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Thermal-hydraulic–iodine chemistry coupling: Insights gained from the SARNET benchmark on the THAI experiments Iod-11 and Iod-12



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HIGHLIGHTS

- The I₂ transport in two multi-compartment THAI tests was analyzed.
- In a benchmark 4 different codes were applied by 7 organizations.
- The I₂ concentrations were mostly overestimated, up to a factor 100.
- Inadequate iodine models and inaccurate thermal-hydraulic parameters were detected.
- The user effect on the quality of the iodine results was large.

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ABSTRACT

In the SARNET2 WP8.3 THAI Benchmark the capability of current accident codes to simulate the iodine transport and behavior in sub-divided containments has been assessed. In THAI test Iod-11 and Iod-12, made available for the benchmark, the distribution of molecular iodine (I₂) in the five compartments of the 60 m³ vessel under stratified and well mixed conditions was measured. The main processes addressed are the I₂ transport with the atmospheric flows and the interaction of I₂ with the steel surface. During test Iod-11 the surfaces in contact with the containment atmosphere were dry. In Iod-12, steam was released, which condensed on the walls.

Nine post-test calculations were conducted for Iod-11 and eight for Iod-12 by seven organizations using four different codes: ASTEC-IODE (CIEMAT, GRS and TUS), COCOSYS-AIM (AREVA, FZ-jülich and GRS), ECART (Pisa University) and MELCOR (Pisa University and VTT). Different nodalizations of the THAI vessel with 20–65 zones were applied.

Generally, for both tests the analytical thermal-hydraulic results are in a fairly good agreement with the measurements. Only the calculated local relative humidity deviates significantly from the measured values in all calculations. The results in Iod-11 for the local I₂ concentration in the gaseous phase are quite diverse. Three calculations show only minor deviations from the measurement, whereas the others are substantially different from the measured I₂ concentrations. For Iod-12, no calculation delivers a satisfactory evolution of the I₂ concentration in all five compartments of the vessel. There are three mediocre results standing out in the Iod-11 exercise which are from the same user–code combinations. The discrepancies derive from various reasons which are discussed in the paper.

In the benchmark a significant user effect was detected, i.e. results achieved with the same code differed considerably.

This work highlights the need of a detailed iodine adsorption/desorption model and precise thermal-hydraulic modeling for an accurate simulation of I₂ transport in a sub-divided containment, as well as experienced users or straight forward user guidelines.

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1. Introduction

In case of an accident in a nuclear power plant iodine is a major contributor to the radioactive source term because of its volatility and the high radiological consequences. Therefore, large efforts have been made to describe the iodine behavior within the containment and the source term to the environment. The model development has been mostly focused on the chemical reactions that iodine species can undergo (Girault et al., 2012; Dickinson et al., 2010).

In the field of iodine chemistry during severe accidents, the containment atmosphere has been mostly assumed to be well mixed and consequently the iodine species were treated as be homogeneously distributed (Clément et al., 2007). From containment thermal-hydraulic analysis and especially the simulation of H₂ and nuclear aerosol distributions, as well as depending on the containment design, it is concluded that atmospheric mixing may be slow or incomplete, resulting in local concentration differences of gases and aerosols. As most of the iodine reaction rates are dependent on the species concentrations, these are key variables whose estimate should be as accurate as possible, particularly the ones corresponding to volatile species such as molecular iodine (I₂). Many iodine reaction velocities depend on local temperature, humidity, wall condensation rate, etc. In other words, iodine behavior within the containment and hence, the iodine source term, are not only dependent on the chemical reactivity, but also on the accuracy of the simulation of the transport and the thermal-hydraulic conditions in the compartments of the containment (Clément et al., 2007).

Several accident codes available have an integrated containment iodine model, like ASTEC-IODE, COCOSYS-AIM, ECART, MAAP-IMPACT, MELCOR-INSPECT, etc. They allow for calculating iodine transport in multi-compartment geometries and the interrelation of iodine chemistry with thermal-hydraulics and aerosol physics. Validation of such coupling effects of iodine chemistry was, however, previously rather limited due to the scarce experimental data available.

Since 1998 the technical-scale THAI test facility (THAI = Thermal-hydraulics, Hydrogen, Aerosols, Iodine) has been operated by Becker Technologies at Eschborn, Germany, in close co-operation with AREVA NP and GRS in order to provide an experimental database for the development and validation of lumped-parameter and CFD containment codes. Up to now, 26 iodine tests have been performed evaluating several phenomena such as iodine/steel and iodine/paint interactions, mass transfer between sump and gaseous phase, and IOx aerosol formation. (Weber et al., 2010). Between 2004 and 2007 the I₂ transport in a 5-compartment geometry was measured in four tests (Funke et al., 2004).

The present paper summarizes the main results obtained in the framework of an open benchmark (i.e. data were known by participants) based on the Iod-11 and Iod-12 tests of the THAI program. The data were released to SARNET by the German Ministry of Economics and Technology (BMWi) (Kanzleiter et al., 2005) and the work was carried out within the Source Term work package of the SARNET network. After introducing both tests, key aspects of the modeling are described to better understand the comparisons set to data. From them, conclusions regarding the current code's capability for simulating the coupling of thermal-hydraulics and iodine chemistry have been drawn and are presented.

2. THAI tests Iod-11 and Iod-12

2.1. THAI vessel and instrumentation

The main component of the THAI facility is the cylindrical stainless steel vessel of 9.2 m height and 3.2 m diameter, with a total

volume of 60 m³ (Fig. 1). To achieve the 5-compartment geometry two intermediate decks made of chromium–nickel–steel-sheets were installed. Five inter-compartmental flow openings in these decks allowed gas exchanges. Two slit shaped openings were at the upper deck, two also slit shaped ones were on the lower deck and one square opening was at the bottom of the inner cylinder. The five compartments were: dome, upper and lower annulus, central compartment (inner cylinder) and bottom compartment including the sump.

The gases were injected at different locations. Helium (simulating hydrogen) was released in Iod-11 into the bottom compartment at 1.6 m against a baffle plate to ensure a fast mixing with the ambient air. In Iod-12 He was released into the dome at 7.75 m and later on into the bottom compartment at 1.8 m. In Iod-11 little steam was injected into the dome at 7.7 m during the setting of test conditions and during the transient phase into the bottom compartment at 1.6 m. Much steam was released again through the bottom compartment pipe at 1.6 m at the end of the test for vessel washing. In Iod-12 the steam was injected into the upper part of the inner cylinder. From there it escaped to the outer compartments and condensed on the cold walls achieving mixing of the vessel atmosphere. Gaseous I₂ was released in both tests into the dome at 8.4 m also against a baffle plate.

Conventional instrumentation was installed to measure the pressure, fluid and structures' temperatures, injection mass flows and the water level in the main sump. The helium concentration was measured at nine locations with an accuracy of ±0.2 vol.%. The dew point temperature (±0.5 °C) was recorded at four levels: bottom compartment (2.1 m), upper annulus (4.9 m) and in the dome (7.7 m and 8.4 m). The relative humidity was calculated from the dew point temperature.

The inactive I₂ injected was traced with some radioactive iodine I-123 and gas and liquid samples were taken at numerous locations. Iodine concentration in the gaseous phase was determined by gamma-ray evaluation of samples from in situ gas scrubbers. The errors of the iodine concentration measurement typically varied between ±15 and ±30%. More details on the iodine instrumentation are given in Funke et al. (2006) and Weber (2012).

A steel coupon with 50 cm diameter was mounted in the dome compartment at 7.7 m. The iodine deposited on the coupon was measured through a glass window by a scintillation detector placed outside the vessel.

2.2. Test Iod-11

The three main phases of test Iod-11 are distinguished by different flow conditions in the vessel atmosphere, which were stratified, transient and mixed (Table 1) (Kanzleiter et al., 2005). At the beginning of the test the vessel was prepared and the initial test conditions, a stratified vessel atmosphere with about 95 °C in the dome and near-ambient temperature in the bottom compartment, were adjusted. The sump was filled with 1.25 m³ of water and a pH ≈ 2 was adjusted in order to avoid I₂ hydrolysis. At the beginning of the *stratified phase*, 8.4E–4 kg of gaseous I₂ was injected into the dome within several minutes. The beginning of injection was the start of the test time ($t = 0$). The iodine spread slowly in the dome atmosphere, which was also slightly stratified, and reached the gas scrubber near the bottom of the dome after about 1½ h. One part of the I₂ was adsorbed on the steel surface. During the stratified phase almost no I₂ reached the lower compartments.

In the *transient phase* the mixing of the vessel atmosphere was stimulated by a controlled injection of heat, helium and steam. The vessel atmosphere was heated by the middle and lower vessel jackets and the sump water was heated electrically without reaching the boiling point. 8.8 standard-m³ of helium were injected into the bottom compartment to support the atmospheric mixing and to

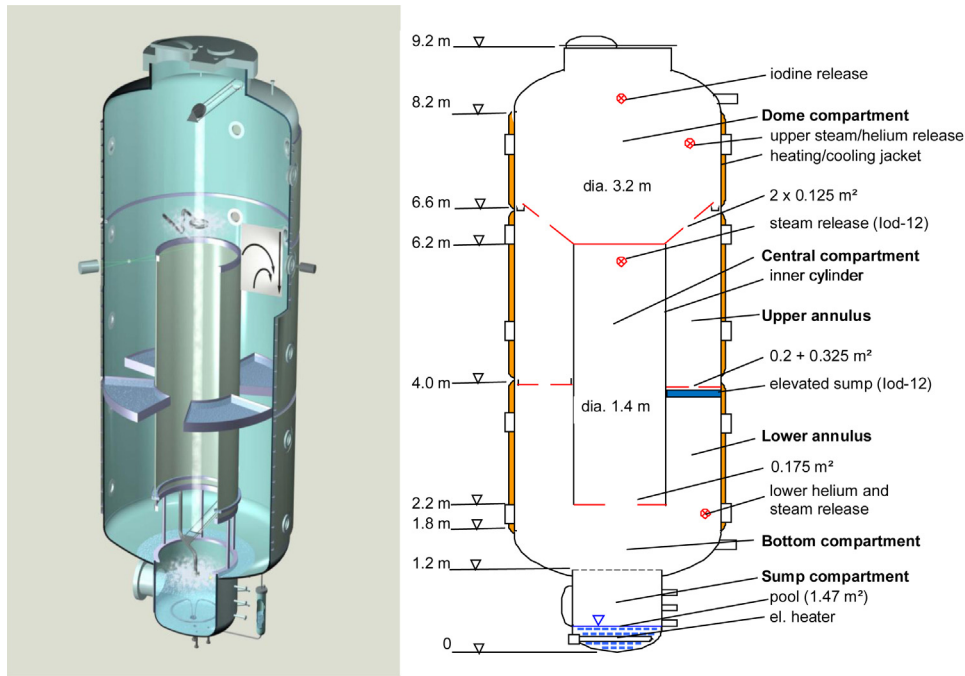


Fig. 1. THAI vessel with standard configuration and with 5-compartment geometry for tests lod-11 and lod-12.

serve as a tracer. Later some steam was injected through the same pipe.

The released helium was rapidly distributed by the convective flows and after three hours it was completely mixed (Fig. 2). Iodine behaved differently. It was transported from the dome into the lower compartments but its concentration was not homogenized in the vessel as the iodine gas scrubber measurements show (also Fig. 2). This incomplete mixing is due to I_2 adsorption and desorption processes onto/from the steel surfaces. This I_2 mass transfer between atmosphere and steel surfaces lasted until the end of the test.

In the *mixed phase* the well mixed conditions were maintained by keeping atmospheric and sump temperatures nearly stationary. The I_2 distribution in the vessel changed only gradually, but the concentration decreased in all compartments due to chemisorption. At the end of the mixed phase the I_2 concentration in the dome was still one order of magnitude higher than in the lower compartments. In the lower compartments the I_2 concentration differed up to a factor 2, which is clearly above the measurement error of $\pm 30\%$.

The aqueous I_2 concentration in the sump remained below the detection limit at all times except in the washing phase.

Table 1
lod-11 and lod-12 test procedures.

Test phase	Duration		Main actions	
	lod-11	lod-12	lod-11	lod-12
1. Vessel preparation	–44.28 h till –5.0 h	–48.0 h till 5.0 h	Dry heating of the closed test vessel by upper vessel jacket and cooling by middle vessel jacket	
2. Setting of test conditions	–5.0 h till 0 h	–5.0 h till 0 h	Setting of test conditions by injection of air, little steam and heating the upper vessel jacket temporarily	Additional injection of little He into the dome
3. Stratified atmosphere	0 h till 4.35 h	0 h till 2.6 h	Iodine injection into the dome; measurement of local iodine concentration in stratified vessel atmosphere	
4. Transient conditions	4.35 h till 8.45 h	2.6 h till 7.0 h	Atmosphere mixing started by heating the middle and lower jackets (strongly reduced at $t = 7.25$ h); injection of He into the bottom compartment	Atmosphere mixing started by steam injection into the inner cylinder; injection of He into the bottom compartment
5. Mixed conditions	8.45 h till 24.67 h	7.0 h till 9.7 h	Vessel atmosphere mixed by continuous jacket and sump heating	
6a. Rest phase	–	9.7 till 24.4 h	–	Without any injections; measurements interrupted
6b. Desorption phase	–	24.4 till 29.9	–	Iodine desorption in the dome by heating the upper jacket
7. Washings	24.67 h till 30.57 h; two further washings	29.9 till 33.67 h; two further washings	Washing of iodine deposits from vessel walls by means of condensing steam	

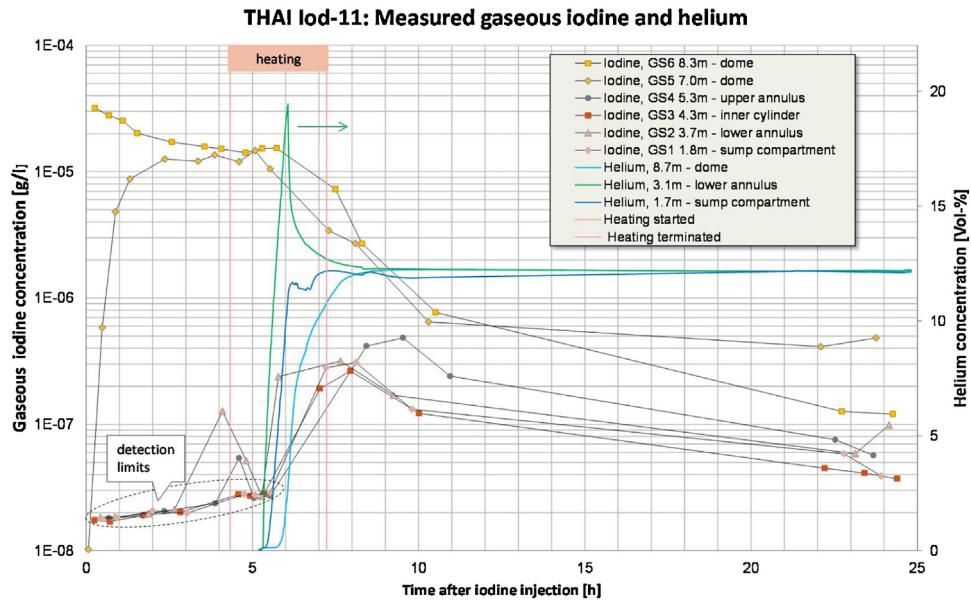


Fig. 2. Gaseous iodine and helium concentrations measured in lod-11.

At the end of the test the deposited iodine was washed down by mean of injected steam, which condensed on the cold walls and structures.

2.3. Test lod-12

The general procedure of the wet test lod-12 with seven phases was similar to that of the dry test lod-11 (Table 1). At the end of the two preparation phases the vessel atmosphere had a temperature stratification of about 100 °C in the dome (rh ≈ 30%) and 25 °C in the sump compartment (rh ≈ 80%). The total pressure was about 1.4 bar. The main sump was initially filled with approximately 0.42 m³ of water and buffered to pH=1.0. At a level of 4.0 m one lateral tray was filled with 0.05 m³ forming the so-called elevated sump, which was also buffered to pH=1.0. A small amount of He was injected into the upper part of the dome in order to measure the

atmospheric mixing there and later on the atmospheric transport into the lower compartments.

At the beginning of the stratified phase 1.18 ± 0.12 g of gaseous I₂ was injected into the dome within 10 min. Continuous iodine concentration measurements by gas scrubbers as well as liquid samplings were made (Fig. 3). The typical gas volume sampled by each of the 6 scrubbers ranged between 1.5 and 15 l. The total atmosphere volume withdrawn was 0.390 m³ at normal conditions.

In the transient phase steam was injected into the upper part of the inner cylinder with a flow rate of about 30 g/s, which was reduced later on. The steam filled up the volume from the top to the bottom. At the lower edge the steam escaped into the lower annulus inducing atmospheric mixing in the whole vessel. The mixing was supported by a helium injection with a total mass of 0.56 kg into the bottom compartment (H=1.6 m). The wall condensate with the iodine washed out was collected in three gutters and conducted

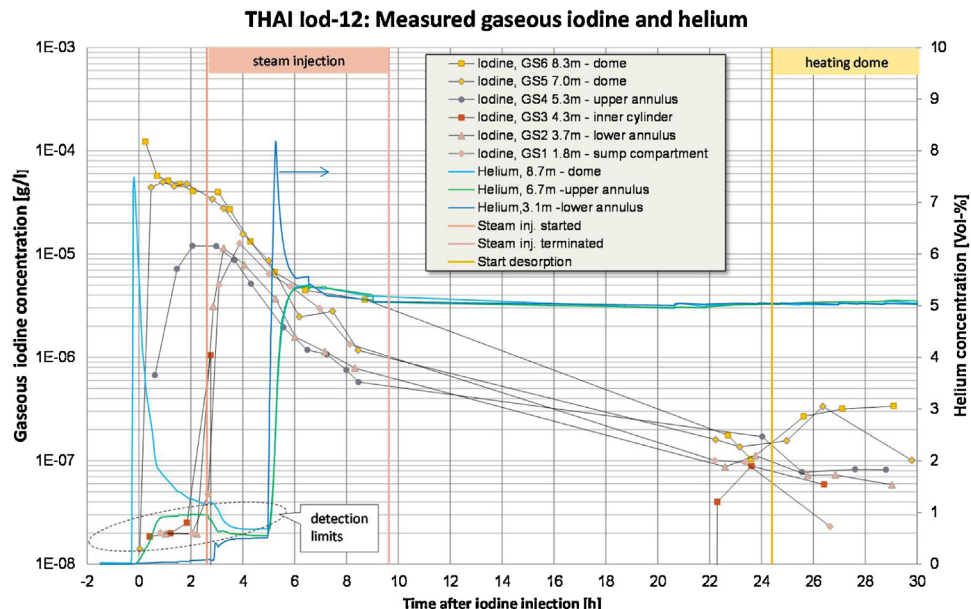


Fig. 3. Gaseous iodine and helium concentrations measured in lod-12.

Table 2
Participants, codes and calculations.

Organization	Code	Nodalization (zones/levels)	Calculations		Remarks
			Iod-11	Iod-12	
AREVA, Germany	COCOSYS V2.4beta – AIM-3	39/11	Yes	Yes	
CIEMAT, Spain	ASTEC V2.0 – CPA/IODE	46/12	Yes	Yes	
GRS, Germany	COCOSYS V2.4 – AIM-3	51/14	Yes	Yes	
	ASTEC V2.0R2 – IODE	51/14	Yes	Yes	With support from IRSN
FZ-JÜLICH, Germany	COCOSYS V2.4 – AIM-3	44/11	Yes	Yes	
TUS, Bulgaria	ASTEC V2.0 – IODE	7/4 (Iod-11) 25/11 (Iod-12)	Yes, only th-hy	Yes	Iod-11: only dome modeled
UNIPI, Italy	ECART FIRE v. 4W0U	65/13	Yes	Yes	
	MELCOR 1.8.6 YV	52/13	Yes	No	
VTT, Finland	MELCOR 1.8.6 YV	20 resp. 21/8	Yes	Yes, only th-hy	

into tanks outside the vessel. The upper and lower annulus and the sump compartment were drained via gutter WB1. The dome compartment was drained via gutter WB2 and the inner cylinder via gutter WZi.

The mixed conditions were maintained in test phase 5 (*mixed phase*) by a continuous steam injection of about 6.0E–3 kg/s. Iodine measurements in the gaseous phase, in the two sumps and from four condensate gutters continued.

During the *rest phase* overnight no injections or measurements were carried out. In the *desorption phase*, I₂ in the dome was desorbed by heating the dome walls by the upper vessel jacket. Finally the iodine deposits from vessel walls were washed down by condensing steam into the sump.

3. Modeling

For the benchmark calculations the participants were provided with the following information: (a) test report and experimental data (Kanzleiter et al., 2005), (b) specification of the benchmark calculations (Weber, 2012) and (c) example input decks for ASTEC/CPA and COCOSYS V2.4/AIM-3 for a simple 6-zone nodalization of the THAI vessel. These input decks were not suitable for analyzing the tests because of too few zones, but they could serve as a basis for more detailed nodalizations, even for other codes.

3.1. Participants and codes

Test Iod-11 was calculated by seven organizations using four different codes. Since UNIPI and GRS made calculations with two different codes each, a total of nine calculations were evaluated (Table 2).

For test Iod-12, eight calculations on the thermal-hydraulic problem and seven on the iodine problem were performed. Because of lack of manpower UNIPI made no analysis with MELCOR. VTT did not calculate the iodine part, because of unsatisfactory results with Iod-11. Therefore, no iodine results with MELCOR are available for Iod-12.

Only lumped parameter codes were used in the benchmark. The integral code ASTEC (Accident Source Term Evaluation Code) (Van Dorselaere et al., 2009) is being developed by IRSN in close cooperation with GRS. ASTEC simulates the complete scenario of a severe accident in a light water reactor and estimates the possible fission product release from the reactor containment. The containment part of ASTEC (CPA) is similar to COCOSYS, except for the iodine model IODE in ASTEC. The IODE module models both gaseous and liquid iodine chemistry with a set of about 30 chemical reactions and some additional equations for the mass transfer processes (Bosland et al., 2010).

The Containment Code System (COCOSYS) is being developed by GRS (Allelein et al., 2008). The main module AFP (aerosols and fission product) contains the Advanced Iodine Model AIM-3. AIM calculates a total of 70 chemical reactions and physical processes for 26 iodine species and 8 non-iodine species in each compartment as well as the iodine transport between compartments by gas and water flows.

The code ECART has been developed at ENEL Milano with the collaboration of Italian Universities (Fontana, 2010). It simulates the transport of nuclear materials within a nuclear power plant and their release to the environment. A central thermal-hydraulic module provides the boundary conditions for the other modules. An iodine model is not available, but simple reactions can be treated.

MELCOR is developed at SANDIA NL for the USNRC (Gauntt et al., 2005). It is a fully integrated computer code whose primary purpose is to model the progression of severe accidents in light water reactor nuclear power plants. An iodine pool model is implemented to predict iodine in the containment. Typically, for plant applications, only CsI aerosol is treated. The capability to model iodine gas chemical behavior is rather limited.

3.2. Iodine/steel model

In Iod-11 and Iod-12 the iodine/steel interaction is an important phenomenon. For the calculations the use of the iodine/steel model in AIM-3 (Weber and Funke, 2009) was recommended. The model was developed on the basis of experimental results from several tests with THAI steel (AISI 316 Ti) and from the OECD BIP project (OECD, 2011). It also underwent substantial validation on other tests in stainless steel vessels, like PHEBUS, RTF, etc. (Bosland et al., 2012; Ball and Marchand, 2004).

The model consists of a physi-sorption step followed by a chemisorption one (Fig. 4). Physi-sorption is a fast and reversible I₂ deposition process onto the steel surface. Physi-sorbed I₂ is loosely bound to the surface striving for a concentration-equilibrium with the gaseous iodine. Chemisorption is the slow chemical reaction of physi-sorbed I₂ with the steel forming non-volatile metal iodides (FeI₂). The model considers also the re-volatilization reaction of FeI₂ with air oxygen to volatile I₂, as originally described in Wren and Glowa (2001).



In Eq. (1) k_{ads} and k_{des} are the adsorption and desorption rate constants. k_{des} is temperature dependent. The reaction rate for chemisorption (k_{chs}) increases with increasing relative humidity (Fig. 4). The re-volatilization reaction of FeI₂ with oxygen (k_{rev}) is

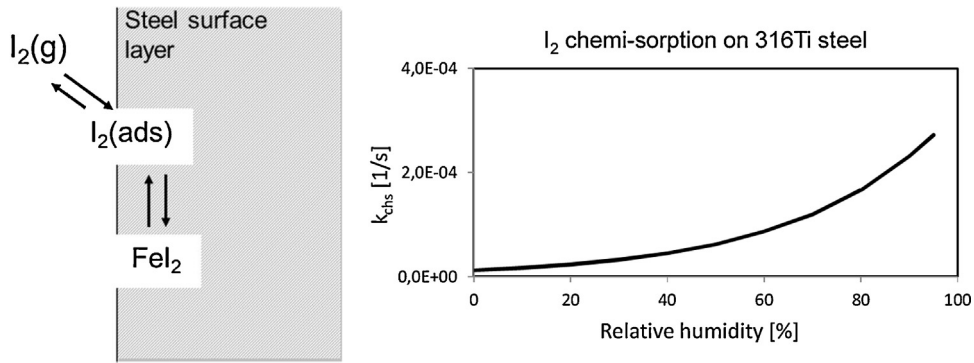


Fig. 4. Iodine/steel reactions (left) and chemi-sorption rate constant.

relatively slow and therefore it is of minor importance in the THAI tests. All the kinetic constants are determined and internally stored in the AIM model, although they might be easily overwritten if needed. The set of constants for THAI steel (316Ti) is also given in the benchmark specification (Weber, 2012).

Because of difficulties in implementing the AIM iodine/steel model in other codes the recommended model was only used with the COCOSYS calculations (Table 3). In IODE, a reversible single step reaction rate is included to describe the iodine interaction with SS surfaces; this approach requires specifying one adsorption and one desorption rate in the input deck. Chemi-sorption is not modeled explicitly. Because of the large area of painted surfaces in most of the PWR containments on which iodine adsorbs much faster than on steel, the iodine–steel model in ASTEC is sufficient. In ECART only chemi-sorption is available in a simplified way, i.e. adsorbed iodine cannot be desorbed again. Two chemi-sorption rates, one for non-oxidized and the other for pre-oxidized steel, were tested. Chemi-sorption was much faster for non-oxidized steel. The MELCOR model has the same restriction. Only chemi-sorption can be treated, desorption processes are not possible.

3.3. Nodalization

The choice of a suitable nodalization is crucial to each lumped parameter analysis. Enough vertical zone levels are needed to simulate atmospheric stratifications and convective flows. High and narrow compartments like the lower and upper annular rooms should be divided radially to allow counter current flows. In the five flow openings counter current flows should also be possible.

The nodalizations used by the participants to model the THAI facility differ from each other. The total number of zones ranges from 20 to 65 (Table 2). All nodalizations enable counter current flows in the upper and lower annulus as well as through the slit shaped openings in the upper and lower deck and in the square

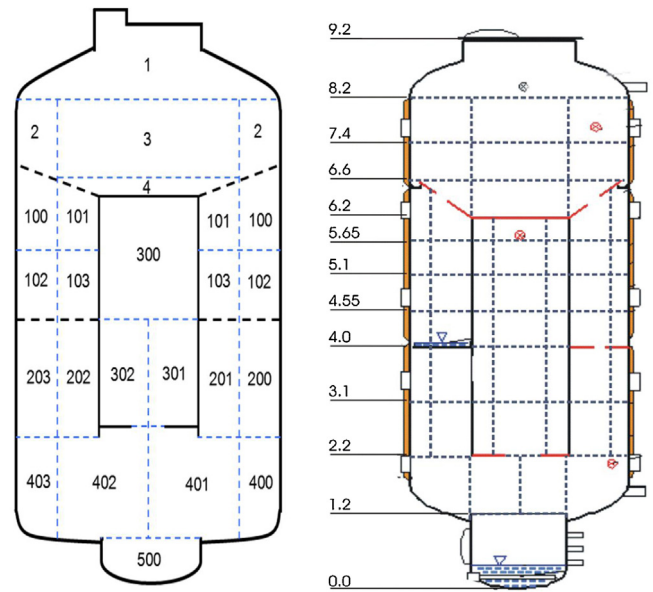


Fig. 5. MELCOR-VTT (left) and ASTEC-CIEMAT nodalizations of the THAI vessel.

opening at the bottom of the inner cylinder. In the ASTEC Iod-11 calculation by TUS only the dome area is simulated with 7 zones arranged in 4 levels. The nodalizations for MELCOR-VTT and ASTEC-CIEMAT are given as examples in Fig. 5; the others are in the THAI Benchmark report (Wren and Glowa, 2001).

4. Data – codes comparison

The comparison of the analytical results with the measured data is made for essential iodine variables and those thermal-hydraulic

Table 3
Iodine–steel model used by the participants.

Code, iodine model	I ₂ /steel model		User	Description and application
	Adsorption/desorption	Chemi-sorption		
ASTEC-IODE	Yes	No	CIEMAT GRS TUS	Adsorption/desorption rates adjusted to the measured concentrations
COCOSYS-AIM-3	Yes	Yes	AREVA GRS FZ-JÜLICH	AIM-3 default AIM-3 default plus extension (reverse reaction to chemi-sorption) AIM-3 default
ECART	No	Yes	UNIPI	Only chemi-sorption modeled; different reaction rates for non-oxidized and pre-oxidized steel
MELCOR	No	Yes	UNIPI VTT	Chemi-sorption without desorption; rate adjusted to measured concentrations So-called iodine pool model not usable; chemi-sorption without desorption applied, rate estimated

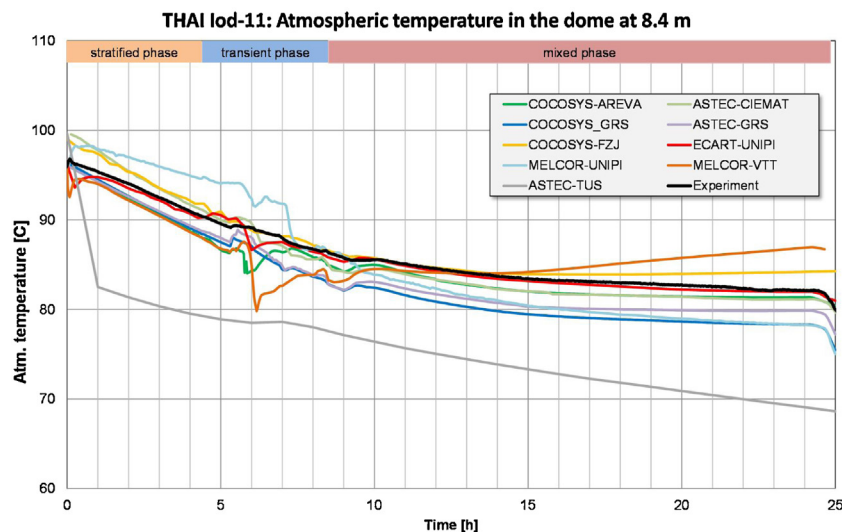


Fig. 6. Atmospheric temperature in the dome at 8.4 m in lod-11.

variables, which have an impact on the iodine result. A representative selection of the comparison plots for both tests will be discussed. Further plots can be found in Weber et al. (2012).

4.1. lod-11

The majority of the participants performed two post-test calculations on test lod-11, the first one on their own and the second (final) one after a discussion of all participants' results. Here only final results are presented.

4.1.1. Total pressure

The pressure has no direct influence on the iodine adsorption and desorption processes, but it can influence the atmospheric transport and hence the local iodine concentration. The calculated pressure deviates from the measurement in all calculations at a maximum of $(-0.1/+0.07)$ bar, which is quite acceptable. In the mixed phase, the pressure increase in all calculations is somewhat faster than in the experiment. Possible reasons are an overestimation of the atmospheric temperature in the annular rooms of approximately 5°C in several calculations and the air sampling for the iodine measurement of about 400 standard-I, which were not considered in most calculations.

4.1.2. Atmospheric temperature

The agreement of the calculated temperatures in the dome with the measurement ($\pm 5^{\circ}\text{C}$) is good for all calculations except ASTEC-TUS (Fig. 6). In this calculation only the dome but not the lower compartments are modeled, which is the reason for the significant underestimation of the temperature. The sudden temperature decrease at about $t = 6$ h in the MELCOR-VTT and to a smaller extent in three other calculations, is due to a temporary increase of the convection flows, just after the helium injection. This atmospheric instability was not measured. At the same time the calculated iodine concentrations in the dome decline (Fig. 9).

In the other compartments, the discrepancy of the calculated temperature is larger than for the dome. For example in the sump compartment the atmospheric temperature is mostly underestimated, maximally by -18°C .

4.1.3. Relative humidity

The spread of the calculated relative humidity (rh) is the largest of all thermal-hydraulic parameters. The deviation from

measurement increases progressively in the test. In the sump compartment it is moderate during the stratified phase ($0/+10$ rh%), becomes noticeably larger in the transient phase ($-40/+10$ rh%) and remains large in the mixed phase ($-30/+20$ rh%) (Fig. 7).

Inaccurately calculated atmospheric temperatures are the main reason for the large rh discrepancy in all compartments except the dome. The temperature in the lower compartments was overestimated in many calculations. For example, in the upper annulus the measured temperature is 84°C and the measured rh is about 50% at $t = 13.9$ h. The temperature in this compartment was overestimated at the same time in all calculations between 5.5 and 9.9°C . This deviation resulted in an underestimation of the relative humidity by -9 to -15% assuming that the steam concentration was calculated correctly.

In the dome compartment the situation is different. In most calculations the temperature is correctly predicted ($\Delta T = -3.7$ to 4.9°C) but the relative humidity is unexpectedly low ($\Delta \text{rh} = -7.5$ to -27%). The most likely explanation is an underestimation of the steam concentration there due to inaccurately simulated atmospheric transport and/or wall condensation processes.

4.1.4. Helium distribution

In test lod-11, the injected helium was homogeneously distributed throughout the whole vessel within 3 h after injection. This mixing period is correctly calculated by COCOSYS-AREVA, ASTEC-CIEMAT, ECART-UNIPI and MELCOR-VTT. In the other calculations the mixing period is two to three times too long. The reason is obviously a too weak atmospheric flow through the two openings in the upper deck. In all calculations the mass flow rates through the lower deck openings are higher than through the upper deck openings, which is plausible during dispersion of the atmospheric stratification.

The helium concentration in the inner cylinder is somewhat overestimated (max. 2 vol.%) in all calculations (Fig. 8). In COCOSYS-AREVA the He concentration is overestimated by 5 vol.% in the injection period due to an overestimated atmospheric mixing. In general the results are good and demonstrate that the counter current flows in and out of the dead end inner cylinder are correctly simulated in all calculations.

4.1.5. Gaseous iodine

The iodine transport within the vessel can be traced back from the evolution of the gaseous iodine concentration in different

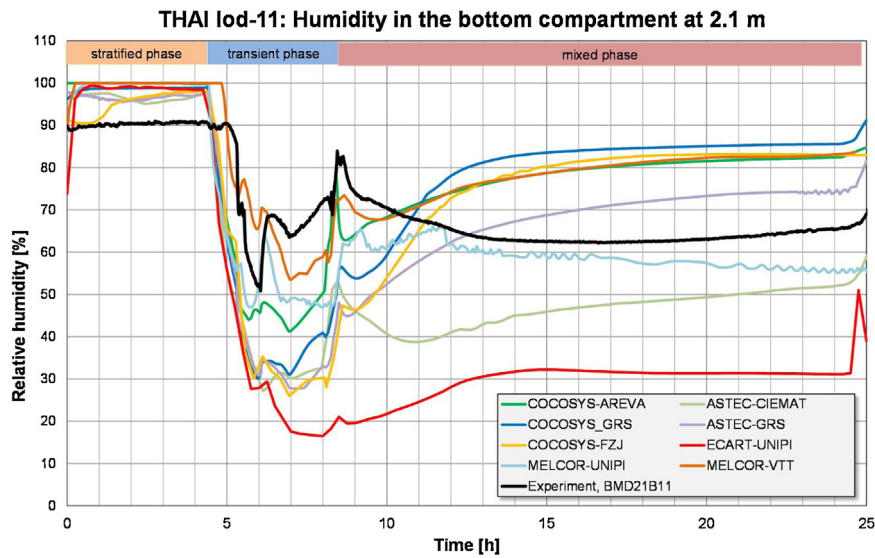


Fig. 7. Humidity in the bottom compartment at 2.1 m in lod-11.

compartments. The discussion below extends to all THAI compartments, although Fig. 9 illustrates a specific location in the dome region; the other plots may be found elsewhere (Weber et al., 2012).

During the *stratified phase* ($t=0-4.35$ h) in the test hardly any I_2 is transported from the dome into the lower compartments. This is almost correctly obtained by three calculations (ASTEC-CIEMAT, COCOSYS-GRS and COCOSYS-FZJ) except in the upper annulus. In all other calculations the concentration in the upper annulus and in the compartments below is overestimated, at least by one order of magnitude. The reason for this artificial I_2 transfer is weaknesses of the nodalizations.

During the *transient phase* ($t=4.35-8.45$ h) the thermal stratification is dispersed and a moderate natural convection loop starts transporting I_2 from the dome into the lower compartments. The atmosphere exchange is slow and overlaid by adsorption/desorption processes on the stainless steel surfaces. At the end of this phase, the measured I_2 concentration in the dome is still a factor of 7 higher than in the lower compartments. The evolution of the I_2 concentrations in different regions of the vessel

is well simulated (factors 0.2/5.0) in three calculations (ASTEC-CIEMAT, COCOSYS-GRS and ASTEC-GRS). In all other calculations, the I_2 mixing in the vessel is too fast, although the helium mixing is too slow. At the end of the transient phase I_2 is calculated to be homogeneously distributed in the vessel. In these calculations (COCOSYS-AREVA, COCOSYS-FZJ, UNIPI-ECART, UNIPI-MELCOR) I_2 adsorption and/or desorption are not modeled correctly. The I_2 deposition is underestimated.

In the *mixed phase* ($t=8.45-24.67$ h), the I_2 distribution within the vessel is governed by moderate convective flows exchanging the atmosphere between the five compartments. These flows are not strong enough to equalize the I_2 concentration in the rooms. At the end of the test ($t=24$ h) the concentration difference between the dome and the bottom compartment is still about one order of magnitude. The main reason is an underestimation of the iodine deposition onto steel caused by inadequate iodine/steel models. In the MELCOR-VTT calculation gaseous I_2 -concentration is strongly underestimated in all phases because I_2 desorption is not considered.

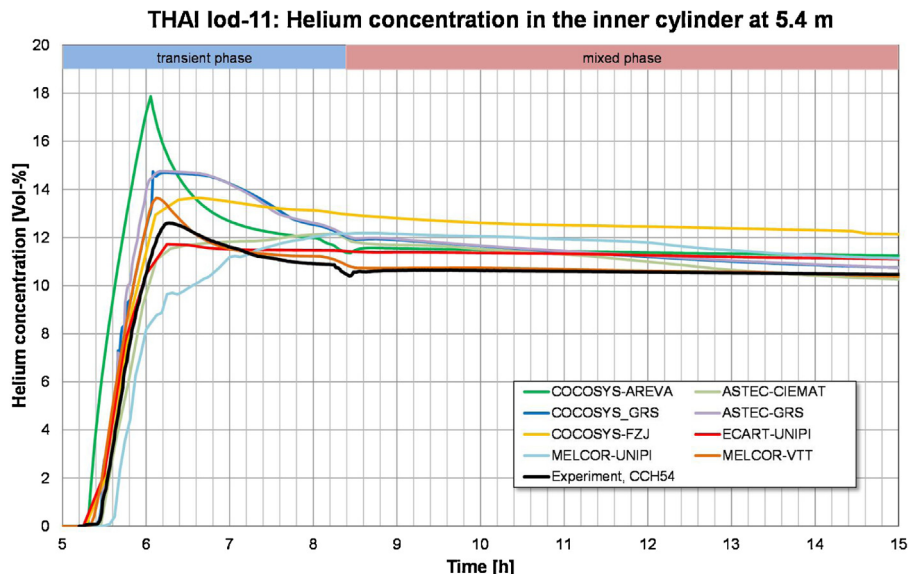


Fig. 8. Helium concentration in the inner cylinder at 5.4 m in lod-11.

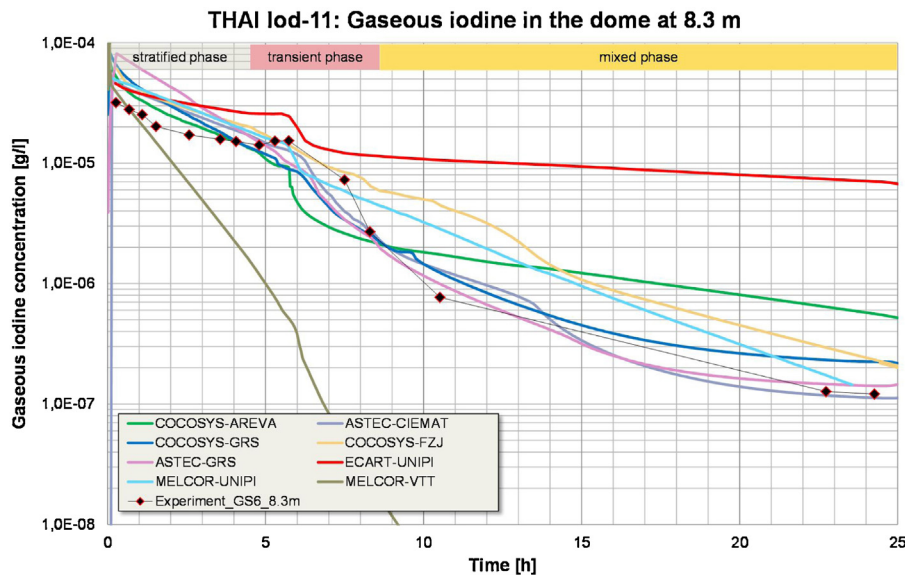


Fig. 9. Gaseous iodine in the dome at 8.3 m in Iod-11.

Only in three calculations (ASTEC-CIEMAT, COCOSYS-GRS and ASTEC-GRS) are essential details, like the I_2 concentration difference and I_2 concentration decrease, reproduced well for the whole test. Three other calculations (COCOSYS-AREVA, COCOSYS-FZJ and ECART-UNIPI) meet the measurement only partially. In the MELCOR-VTT calculation the I_2 concentration is again extremely underestimated and the concentration differences between the dome and the lower compartments are too small. The main reasons for the discrepancies are unfavorable iodine/steel models.

Fig. 9 shows the comparison of all calculated I_2 concentrations in the *dome*. ASTEC-CIEMAT, COCOSYS-GRS and ASTEC-GRS agree well with the measured I_2 evolution (factors 0.3/2.0). In COCOSYS-AREVA, COCOSYS-FZJ and MELCOR-UNIPI the I_2 concentration is temporarily overestimated in the mixed phase. In ECART-UNIPI the measured I_2 concentration is strongly overestimated and in MELCOR-VTT strongly underestimated for reasons already explained.

The results for the I_2 concentration in the *bottom compartment* show a quantitatively similar spread. ASTEC-CIEMAT, COCOSYS-GRS and ASTEC-GRS again agree well with the measured I_2 evolution. For ASTEC-CIEMAT the I_2 concentration is temporarily up to 1 order of magnitude too high. Two peaks occur with the correction of the adsorption/desorption rates at the beginning and the end of the transient phase. However, COCOSYS-AREVA, COCOSYS-FZJ and MELCOR-UNIPI overestimated the measured I_2 concentration by 1–2 orders of magnitude, which is clearly more than for the dome.

A significant user effect was detected, i.e. considerably different results were achieved with the same code ($2 \times$ ASTEC, $2 \times$ MELCOR, $3 \times$ COCOSYS). The main reasons are different nodalizations, different details in the thermal-hydraulic modeling (e.g. heating of the jackets) and different attempts to overcome the deficits in iodine/steel models.

4.2. Iod-12

The differences in the Iod-12 iodine behavior with respect to Iod-11 concern the additional transport of I_2 with the condensing steam onto the vessel walls and the subsequent partial wash down of deposited iodine from the wet walls by the draining condensate.

4.2.1. Thermal-hydraulics

The calculated total pressure lies within an error band of ($-0.2/+0.3$ bar) along the measurement. It is larger than for Iod-11, but still acceptable. In nearly all calculations, the trend throughout the complete test is quite well reproduced, indicating that heat and mass transfer are simulated well. Only in the ASTEC-calculation carried out by TUS is the pressure significantly underestimated. The main reason for this discrepancy is an insufficient nodalization. The dome is correctly modeled in 7 zones but none of the lower compartments (upper and lower annulus, inner cylinder and bottom compartment) are sub-divided. This prevents a correct simulation of the circulating atmospheric flows.

The sump temperature is overestimated in all calculations, mostly by MELCOR-VTT and ASTEC-TUS by about 20°C . In spite of the high temperature, the sump is still far from boiling. The atmospheric temperature is well simulated in all calculations except ECART-UNIPI and ASTEC-TUS. The spread of the calculated temperature in the stratified, transient and mixed phases is ($-5/+7^\circ\text{C}$). In the desorption phase the deviation from the measured temperature is much higher ($-20/+15^\circ\text{C}$). Only ASTEC-CIEMAT, COCOSYS-GRS and ASTEC-GRS match the measured temperature quite well in the desorption phase. In the bottom compartment, the atmospheric temperature is generally overestimated up to 10°C . Only ECART-UNIPI and ASTEC-TUS show larger deviations (Fig. 10).

As in Iod-11 the relative humidity is the thermal-hydraulic parameter with the largest deviation from the measurement. In the upper annulus during the stratified phase the spread of the calculated rh is about ($-20/+7$ rh%). During the transient phases rh in the upper annulus is underestimated up to -50% (Fig. 11). In the mixed, rest and desorption phases the agreement is better than in the transient phase. The deviations in Iod-12 are in general smaller than in Iod-11. This may be connected to the steam injection pushing the relative humidity up to over 65%.

The first He injection (Fig. 12) was placed into the upper part of the dome at the beginning of the stratified phase. The relatively fast depletion of the He concentration there is mostly underestimated, i.e. the atmospheric stratification is more stable in the calculations than in the experiment. This gas transport from the dome into the upper annulus can be recognized also by the iodine measurement given in Fig. 13.

The analyzed distributions of the He from the second injection at $t=5$ h (Fig. 12) are, in general, in good agreement with

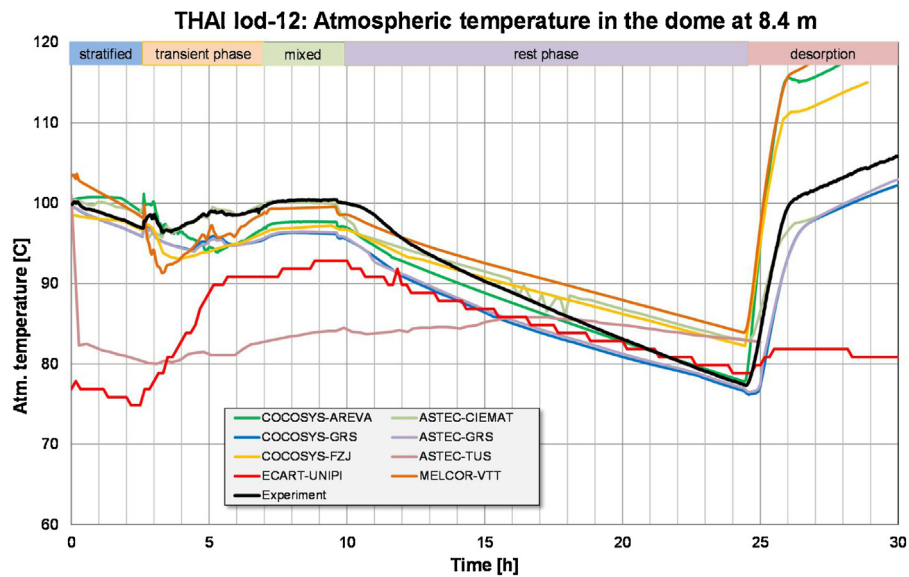


Fig. 10. Atmospheric temperature in the dome in lod-12.

the measurement ($-0.5/+0.5$ vol.%). The measured average concentration after mixing of about 3.5 vol.% is well matched in most calculations.

For the calculation of the iodine concentration in the draining condensate (see below) accurate condensate flow rates in the three gutters are needed. Only some calculated flow rates for the gutters WB1 and WZi are in an acceptable agreement with the measurement. The other flow rates are too low or zero. The reason for this discrepancy is mainly a weakness of the nodalizations applied. In some calculations condensate is not allowed to drain. In other calculations the condensate is conducted into the main sump and not via the gutters into the external tanks.

4.2.2. Iodine behavior

In the *stratified phase* I_2 was released into the dome and some I_2 reached the upper annulus. The I_2 concentration in the dome (Fig. 13) and in the bottom compartment is well simulated in 5 out of 7 calculations. Only in ASTEC-TUS and ECART-UNIPI is the I_2 concentration at 1.8 m elevation overestimated. The concentration in

the upper annulus is generally underestimated. In the test, some I_2 entered the upper annulus, which was not reproduced in the calculations. However, this local effect is of a minor importance. In the *transient phase*, when the atmosphere is mixed by the injected steam, $I_2(g)$ is acceptably simulated in most calculations except in ASTEC-GRS, COCOSYS-GRS and COCOSYS-FZJ at 1.8 m. In these calculations I_2 is underestimated more than 1½ order of magnitude in the bottom compartment. In the *mixed* and the *rest phases* a permanent concentration difference between the dome and the lower compartments (factor 2–9) and a moderate I_2 concentration decrease were measured. This local concentration difference between compartments in the vessel was simulated satisfactorily only in the COCOSYS-AREVA calculation. In all other calculations, the measured values were over- or underestimated and/or the concentration difference is zero.

In the *desorption phase*, the dome jacket was heated and some deposited I_2 was desorbed again. This process is correctly calculated only by COCOSYS-AREVA and COCOSYS-GRS. In ASTEC-GRS desorption is too high because chemi-sorption on steel is not treated in

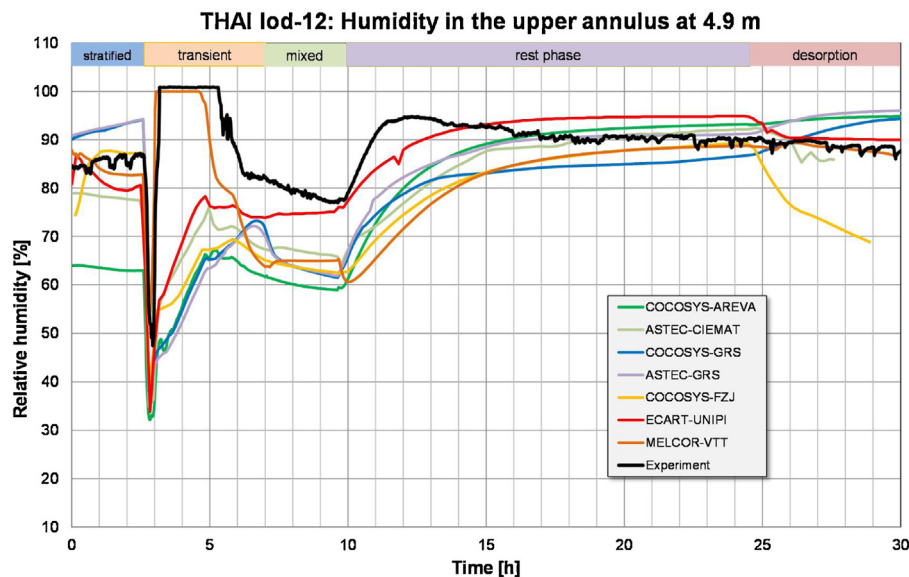


Fig. 11. Humidity in the upper annulus at 4.9 m in lod-12.

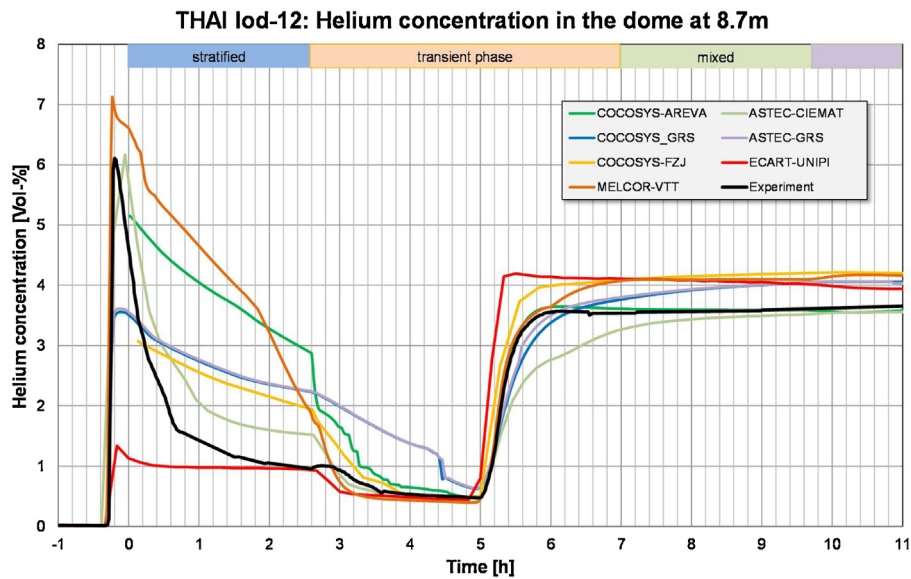


Fig. 12. Helium concentration in the dome at 8.7 m in lod-12.

IODE. The desorption constant used in the ASTEC-GRS calculation is the same as in the COCOSYS calculations, but at the start of resuspension all I_2 deposited on steel is still in a volatile form. An adapted temperature dependence of the desorption kinetics would reduce the discrepancy in the ASTEC-GRS calculation. In the other calculations this last test phase is not simulated or the I_2 desorption process is not modeled at all, like in ECART and MELCOR.

The gas scrubber measurement in the *inner cylinder* showed that this dead-end compartment was well mixed with the residual parts of the vessel. This was well matched only by ASTEC-CIEMAT. All other calculations failed strongly to reproduce the measured concentration. This indicates that the natural convection counter current flows in and out of the inner cylinder are not adequately simulated.

In the test, iodine was delivered into the main sump only by mass transfer from the bottom compartment atmosphere. The iodine

collected in the wall condensate was transported via the gutters in to the external tanks. As the main sump, pH = 1 no hydrolysis took place. However, $I_2(w)$ in contact with the immersed steel surface was converted to $I^-(w)$, which was accumulated in the sump. Therefore, the total iodine concentration is higher than the $I_2(w)$ equilibrium concentration due to the mass transfer in the long term. Three calculations show very little iodine in the main sump (COCOSYS-AREVA and COCOSYS-FZJ). Obviously the $I_2(w)/I^-(w)$ conversion is not treated correctly. In ECART-UNIPI aqueous iodine is not modeled at all. The other three results (COCOSYS-GRS, ASTEC-GRS and ASTEC-CIEMAT) are within a factor (0.2/5.0) of the measured value.

No calculation determines the iodine concentration accurately in all three condensate gutters. For gutter WB1, acceptable results are reached by COCOSYS-GRS and ASTEC-GRS. In all other calculations the concentration was too low or zero, also for WB2 where

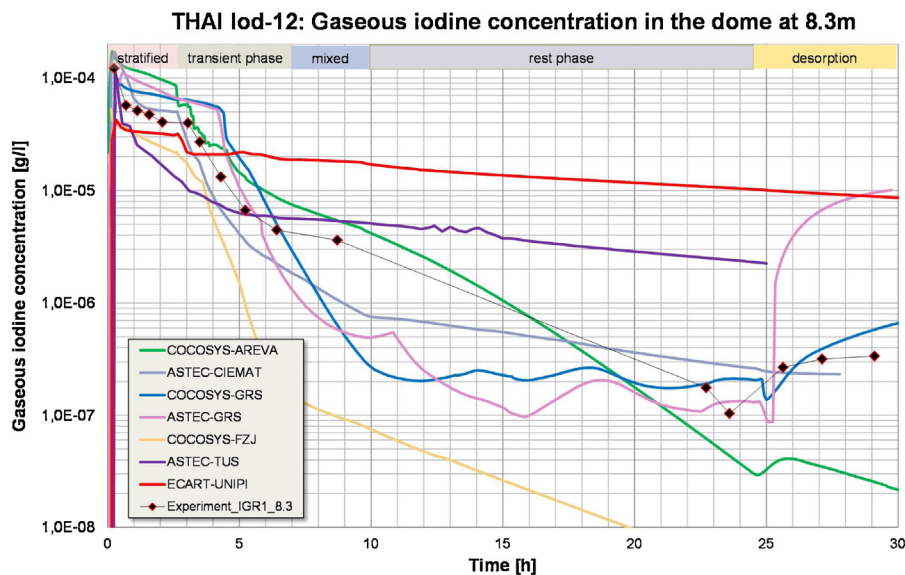


Fig. 13. Gaseous iodine in the dome at 8.3 m in lod-12.

the highest iodine concentration was measured. A more detailed interpretation of these uncertainties including the I_2 /wet surface modeling would be out of the scope of this paper.

4.3. Summary and assessment

In the THAI benchmark, analytical results of 9 calculations using 4 different codes (ASTEC, COCOSYS, ECART and MELCOR) were evaluated for the dry test Iod-11 and 8 for the wet test Iod-12. The relatively large THAI vessel with its five compartments allows many suitable possibilities for modeling. The number of control volumes in the nodalizations applied varied between 20 and 65 zones. The essential features, e.g. enough vertical levels, openings prepared for counter current flows, were mostly taken into account.

The main parameters looked at in the exercise were total pressure, temperature, relative humidity and helium concentration for the thermal-hydraulic part and iodine concentration in the gaseous phase, the sumps and the wall condensate for the iodine part.

4.3.1. Thermal-hydraulic modeling

In many calculations, the total pressure and the atmospheric temperature are well simulated, i.e. $\Delta p \leq 0.1$ bar respectively $\Delta T \leq 5$ °C. However, in some calculations the calculated local temperature exceeds the margin considerably. The consequence is a reduction in the accuracy of the humidity and convection flow rate parameters.

The sump temperature is overestimated significantly in some calculations, but this has only minor consequences on the iodine results since the boiling point is not reached throughout the calculations.

In none of the calculations is the measured relative humidity (rh) reproduced satisfactorily, i.e. the deviation from the measured values is at least temporarily larger than $\Delta rh = \pm 10$ rh%. Especially in the transient and the mixed phases, in which the vessel atmosphere is relatively well mixed, the deviations are rather large, typically $\Delta rh = \pm 20$ rh%. The reasons for this discrepancy are mainly inaccurately calculated local atmospheric temperatures and local steam concentrations respectively. This deviation is particularly important since it was found experimentally iodine interactions with surfaces are dependent on the local relative humidity (Weber and Funke, 2009; Nugraha, 1997).

In principle, the local He concentrations and the He distribution within the vessel are well calculated. However, in half of the calculations the mixing is too slow because of too weak convective flows. This can be attributed to shortcomings in the nodalization and the flow junction modeling.

4.3.2. Iodine behavior modeling

In several calculations for both tests, too much gaseous I_2 is transported from the dome into the lower compartments during the *stratified phase*, i.e. the atmospheric mixing is overestimated. This results probably from a weakness of the nodalization since the initial thermal stratification is correctly modeled in all calculations. In the *transient phase* the gaseous I_2 mixing between the dome and lower compartments is often too fast, although the helium mixing is too slow. This indicates that I_2 adsorption and/or desorption onto/from are not modeled correctly. The I_2 net deposition is underestimated by several codes. In the *mixed phase* the gaseous I_2 -concentration is overestimated in the whole vessel in half of the calculations. The main reason is an underestimation of the iodine deposition onto steel caused by inadequate iodine/steel models and an inaccurate simulation of the local humidity.

The iodine concentration in the wall condensate is underestimated in several calculations because of a faulty iodine wash-down modeling and inaccurate condensate flow rates. The iodine concentration in the two sumps is often underestimated, mainly because

the $I_2(w)$ to $I^-(w)$ conversion on immersed steel walls is not considered.

5. Lessons learned

The THAI benchmark on the multi-compartment iodine tests Iod-11 and Iod-12 delivered a wide spectrum of different results ranging from a good to a less good agreement. It shows the current status of the applicability of the models implemented into different codes, the importance of appropriate user experience with this complex phenomena and the need for a further model improvement. The major findings are:

- The local I_2 concentrations measured are mostly overestimated by up to two orders of magnitude. The main reasons for this discrepancy are: (a) inadequate iodine/steel models and (b) inaccurately calculated relative humidity and atmospheric flow rates.
- An adequate treatment of iodine interaction with the steel surface requires a model which can handle both physi-sorption and chemi-sorption reactions, although simpler models can be suitable if the rate constants are adjusted to the problem. In COCOSYS, physi-sorption and chemi-sorption are modeled. In ASTEC only physi-sorption is modeled and in ECART and MELCOR only irreversible iodine deposition processes can be treated. It is recommended to improve the incomplete iodine/steel models for dry and wet conditions. Under wet conditions, i.e. with wall condensation, iodine wash-down has to be considered. In several calculations the wash-down was underestimated or not simulated at all.
- The iodine/steel reaction and other iodine reactions depend on the relative humidity. The error of the local relative humidity was high in many calculations. The main reasons are inaccurate estimates of temperature and weak simulations of gas flows, which prevent a correct modeling of stratification or mixed conditions. With respect to an accurate iodine transport simulation these inadequacies in the thermal-hydraulic modeling should be reduced.
- In the THAI Benchmark the user effect is rather large, i.e. considerably different results were achieved with the same code ($2 \times$ ASTEC, $2 \times$ MELCOR, $3 \times$ COCOSYS). However, it is not so much different as in comparable International Standard Problem exercises on complex phenomena of severe accidents (Firnhaber et al., 1996; Clément and Haste, 2003). It may be reduced by an appropriate training comprising thermal-hydraulic and iodine issues. Nevertheless, the analysis of the multi-compartment iodine tests Iod-11 and Iod-12 has been an important validation step for the codes applied.

In a LWR containment the wall surfaces are usually coated with paint and only the main components are thermally insulated with a metal surface on top. In general, the I_2 transport processes between different compartments are expected to be comparable to those in the THAI steel vessel. The main differences are the much faster chemi-sorption reaction with the paint and the differences in the room volume/surface area ratio. As a next step the conduction of a multi-compartment test similar to Iod-11 and Iod-12 with (partly) painted surfaces, at dry and wet conditions, and their analyses is suggested. Also, a real plant application of the same codes with well-defined design specifics of the containment and boundary conditions similar to the THAI tests could prove useful for further defined future code development needs.

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