatively with the steel wall of the primary vessel or metal ion impurities in the aqueous phase during heat-up to produce the non-volatile iodide form.

4.5 Laboratory tests

The depletion of molecular iodine from an atmosphere by interaction with non-reactive and reactive aerosol was studied in tests Iod-25 and Iod-26. In parallel, a laboratory-scale test programme was run to support the design and procedure of THAI-tests Iod-25 and Iod-26, and to study a number of chemical parameters that could influence the iodine/aerosol interaction such as temperature, aerosol material and gaseous iodine concentration. Test results will enlarge the data base for model development and validation. More details can be found in the respective Technical Report [Lan 2012].

The aerosol particles selected reflect the different chemical behaviour of these substances:

- Fe was considered as: "metallic, I_2 reactive, non-soluble",
- Ag   as: "metallic, I_2 reactive, non-soluble",
- SnO_2 as: "oxidic, non-reactive, non-soluble" and
- CsI   as: "soluble, wet/hygroscopic aerosol".

The test aerosols were deposited on a coupon and were exposed to a carrier gas containing I_2. The absorption of I_2 by the aerosol particles was measured online. All tests were performed at ambient pressure. Temperature was varied systematically. The gaseous iodine concentration was kept in the range between 1.0E-06 and 2.6E-6 mol/L, the gas flow rate through the tube was kept at 1.2 to 1.4 L/min. The temperature of the gas atmosphere was 80 °C, 120 °C and 140 °C.

For additional tests with Ag-aerosol the gas composition (steam/air and dry nitrogen) and the concentration of the gaseous iodine were varied.

At 80 °C Ag and CsI are the most reactive in an inert, dry atmosphere and have the highest iodine loading on the surface. The remarkable desorption of iodine from CsI during the desorption phase indicates that the I_2 is physisorbed on the CsI in inert, dry conditions and therefore easily released after terminating the I_2 supply. Ag shows a similar reactivity towards iodine, but the data indicate that the iodine is chemisorbed on the silver surface since no release of iodine could be measured. The observed iodine loading on Fe aerosols is by about one order of magnitude lower than for Ag and CsI, i.e. Fe is less reactive than Ag and CsI. The very low release of iodine from the Fe surface is explained by the predominance of chemisorption over physisorption. No significant iodine loading was observed for the oxidic SnO_2 particles.

At 120 °C, a deposition of gaseous iodine on CsI could not be measured supporting the hypothesis that the nature of interaction of I_2 with CsI under inert, dry conditions is physisorption. But Ag still shows a high tendency to adsorb iodine from the gas phase with an iodine loading being the same as for the 80 °C tests. This evidences chemisorption for the interaction of I_2 with Ag. The two tests with Fe powder at 120 °C revealed a weaker but still observable interaction with I_2. The low release of iodine in the desorption phase supports the assumption of a chemisorption mechanism. The final iodine loadings on iron were lower by a factor of about 3-5 than at 80 °C, demonstrating a temperature dependence of this interaction.

At 140 °C, any iodine loading on the aerosols could only be measured in the tests with Ag, being at the same level as for 80 °C and 120 °C. With all other materials, no signals above the detection limits could be measured.

Adsorption rates had been derived from the initial increases of the specific iodine loadings. The data do not show a temperature dependency, which is also known from I_2 adsorption onto other surfaces, such as decontamination paint.

The rate constants for I_2 adsorption onto decontamination paint determined for a variety of paint types and manufacturers agree well with the adsorption rate of the I_2 / Ag aerosol system obtained from the present tests.

Table 0-1:   HD-tests: Initial test conditions as specified and as measured

Test parameters		HD-30	HD-7	HD-31	HD-31-SE	HD-32	HD-32.1	HD-33	HD-22	HD-34	HD-35	HD-23
Vessel pressure [bar]	spec.	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
	meas.	1.493	1.480	1.470	1.495	1.474	1.492	1.506	1.487	1.495	1.497	1.465
Gas temperature [°C]	spec.	20	25	90	90	90	90	90	90	90	90	90
	meas.	24.5	17	89.8	89.5	89	89.5	89.6	92	90.0	89.4	91
Hydrogen content [vol %]	spec.	10.0	10.0	10.0	-	10.0	10.0	10.0	10.0	10.0	12.0	12.0
	meas.	9.5	9.9	9.6	-	7.5	9.2	9.1	9.9	9.1	11.7	12.0
Steam content [vol %]	spec.	-	-	25	25	25	25	25	25	25	25	25
	meas.	-	-	31	27	46	25.3	25.1	25	25.1	24.9	25
Spray water Temperature [°C]	spec.	20	-	20	20	90	90	90	-	20	90**	-
	meas.	***	-	***	***	***	***	***	-	***	***	-
Spray nozzle*		s		s		s	s	s		l	s	
Burn direction		↑	↑	↑		↑	↑	↑	↑	↑	↓	↓

* s = small droplets; l = large droplets
** = spray operation 60 s prior to ignition
*** = see respective diagrams in the Technical Report [Gup 2014c]

Table 0-2:   HR-tests: Initial test conditions as specified and as measured

Test ID	p bar		T °C		Steam content vol %		O_2 content vol %		H_2 content vol %		PAR type
	spec.	meas.	spec.	meas.	spec.	meas.	spec.	meas.	spec.	meas.	
HR-33	1.5	1.49	105	104.1	-	-	< 0.5	< 0.5	4	4.6*	AREVA
HR-34	2.0	1.87	105	104.4	40	41	< 0.5	< 0.5	4	4.7*	AREVA
HR-35	3.0	3.12	117	118.8	60	58	< 0.5	< 0.5	4	4.4*	AREVA
HR-36	1.5	1.48	69	71	20	19	< 0.5	< 0.5	10	10.1*	AREVA
HR-37	1.5	1.49	71.2	91.4	30	30	< 0.5	< 0.5	10	10.1*	AREVA
HR-38	2.0	1.91	105	104.6	40	40	< 0.5	< 0.5	4	3.7	NIS
HR-39	3.0	2.99	117	117.5	60	61	< 0.5	< 0.5	4	3.8	NIS
HR-40	1.5	1.49	117	115.5	-	-	air	air	-	-	NIS
HR-41	2.0	1.99	105	104.4	40	39.5	air	air	-	-	NIS
HR-42	3.0	3.0	117	118	60	62	air	air	-	-	NIS

* = average concentration above sump compartment

Table 0-3:   Tests Iod-25 and Iod-26: Initial conditions and test parameters as specified and as measured

Test parameter	Specified	Measured	Specified	Measured
	Iod-25		**Iod-26**	
Preconditioning				
Vessel pressure [bar]	1.30	1.36	1.30	1.32
Gas temperature [°C]	70	68.2	70	69.87
Wall temperature [°C]	70	69.8	70	71.00
Test phase 1: Iodine release (duration 15 min)				
Released iodine mass (g)	1.0	1.0	1.0	1.0
Test phase 2: Rapid aerosol release				
Aerosol injection duration (min)	10	10	6	6
Aerosol mass concentration (g/m^3)	~ 2.0	2.0 (max.)	~ 1.0	1.28 (max.)
Test phase 3: Constant aerosol concentration				
Aerosol injection duration (min)	300	300	~ 300	337
Aerosol mass concentration (g/m^3)	1 - 1.5	1.15 (aver.)	0.5	0.54 (aver.)

Table 0-4:   Test Iod-29: Initial and post-test conditions in the THAI vessel and in the pressure vessel (PV)

Test parameter	Specified	Measured	Measured
Preconditioning			**End of Flashing***
THAI vessel pressure [bar]	1.50	1.497	2.848
THAI vessel fluid temperature [°C]	100	100.3	122.9
THAI vessel wall temperature [°C]	100	101.45	-
PV pressure [bar]	40	40.17	38.64
Water temperature in PV [°C]	250	247	245.6
Chemical boundary conditions (pressure vessel)			
pH at ambient conditions	7.2	7.0 - 7.2	
Buffer concentration [mol/L] NaOH KH_2PO_4	 6.0E-3 1.0E-2	 6.67E-3 1.0E-2	
I_2 injected into PV water [g] I_2 inv. before flash. based on sampl. [g]	19.0	19.1 13.9	
Iodine concentration in PV water [mol/L] Based on sampl. [mol/L]	1.0E-4	1.0E-4 0.73E-4	

* Flashing duration: 3.03 min

5. CONCLUSIONS

5.1 HD-experiments

- For a H_2 concentration up to 10 vol %, all HD-tests with spray and upward combustion exhibited clearly a suppressing effect with respect to peak pressures and peak temperatures which becomes more pronounced for increasing steam content. For test HD-35 with a H_2 concentration of 12 vol % and 25 vol % steam and downward burn direction, the peak pressure exceeded that of the quiescent (without spray) test by 10 %. Test HD-35 was the only test for which spray operation proved to be disadvantageous with respect to peak pressure.

- For the HD-tests with spray, the combination of an elevated initial gas temperature and a steam concentration of 25 vol % (superheated) provided the most dominant influence on peak pressure and peak temperature reduction.

- Flame quenching by cooling of the reaction zone (locally even below the auto-ignition temperature required to sustain chain reaction) is the dominant effect of the spray. In addition, the steam produced by vaporisation of water droplets acts as a heat sink (higher specific heat than air) which reduces peak pressure and peak temperature.

- By use of (cold) spray in a saturated atmosphere steam condensation takes place and may render non-flammable mixtures flammable. Combustion near the flammability limit is benign with very moderate pressure increase for low and intermediate H_2 concentrations. Early spray activation is favourable, since for an accident scenario with a H_2 concentration exceeding 8 vol % (in ignitable atmosphere) despite PAR operation, several sources of ignition are present in a containment building, e.g. random ignition (TMI, non-nuclear industry experience), by an operating PAR, or by glow plugs. For this case (H_2 concentration in the range of 8 vol %) the flame suppressing effect, i.e. peak pressure reduction, of the spray is beneficial.

5.2 HR-experiments

Tests HR-33 to HR-35 (AREVA PAR)

From the results of the experiments HR-33 to HR-35, the following conclusions can be drawn:

- PAR starts operation at very low oxygen concentrations (O_2 concentration at PAR inlet < 0.5 vol %), which corresponds to the O_2 surplus ratio far below the stoichiometric oxygen surplus ratio of $\Phi = 1$.

- Three H_2 recombination regimes are observed during the tests. For an oxygen surplus ratio value $\Phi \leq 1$, an oxygen lean gas mixture exists at the PAR inlet and the rate of oxygen diffusion through the catalyst boundary layer mainly governs the recombination rate. For $1 < \Phi \leq 2$, a transition in hydrogen recombination rate occurs and H_2 recombination is governed by both oxygen and hydrogen diffusion through the catalyst boundary layer. For oxygen surplus ratio value $\Phi > 2$, a hydrogen-lean gas mixture prevails at the PAR inlet and the rate of recombination is mainly governed by the rate of hydrogen diffusion through the catalyst boundary layer.

- Test data indicate that for a given H_2 concentration of approximately 4 vol % at PAR inlet and an O_2 surplus ratio below 2, the hydrogen recombination rate as well as the hydrogen depletion efficiency γ increase almost proportional to the oxygen surplus ratio (or to oxygen concentration at the PAR inlet). Above an oxygen surplus ratio of $\Phi = 2$, further increase of hydrogen depletion efficiency γ becomes very small and remains between 50-70 %, and is hydrogen controlled.

- The comparison of the measured recombination rates with those calculated with the empirical correlation provided by the PAR vendor AREVA reveals some need for further improvement of the correlation for the oxygen starvation regime.

Tests HR-36 to HR-37 (AREVA PAR)

In the case of reduced O_2 content in air (oxygen surplus ratio $\Phi < 1.5$ for both tests), but with the H_2 concentration sufficiently high, ignition in the vessel occurs if

- the steam content is below a certain (inertisation) limit for a given ratio of O_2/N_2,

- the oxygen transient renders the gas mixture flammable at PAR inlet, above the catalyst within the PAR, and in the vessel,

- the preceding recombination had produced a catalyst surface temperature high enough to ignite the mixture,

Since ignition in the vessel occurs once the O_2 enrichment transient causes the mixture to approach the flammability limit at all of the a. m. three locations, slight deviations in gas composition or temperatures can result in either ignition or no ignition.

Tests HR-38 to HR-39 (NIS PAR)

From the results of the experiments HR-38 to HR-39, the following conclusions can be drawn:

- PAR onset indicated by a marked rise in catalyst temperature occurs at oxygen concentrations at PAR inlet < 0.5 vol %, far below an oxygen surplus ratio of $\Phi = 1$.

- Three H_2 recombination regimes are observed during the tests. For an oxygen surplus ratio value $\Phi \leq 1$, an oxygen lean gas mixture exists at the PAR inlet and the rate of oxygen diffusion through the catalyst boundary layer mainly governs the recombination rate. For $1 < \Phi \leq 2$, a transition in hydrogen recombination rate occurs and H_2 recombination is governed by both oxygen and hydrogen diffusion through the catalyst boundary layer. For oxygen surplus ratio value $\Phi > 2$, hydrogen lean gas mixture prevails at the PAR inlet and the rate of recombination is mainly governed by the rate of hydrogen diffusion through the catalyst boundary layer.

- For a given H_2 concentration of approximately 4 vol % at PAR inlet and an O_2 surplus ratio below 2, the hydrogen depletion efficiency γ increases almost proportional to the oxygen concentration at PAR inlet. Above an oxygen surplus ratio of $\Phi = 2$, further increase of hydrogen depletion efficiency γ becomes very small; γ remains between 40-60 %, and is hydrogen controlled. Comparison with other NIS-PAR tests indicate that these results are almost independent from initial test conditions, e.g. pressure, temperature, steam and hydrogen contents.

Tests HR-40 to HR-42 (NIS PAR)

With respect to PAR performance it was found, that:

- The onset of H_2 recombination – indicated by the first recognizable increase of PAR catalyst temperatures - occurs at H_2 concentrations ranging between 0.8–1.2 vol %. Due to the hydrophobic coating, onset of recombination is only slightly faster in non-condensing

atmosphere than at saturation. Onset of recombination is faster for new than for used catalyst.

- The effect of O_2 surplus ratio on H_2 recombination rate and catalyst surface temperatures was clearly visible. As soon as the O_2 surplus ratio falls below $\Phi = 2$, both H_2 recombination rate and catalyst surface temperatures decrease significantly.

With respect to PAR ignition behaviour it was found, that:

- In HR-40 test (dry mixture), multiple ignitions were observed outside the PAR in parts of the vessel with flammable gas mixture. Test data indicate that ignition is initiated by "glow worms" for a H_2 concentration of 4.1 vol %. Since the ignitions took place in H_2-lean gas mixtures, combustion was benign with low pressure peaks only.

- For steam-rich gas mixtures (HR-41 and HR-42), no ignition was observed even though the gas mixture was in the flammable range outside the PAR in the vessel. No "glow worms" had been observed during these tests. One of the potential reasons might be the minimum required ignition energy for the steam-rich mixture, which could not have been provided by the "glow worms".

5.3 Tests Iod-25 and Iod-26

Test Iod-25

The aerosol-bound iodine fraction was found to be low during Iod-25, however, the atmospheric iodine concentration determined from numerous samplings from the vessel atmosphere decreases clearly due to adsorption on "inert" SnO_2 aerosol and subsequent depletion by aerosol sedimentation. The evaluation of these data with respect to application in severe accident containment models will require detailed kinetic models on the iodine and on the aerosol behaviour, as well as a coupled iodine / aerosol interaction model. The iodine loading onto a steel deposition coupon coated with a thin SnO_2 aerosol layer is similar to that on the stainless steel coupon.

Test Iod-26

The gaseous iodine concentration is efficiently decreased due to the removal by the reactive Ag aerosol. Sedimentation of iodine loaded aerosols is the dominating process for the removal of iodine from the vessel atmosphere. The depletion of gas-borne iodine during both aerosol phases is 25 times faster as compared to test Iod-25 with the inert SnO_2 aerosol. Adopting a number of assumptions, an I_2 / Ag adsorption velocity of 1.2E-3 m/s is derived, which means that the reaction is limited by mass transfer. Thus, the effect of the removal of gaseous molecular iodine from the vessel atmosphere by interaction with an injected reactive aerosol was quantified and the data can be used for further code development.

Test Iod-25 and Iod-26 modelling aspects

Tests Iod-25 and Iod-26 can be considered to provide the bounding cases for the effect of I_2 removal from atmospheres by interaction with aerosol, the reactive Ag aerosol providing the maximum and the inert SnO_2 the minimum removal rates under the given boundary conditions of the tests. Real containment aerosol can be expected to show removal effects between these bounding cases.

Tests Iod-25 and Iod-26 were generic tests on the investigation of iodine-aerosol interaction processes not quantified to date. For the test conditions as used in test Iod-25 and Iod-26, the results can be used in the respective models. For different conditions with respect to e. g. thermal hydraulics or concentrations, quantitative data has to be determined. First indications about the dependency of the iodine-aerosol interaction from the prevailing thermal hydraulic conditions, e.g. relative humidity, are provided by the corresponding laboratory tests.

Therefore, the determined deposition rate should not be transferred directly to other tests or to general application in severe accident containment codes without specific assessment.

5.4 Test Iod-29

The iodine measurements in the THAI vessel (gas space and sump) and in the primary vessel consistently show that there was no gaseous iodine released from the flashing jet under the investigated test conditions. The iodine mass balance was found to be closed.

Phosphor concentration measurements (from KH_2PO_4 used as pH buffer together with NaOH in the pressure vessel water) also acted as a tracer to confirm if droplets containing iodine left the droplet separator. Measured concentrations in the THAI vessel are at a very low concentration level, showing that the droplet separator was very efficiently removing droplets, and that any iodine in gas phase samplings above the iodine detection limit would have to be attributed to the gaseous form.

The observation that no gaseous iodine existed in the THAI vessel is consistent with measurements on iodine speciation in the water of the pressure vessel, sampled during heat-up just before and just after the flashing process. Although 19 g of molecular iodine were injected before heat-up, only the iodide form was found, and iodate as a product of iodine hydrolysis was not detected above the detection limit of 5.4 % with respect to the sum of all iodine species. This indicates that the injected molecular iodine reacted quickly with the steel wall of the primary vessel during heat-up and thus, had produced the non-volatile iodide form. The analytic work performed on this experiment also indicated lack of validated iodine chemical models for the investigated DBA relevant test conditions.

5.5 Laboratory test

The laboratory tests on I_2 / Ag interaction within the current THAI-2 project are qualitatively consistent with Iod-26 results, especially with respect to the iodine loading on the Ag surface. They provide additional data for parameter variations like relative humidity, iodine concentration in the gas phase, particle mass distribution, and for the behavior of other aerosol material like CsI and Fe. The rates measured in the laboratory tests and the THAI tests include different processes and are thus not directly comparable. This is a further requirement for a detailed iodine / aerosol interaction model.

5.6 Application of experimental results to accident scenarios

In this section, the application of the experimental results as achieved under the test conditions and test parameters investigated is discussed.

HD-Tests

For almost all of the tests, the operation of the spray reduces peak pressure and peak temperature due to local flame quenching. Only for the test for which flame travel and spray induced flow had the same direction (downward burn), combustion peak pressure was about 10 % higher than for the reference test without spray, but clearly below the respective AICC-pressure. This peak pressure increase is not excessive and considering all other beneficial effects of the spray, spray operation can be regarded as favourable.

The spray nozzles used for the tests provided the same droplet size distribution and mass flow rate as those used in real containments. These nozzles produce a sufficient droplet surface to support flame quenching for the given mass flow rate and thermal hydraulic conditions. Solely for the purpose of suppression of a H_2 deflagration, a finer droplet size distribution could be considered, since this would provide

- for the same mass flow rate more droplet surface,
- due to a smaller falling velocity a higher mass of airborne droplets, and

- a flow field less turbulent.

HR-Tests

Both PAR types tested start operation reliably also at very low oxygen concentrations, a situation which may occur in the late phase of a severe accident where combustible gases are already present in the containment atmosphere. Ignition occurs once the mixture becomes combustible in the transient starting from the oxygen starvation regime, and produces moderate peak pressures only. This is the same behaviour as was found for the transient with a mixture starting from the "hydrogen starvation" regime.

Correlations used for capacity design of PAR can be validated also for the oxygen starvation regime, and for this regime an ignition criterion is now available.

Tests Iod-25 and Iod-26

The adsorption of I_2 by the inert SnO_2-aerosol is small considering other potential sinks of gaseous I_2 (e. g. decontamination paint) and the influence of this process on the radiological source term is expected to be small as well.

The adsorption of I_2 by the reactive Ag-aerosol is high and in the same range as the I_2 adsorption by decontamination paint under the thermal hydraulic conditions prevailing in test Iod-26 and the depletion of gaseous iodine from the atmosphere is governed by the aerosol settling process. For the application of this data to severe accident scenarios, further validated source term codes are to be used.

By laboratory tests, the results of tests Iod-25 and Iof-26 have been qualitatively confirmed for different thermal hydraulic conditions and concentrations.

From a pure chemical point of view, the high efficiency of Ag to bind gaseous iodine and to convert it into an aerosol can be used as an accident management measure.

The results may be of significance for the design of filtered venting devices and their operation strategy.

Test Iod-29

In test Iod-29, a PWR-design basis accident condition (rupture of a steam generator tube under reactor shutdown) was simulated with respect to I_2 release from the primary to the secondary side with a pressure difference of 40 bar. No gaseous iodine was measured in the gas plenum of the THAI vessel (which simulated the secondary side of the steam generator), since obviously the I_2 present in the pressure vessel water reacted with the pressure vessel steel walls and formed non-volatile iodate under the prevailing thermal hydraulic conditions.

The predicted release of gaseous iodine based on high temperature iodine/water chemistry was overestimated. Improved post-test calculations confirmed that under the investigated conditions a significant gaseous iodine fraction should not exist, which indicates lack of validated iodine chemical models for the investigated DBA relevant test conditions.

6. REFERENCES

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[Fre 2013]   Freitag, M.; Sonnenkalb, M.: Comparison report for blind and open simulations of HR-35. OECD-NEA THAI-2 Project, Report No. 1501420 – HR35 – AWG (VB) (December 2013), Becker Technologies GmbH, Eschborn, Germany, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Köln, Germany

[Fun 2014a]   Funke, F. et al.: Iodine release from a flashing jet – Test Iod-29. OECD-NEA THAI-2 Project, Report No. 1501420–Iod-29-TR (June 2014), Becker Technologies GmbH, Eschborn, Germany

[Fun 2014b]   Funke, F. et al.: Deposition of molecular iodine on aerosol particles Iod-25 (Test using non-reactive aerosol SnO_2). OECD-NEA THAI-2 Project, Report No. 1501420–Iod-25-TR (July 2014), Becker Technologies GmbH, Eschborn, Germany

[Fun 2014c]   Funke, F. et al.: Deposition of molecular iodine on aerosol particles Iod-26 (Test using reactive aerosol Ag). OECD-NEA THAI-2 Project, Report No. 1501420–Iod-26-TR (November 2013), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014a]   Gupta, S.; Langer, G.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-33-35 (June 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014b]   Gupta, S., Schmidt, E.; Langer, G.: Effect of reduced oxygen content in air on ignition potential of Passive Autocatalytic Recombiners (PAR); Tests HR-36 and HR-37. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-36-37 (June 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014c]   Gupta, S. et al.: Hydrogen combustion during spray operation; Tests HD-30 to HD-35. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HD-30-35 (September 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014d]   Gupta, S.; Schmidt, E.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-38-39 (December 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014e]   Gupta, S.; Schmidt, E.; Langer, G.: Hydrogen recombiner tests with NIS-PAR (Tests HR-40, HR-41, HR-42). OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-40-42 (July 2014), Becker Technologies GmbH, Eschborn, Germany

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[Kan 2009]   Kanzleiter, T.: Recombiner Tests HR-14 to HR-16. (Tests using a NIS PAR) Areva, AECL and NIS PAR Comparison. Quick Look Report Hydrogen Reactor Safety Research Project 150 1326 OECD-NEA, THAI Project Report No. 1501326-HR-QLR-4 (October 2009), Becker Technologies, Eschborn, Germany

[Kan 2010a]   Kanzleiter, T.; Langer, G.: Hydrogen deflagration tests in the THAI test facility. Reactor Safety Research Project 150 1326, OECD-NEA THAI Project, Technical Report No. 150 1326-HD-2 (January 2010), Becker Technologies GmbH, Eschborn, Germany

[Kan 2010b]   Kanzleiter, T, et al: Hydrogen and fission product issues relevant for containment safety assessment under severe accident conditions. Project final report 1501326-FR 1, June 2010

[Lan 2012]   Langrock, G.: Laboratory-scale tests on the interaction of gaseous I_2 with aerosol particles. OECD-NEA THAI-2 Project, Repot No. PTCC-G/2012/en/0315 (December 2012), AREVA GmbH, Erlangen, Germany

[Mey 2012]   Meynet, N.; Bentaib, A.; Candan, O.: Impact of oxygen starvation on PARs hydrogen ignition limit. 3rd PRG Meeting of the OECD THAI-2 Project, Paris, 19-20 November 2012

[Web 2014]   Weber, G.: Conclusions drawn for Iod-25 and Iod-26 experiments on iodine deposition on aerosols. OECD-NEA THAI-2 Final Seminar, Frankfurt, 18-19 November 2014

APPENDIX A: LIST OF PROJECT REPORTS

The below listed reports are restricted to the project signatories. However, the Management Board of the THAI Project has agreed that these Quick Look Reports and Technical Reports will be disclosed for non-signatories after 31 July 2017 and can be obtained after this date upon request to the NEA secretariat.

Project deliveries

Quick Look Reports (QLR)

[Gup 2012/1]   Gupta, S.; Balewski B.; Kühnel A.; Funke F.; Zeh P.; Burchardt C.; Stahl A.: Deposition of molecular iodine on aerosol particles - Iod-25 Test using non-reactive aerosol SnO2. OECD-NEA THAI-2 Project, Report No. 1501420–Iod-25-QLR (January 2012), Becker Technologies GmbH, Eschborn, Germany, AREVA GmbH, Erlangen, Germany

[Gup 2012/2]   Gupta, S.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–HR-33/34-QLR (October 2012), Becker Technologies GmbH, Eschborn, Germany

[Gup 2012/3]   Gupta, S.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–HR-35-QLR (November 2012), Becker Technologies GmbH, Eschborn, Germany

[Gup 2013]   Gupta, S.; Balewski B.; Kühnel A.; Funke F.; Langrock G.: Deposition of molecular iodine on aerosol particles - Iod-26 Test using reactive aerosol Ag. OECD-NEA THAI-2 Project, Report No. 1501420–Iod-26-QLR (February 2013), Becker Technologies GmbH, Eschborn, Germany, AREVA GmbH, Erlangen, Germany

[Gup 2013]   Gupta, S.; Langer, G.: Hydrogen combustion during spray operation - Tests HD-30, HD-31 and HD-32. OECD-NEA THAI-2 Project, Report No. 1501420-QLR-HD30-32 (November 2013), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014/1]   Gupta, S.; Schmidt, E.: Effect of reduced O_2 content in air on PAR H_2-ignition limit - HR-36 and HR-37, Tests with AREVA PAR. OECD-NEA THAI-2 Project, Report No. 1501420–HR36/37-QLR (January 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014/2]   Gupta, S.; von Laufenberg B.; Schmidt E.; Freitag F.; Funke F.; Langrock G.: Iodine release from a flashing jet (Test ID: Iod-29). OECD-NEA THAI-2 Project, Report No.1501420–Iod-29-QLR (April 2014), Becker Technologies GmbH, Eschborn, Germany, AREVA GmbH, Erlangen, Germany

Technical Reports (TR)

[Fre 2013]   Freitag, M.; Sonnenkalb, M.: Comparison report for blind and open simulations of HR-35. OECD-NEA THAI-2 Project, Report No. 1501420 – HR35 – AWG (VB) (December 2013), Becker Technologies GmbH, Eschborn, Germany, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Köln, Germany

[Fun 2013]   Funke F.; Langrock G.; Dorsch T.; Gupta S.; von Laufenberg B.; Kühnel A.; Weber G.: Deposition of molecular iodine on aerosol particles - Iod-26 (Test using reactive aerosol Ag). OECD-NEA THAI-2 Project, Report No. 1501420–Iod-26-TR (November 2013), AREVA GmbH, Erlangen, Germany, Becker Technologies GmbH, Eschborn, Germany, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Garching, Germany

[Fun 2014a]   Funke F.; Langrock G.; Gupta S.; von Laufenberg B.; Schmidt E.; Freitag F.; Kühnel A.: Iodine release from a flashing jet – Test Iod-29. OECD-NEA THAI-2 Project, Report No. 1501420–Iod-29-TR (June 2014), Becker Technologies GmbH, Eschborn, Germany, AREVA GmbH, Erlangen, Germany

[Fun 2014b]   Funke, F.; Gupta S.; Balewski, B.; Weber G.: Deposition of molecular iodine on aerosol particles - Iod-25 (Test using non-reactive aerosol SnO2). OECD-NEA THAI-2 Project, Report No. 1501420–Iod-25-TR (July 2012), AREVA GmbH, Erlangen, Germany, Becker Technologies GmbH, Eschborn, Germany, Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) gGmbH, Garching, Germany

[Gup 2014]   Gupta, S.; Colombet, M.; Langer, G.: Hydrogen combustion during spray operation - Tests HD-30, HD-31 and HD-32. OECD-NEA THAI-2 Project, Report No. 1501420-TR-HD-30-32 (April 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014a]   Gupta, S.; Langer, G.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-33-35 (June 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014b]   Gupta, S. , Schmidt, E. ; Langer, G.: Effect of reduced oxygen content in air on ignition potential of Passive Autocatalytic Recombiners (PAR) - Tests HR-36 and HR-37. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-36-37 (June 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014c]   Gupta, S. et al.: Hydrogen combustion during spray operation; Tests HD-30 to HD-35. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HD-30-35 (September 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014d]   Gupta, S.; Schmidt, E.: Onset of PAR operation in case of low oxygen concentration. OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-38-39 (December 2014), Becker Technologies GmbH, Eschborn, Germany

[Gup 2014e]   Gupta, S.; Schmidt, E.; Langer, G.: Hydrogen recombiner tests with NIS-PAR (Tests HR-40, HR-41, HR-42). OECD-NEA THAI-2 Project, Report No. 1501420–TR-HR-40-42 (July 2014), Becker Technologies GmbH, Eschborn, Germany

[Lan 2012]   Langrock, G.: Laboratory-scale tests on the interaction of gaseous I_2 with aerosol particles. OECD-NEA THAI-2 Project, Repot No. PTCC-G/2012/en/0315 (December 2012), AREVA GmbH, Erlangen, Germany