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Efficiency of selected methods of hydrogen removal from a nuclear reactor's containment building

Tomasz Bury*

Institute of Thermal Technology, Silesian University of Technology, Konarskiego 22, 44-100 Gliwice, Poland

Abstract

Large amounts of gaseous hydrogen may be released into the containment building during a severe accident in a water cooled nuclear reactor. The main methods of hydrogen removal from the containment are described in brief in this paper. HEPCAL—an in-house lumped parameter computer code—was used for simulation purposes and the results were used to evaluate the efficiency of various hydrogen removal systems.

Keywords: Nuclear reactor, Containment, Hydrogen, Hydrogen removal systems, Thermal-hydraulic analysis

1. Introduction

There are currently 434 nuclear power reactors in operation around the world and 402 units are water cooled and moderated (Boiling Water Reactors—BWR, Heavy Water Reactors—HWR and Pressurized Water Reactors—PWR) [1]. 62 of the 69 reactors under construction at present are of the water type.

Two main problems related to nuclear reactor operation are the presence of a large amount of highly radioactive material in the core and after-heat power.

A system of barriers is constructed to prevent a release of radioactive materials to the environment from the core. With water reactors these barriers take the form of: the nuclear fuel structure, fuel cladding, walls of the primary cooling circuit and the containment building. Individually they all feature very high reliability and are extremely unlikely to fail simultaneously.

Shutting down a nuclear reactor means stopping the chain fission reaction, but heat is still produced by decaying fission products. This is termed residual heat or after-heat power [2]. Therefore it is very important to assure sufficient core cooling in all operating modes and during accidents too.

Gaseous hydrogen may be generated in the overheated core region. The main source of this gas in the core region is the exothermic reaction of steam with the nuclear fuel cladding (zirconium alloy). The amount of hydrogen produced by this reaction is proportional to the mass of zirconium reacted [3]. This gas may be released into the containment building either through a break in the primary cooling circuit during a loss-of-coolant accident, or by a safety relieve valve (as in the Fukushima Dai-ichi nuclear power plant [4]). The mixing of hydrogen with the air in the internal atmosphere creates flammable mixtures. The problem of hydrogen combustion and detonation is a crucial issue for containment integrity, hence the need for prevention and gas removal measures.

The efficiency of two methods of hydrogen removal from the containment structure was assessed based on simulations made using the in-house com-

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*Corresponding author

puter code HEPCAL. This is a lumped parameter code for analysis of PWR type nuclear reactor containment transient response. Real systems utilizing the described hydrogen removal methods were simulated in the first step by applying design-basis accident scenarios. As the results could not be directly compared, in the second step a virtual experiment was simulated too.

2. Characteristics of hydrogen control systems in the containment building

There are five hydrogen control systems available:

- controlled combustion of hydrogen through intentional ignition,
- recombination of hydrogen by catalytic devices,
- removal of oxygen by pre-inertization,
- dilution of the atmosphere by post-accident injection of inert gas,
- dilution of the atmosphere by increasing the containment volume.

The first two methods from this list remove hydrogen from the containment. The other three methods decrease the hydrogen concentration in the internal atmosphere, but the amount of hydrogen in the containment remains the same.

Mixing the containment atmosphere in order to prevent high local concentrations of hydrogen is a key preventive measure as regards unintentional hydrogen combustion. Various factors affect the mixing process: notably, the arrangement of the containment building—its subcompartments, interconnections and layout of structures and equipment. The mixing of the containment atmosphere may be a passive process occuring due to gas flows inside the containment. These flows may be induced by coolant break flow, buoyancy effects originating from density differences in various areas and buoyancy effects due to the recombination of hydrogren in catalytic devices [5]. Cooling fans may also be used for mixing.

The choice of mitigation strategy depends on the design of the containment. At this point the following general observations are apposite [6]:

- pre-inertization is applied in the small Mark I and Mark II containments of most older boiling water reactors,
- more recent BWRs with Mark III containments, multi-units CANDU, and pressurised water reactors with ice condensers use igniters and sometimes PARs too,
- modern PWRs with large dry containments are equipped with catalytic recombiners.

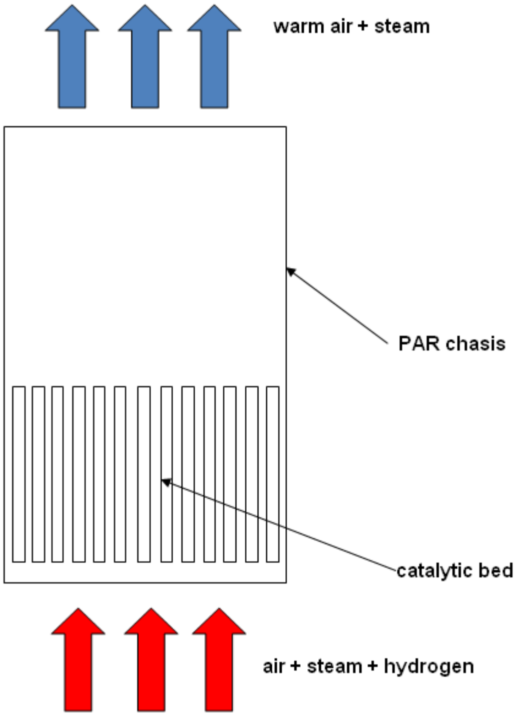

Figure 1: Passive Autocatalytic Recombiner—scheme of construction

In the European Union and Canada the main tendency for hydrogen hazard mitigation is to install catalytic recombiners in existing nuclear power plants for design-basis accident and beyond-design-basis accident. In contrast, passive catalytic recombiners are used for design-basis accident in Eastern Europe and the United States. A catalytic recombiner is a passive device—no external energy is needed for operation—and it is self starting even at low temperatures and in wet conditions. The recombiner consists of a vertical channel or stack equipped with a catalyst cartridge in the lower part. This design creates a chimney effect—a gas mixture flows through the recombiner by means of natural circulation. Underpinning the operation of a passive autocatalytic recombiner is the exothermic reaction of hydrogen and oxygen—present in the containment atmosphere—taking place on the surface of the catalyst. The catalytic cartridge contains plates or spheres coated with noble metals: palladium or platinum. The simplified diagram of a PAR is shown in Fig. 1.

Pre-inertization is a characteristic feature of BWR containments. The aim of this method is to remove or dilute oxygen by injecting inert gases such as carbion dioxide or nitrogen. Combustion prevention at all possible hydrogen concentrations requires the minimum initial concentrations of carbon dioxide or steam of 60% vol. and about 75% vol. for nitrogen [7]. This method is applied in Japan, the United States and Sweden.

Japanese BWRs also use igniters [6] in order to ignite a flammable mixture and thus consume hydrogen at lower concentrations. This action leads to local increases in temperature and pressure, but it is expected that the peak values of these parameters would be relatively low assuming that slow deflagration combustion will take place [5]. Distribution of igniters is crucial to prevent high local concentrations of hydrogren. Therefore, detailed knowledge is required of hydrogren distributions in the containment as well as places of possible rapid steam condensation. The igniter usually takes the form of a glow plug, similar to the ones used in diesel engines.

3. Numerical tool

The results were obtained from simulations performed using the HEPCAL-AD lumped parameter code. This code was developed at the Institute of Thermal Technology of the Silesian University of Technology [8]. The code is designed to predict changes in thermodynamic parameters within containment during a loss-of-coolant accident (LOCA). The whole containment is simulated by a couple of interconnected zones (volumes). Usually, the geometry and dimensions of a control volume correspond to the real dimensions of the specified compartment of the accident localization system. The control volumes are connected through open channels, orifices, valves, membranes or siphon closures. Homogeneous conditions (perfect mixing) are assumed for each zone. Hence, the applied model is discrete in reference to space and time as well. Its bases are the energy balance equations written for each specified control volume in the given time span (time step) Dt.

Energy streams flowing in and out of the control volume are associated primarily with heat transfer to walls and structures and intercompartment flows of media. When modeling it is crucial to take into account the operation of safety systems. Mass and energy streams resulting from the operation of pumps, fans and other devices should be considered in the energy balance. These quantities as well as the initial internal energy U_1 are determined based on the values of thermodynamic parameters at the beginning of the time step.

Transforming the general relationship for the energy balance in a control zone, one obtains:

$$U_2 = (\dot{E}_{in} - \dot{E}_{out})\Delta\tau + U_1 \tag{1}$$

where $\dot{E}_{in}$ and $\dot{E}_{out}$ are the energy flow rates flowing into and out of a control zone respectively.

The right hand side components of the equation (1) are known, so the internal energy at the end of the time step can be calculated. Unknown thermodynamic parameters at the end of the time step are functions of the internal energy U_2.

This approach enables one dimensional analysis and determination of time dependent changes of basic thermodynamic parameters (temperature, pressure) within the containment building. It should be clearly stated that the model does not include processes taking place within the primary cooling circuit. Data relating to coolant leakage (mass flow rates and specific enthalpy) are the boundary conditions for the HEPCAL code. These pieces of information are taken from external programs.

The mathematical basis of the model describing changes in thermodynamic parameters consist of the equations of mass and energy balance for specified phases and equations of state [8–10]. The equations of mass and energy balance apply to the time step $\Delta\tau$, but the equations of state concern the end of each time step. All the equations are nonlinear and their form depends on the state of the specified agents in the control volume. The basic set of equations constituting the mathematical model consists of:

- equations of the energy and mass balance for each control volume,
- equations describing intercompartment flows,
- equations of state for the specified gaseous agents (air, steam, hydrogen),
- equations describing additional phenomena, e.g., heat transfer to the walls and structures, operation of safety systems.

As thermodynamic nonequilibrium between states is assumed, the basic equations mentioned earlier may take a different form, depending on the actual state of water and steam within the control zone. The model includes six possible cases:

- lack of water, superheated steam,
- subcooled water, superheated steam,
- subcooled water, saturated steam,
- saturated water, superheated steam,
- saturated water, saturated steam,
- lack of water, saturated steam.

Determination of unknown parameters is a gradual process. In the first step mass and energy streams are determined, e.g., leakage of coolant from the primary circuit, media flows through the intercompartment junctions, mass flow rate of water from the spraying system, accumulation of heat in walls and structures. Heat transfer between phases is also calculated in this step. All these quantities relate to the beginning of the time step and enable one to determine the internal energy of gas and liquid.

Taking this into account the amounts of media, as well as the internal energy at the end of the time step are computed in the second step. The amounts of steam and water and their internal energies initially are determined neglecting the phase changes during the time step.

In the third step the equations listed above are used to calculate the values of the basic thermal parameters. Only the values which are valid for the actual state of media within the control volume are chosen. Eventually one obtains a system of nonlinear equations, which is solved using the Newton-Raphson method. A number of equations in the system depends on the current state of agents in the control zone. The calculating process is repeated in each time step for every control zone as long as the desired accuracy is achieved.

In the last step the values of the remaining unknown parameters (pressures, volumes and final masses of agents) are computed from basic thermodynamic laws and geometrical relationships.

The model applied in the HEPCAL code allows one to determine the thermal parameters (temperature, pressure, density) in the specified volumes and the mass flow rates as well as the energy transfer rates between the control zones. The spraying system work is taken into account as well as the heat transfer between phases and heat accumulation in the structures of the containment.

4. Numerical simulations

4.1. Analyzed systems

The main problem in selecting a system for analysis comes from the the use of different hydrogen removal methods (such as pre-inertization or igniters), mainly for boiling water reactors. In order to make a meaningful comparison of operating results, the analysis should be performed for similar systems. It was decided that the containment structure for one selected reactor would be analyzed, but with different hydrogen removal systems in subsequent simulations. The VVER-440/213 reactor was chosen, as its containment construction comprises some features that are characteristic of both boiling water and pressurized reactors.

Pressurized water reactors of the VVER- 440/213 type have a containment building which is connected with a bubble condenser. The bubble condenser acts as a pressure suppression system by condensation of released steam. Specific features of the VVER 440/213 containment are the subdivided rectangular building and the localization tower includ-

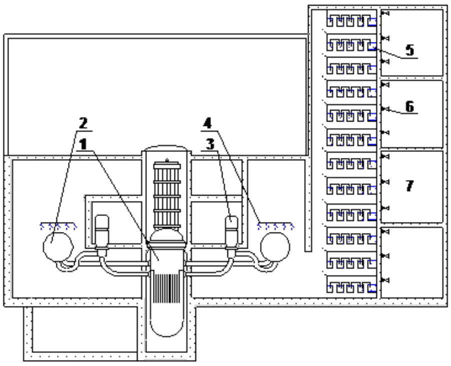

Figure 2: Simplified sketch of the VVER 440/213 reactor containment (1—reactor pressure vessel, 2—steam generators, 3—reactor coolant pumps, 4—spraying system, 5—water trays, 6—check valves,7—air traps)

ing the bubbler trays and air traps (Fig. 2). The containment is designed to prevent the escape of steam and fission products in any loss of coolant accident cases, including the double ended guillotine rupture of a 500 mm diameter main circulation pipe (this is the design-basis accident—DBA) [11]. The design pressure of the containment is 0.25 MPa.

The accident localization system consists of the bubble condenser and the air traps. The aim of the localization system is to decrease the maximum pressure and to ensure near atmospheric pressure after 5 to 10 min of the pipe break [12]. The localization tower contains about 1500 m^3 of water distributed among 12 levels of trays. The air volume of trays is connected to four air traps through the check valves. The steam condenses, flowing to the water trays through a layer of water. Non condensable gases and air accumulate in the gaseous space of the water trays and, after crossing the border pressure, flow to the air traps.

Four variants of hydrogen removal system were analyzed for the described containment.

The first variant assumes that the system under consideration is equipped with 28 passive autocatalytic recombiners of type FR1-1500T and 4 devices of type FR1-750T produced by AREVA. The nominal capacity of these PARs is 160 kg of hydrogen per hour for reference conditions (absolute pressure 150 kPa, temperature 60°C and hydrogen concentration of 4%). The PARs start operation at a hydrogen concentration of 2% (volume fraction) [13].

Variants two and three areoxygen dilution methods. In the second variant it was assumed that the internal atmosphere is pre-inerted with nitrogen. The initial concentration of nitrogen is 75% by volume. The third variant assumes post-accident injection of inert gas (nitrogen). The injection rate is a constant 5 kg/s during the accident. As nitrogen is an inert gas and it impairs the heat transfer conditions, the injection was assumed to start when the first portion of hydrogen is released into the containment building from the primary circuit.

The fourth variant is a hydrogen removal system based on hydrogen igniters. This case is the most problematic due to a lack of sufficient data. According to [5] it was assessed that the analyzed containment system may need about 100-120 igniters. It is devoid of purpose to apply this number to the lumped parameter code HEPCAL, as the code operates on average values of parameters within each control zone (perfect mixing conditions). Therefore, it was assumed that instead of a large number of igniters only one “lumped” igniter is present in each control zone and that it needs 15 seconds to achieve the required temperature of ignition. The other problem is that the igniters should prevent high local concentrations of hydrogen and such information is unavailable in the lumped parameter approach. An optimized arrangement of igniters means that combustion starts as soon as possible [5]. Taking this into account it was assumed that in each control zone the igniter will be activated every five minutes and it will cause burn out of 80% of the hydrogen present in the zone. The first activation will take place five minutes after the first portion of hydrogen appears within the containment. In view of the above, significant uncertainty is expected with regard to the results of the simulation for this case.

4.2. Numerical model and boundary conditions

According to the requirements of the applied mathematical model the containment structure under consideration was divided into nine control volumes. The nodalization scheme is shown in Fig. 3. and is as follows: zone 1—volume of 6370 m^3 (half of the steam generator boxes); zone 2—volume of 6370 m^3 (half of the steam generator boxes); zone 3—volume of 2000 m^3 (connecting channel); zone 4—volume of 3000 m^3 (the shaft of the accident lo-

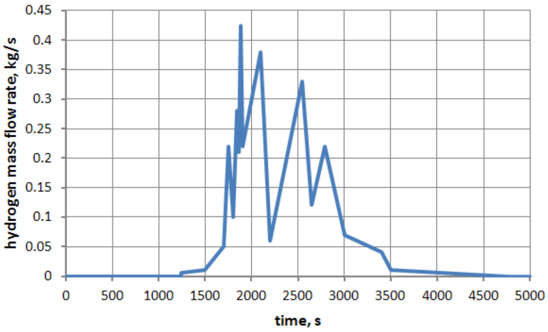

Figure 3: Nodalization scheme of the VVER-440/213 containment building

calization tower); zone 5—volume of 2667 m^3, including 500 m^3 of water (water trays—4 lower levels); zone 6—volume of 5333 m^3, including 1000 m^3 of water (water trays—remaining levels); zone 7—volume of 4200 m^3 (first air trap); zone 8—volume of 12 600 m^3 (other air traps); zone 9—volume of 6000 m^3 (closed subcompartments connected to the steam generator boxes with open channels of constant flow cross-section area).

In the above figure the continuous lines denote junctions by open channels of constant flow cross-section area and dashed lines denote junctions by siphon closures.

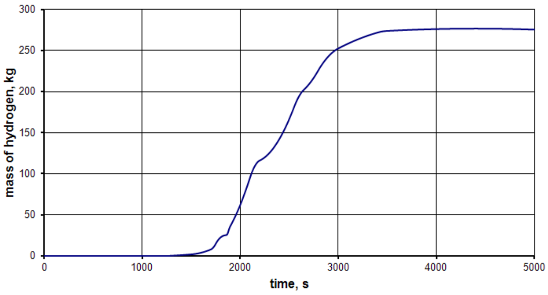

Figure 4: Mass flow rate of hydrogen flowing into the break zone

The analyzed accident scenario is medium break loss-of-coolant accident (LOCA). The accident is initiated by a rupture of the primary circuit pipe with an effective diameter of 100 mm. The break takes place in the steam generator boxes (zone 1). The low and high pressure emergency coolant injection as well as the active spraying system are unavailable during the accident [14]. Hydrogen is produced in the steam-zirconium reaction within the core region and is then released into the containment via the break. Fig. 4 presents the mass flow rate of hydrogen released to the break zone. Hydrogen temperature was assumed constant at 500°C.

4.3. Results of simulations

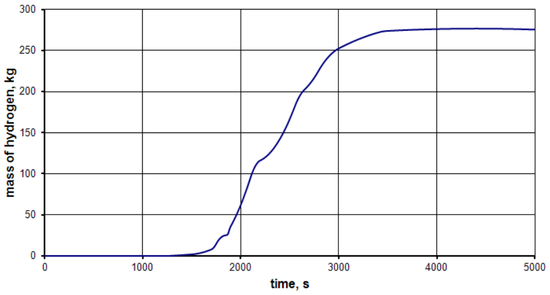

Figure 5: Mass of hydrogen accumulated within the break zone—simulation without operation of hydrogen removal system

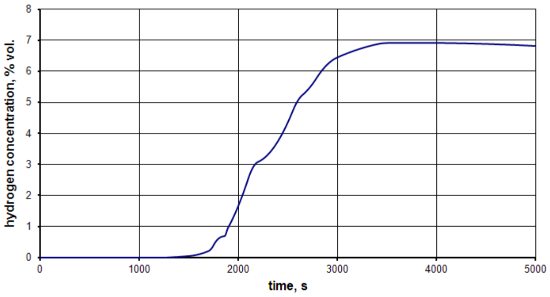

Figure 6: Hydrogen concentration trend within the break zone—simulation without operation of hydrogen removal system

The most interesting results are the mass of hydrogen released into the containment and its concentration. The time dependent trends of these parameters are presented in Figs 6, 7, 8. The results concern control zone number 1 where the rupture of the primary circuit was assumed.

The first stage of simulations were made without operation of the hydrogen removal system. The results are shown in Figs 5 and 6. Taking into account a 4% flammability limit it can be seen in Fig. 6 that this limit is achieved within about 20 minutes after the first portion of hydrogen appears in the steam generator boxes.

The PARs based hydrogen removal system operation was simulated in the next step. Computations were accomplished assuming constant capacity of the catalytic devices. As mentioned earlier, the PARs

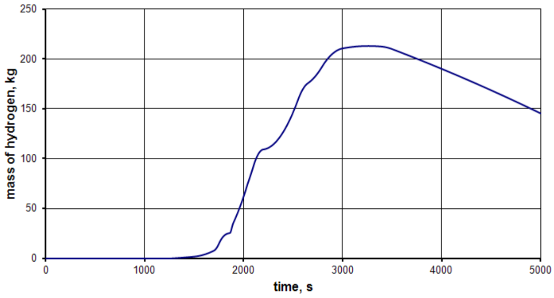

Figure 7: Mass of hydrogen accumulated within the break zone—simulation with PARs operation

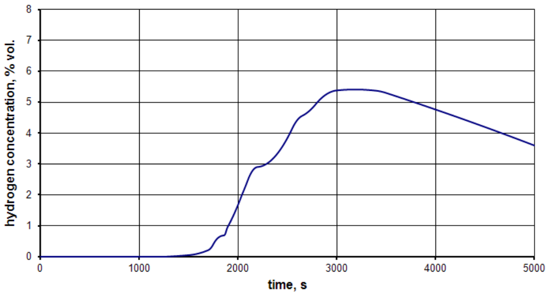

Figure 8: Hydrogen concentration trend within the break zone—simulation with PARs operation

are activated after crossing the 2% hydrogen concentration limit. The hydrogen removal system operation enables over 50 kg of this gas to be recombined within the analyzed time period, as shown in Fig. 7. The hydrogen flammability limit is crossed in this case too (see Fig. 8), but the concentration of hydrogen falls below this limit within about 37 minutes.

The third analysed case is the hydrogen removal system variant based on pre-inertization of the internal atmosphere with nitrogen. The initial nitrogen concentration in the air and nitrogen mixture is 75% and this means that the initial oxygen concentration is reduced to a level of about 5.2%. The mass of hydrogen accumulated within the break zone in this case is the same as in the first analyzed scenario without hydrogen removal (see Fig. 5), as this gas is not removed from the containment.

The hydrogen concentration trend is shown in Fig. 9 for that case and is compared with the post-accident nitrogen injection case. As can be seen, in both cases the maximum hydrogen concentration is far below the flammability limits.

The results for the last analyzed case are presented in Figs 10and11. Some simplifications were made in

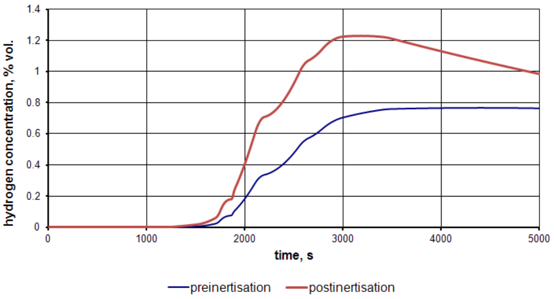

Figure 9: Hydrogen concentration trend within the break zone—comparison of preinertization and postinertization cases

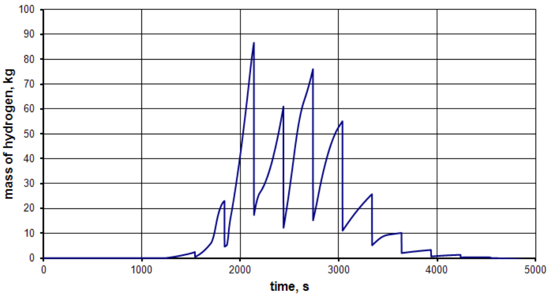

Figure 10: Mass of hydrogen accumulated within the break zone—hydrogen igniters case

modeling the combustion of hydrogen—a very simple model of reaction kinetics was applied. The presented trends clearly show that the igniters should not be time controlled. Such a solution may lead in some cases to the combustion of large amounts of hydrogen (see Fig. 10). Igniter initiated hydrogen combustion should not cause excess pressure loads of more than 30 kPa [5]. In the analyzed case these loads reached almost 180 kPa. However, this could be an effect of combustion modeling—it was assumed that hydrogen is burnt up immediately after ignition.

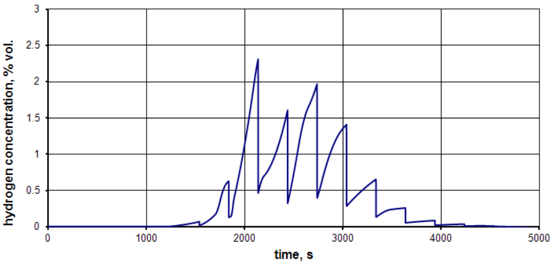

Figure 11: Mass of hydrogen accumulated within the break zone—hydrogen igniters case

The results shown in Fig. 11 suggest that autoignition of hydrogen would not take place (the 4\%

flammability limit was not exceeded).

5. Conclusions

Some "artificial" systems were included in the simulations reported in this paper for the express purpose of comparing different methods of hydrogen risk mitigation. In particular, the inertization methods are not practical for the considered containment of the VVER-440/213 reactor due to the large volume of this system.

According to the results of the analyzes presented here it may seem that oxygen dilution by pre-inertization or post-inertization is the most efficient way to mitigate the hydrogen hazards within containments of water reactors. The hydrogen igniters seem to be a good solution too. However, these devices need an electric power supply to operate and very detailed information on hydrogen behavior in the containment in order to properly distribute them. Passive autocatalytic recombiners look the worst solution, but it should be noted that they do not need any external supply and they are self-initiating devices.

When evaluating these results, it should be clearly noted that the analyzes were performed with a lumped parameter code. Such a code assumes perfect mixing within the control volume. Therefore the results of simulations are subject to large uncertainties. It is self-evident that near the break the hydrogen concentration will cross the flammability limits much earlier than is predicted by the code.

Crossing the flammability limits does not mean automatic hydrogen auto-ignition—when there is a large amount of steam it prevents the combustion of hydrogen. On the other hand, rapid condensation of steam may lead to high local concentrations of hydrogen and may create detonable mixtures. In order to obtain knowledge of local distributions of hydrogen, more detailed modeling is required than is available under a lumped parameter approach.

There is another problem in relation to the hydrogen risk: hydrogen combustion may be caused by an electric spark or when the gas stream hits a hot surface. These are stochastic events and it is impossible to factor them in with any semblance of satisfactory accuracy.

In summary, the lumped parameter approach for modeling containment thermal-hydraulic distributions may lead to large uncertainties in some cases. Evaluating these uncertainties is extremely problematic.

Acknowledgments

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References

[1] International Atomic Energy Agency: Power reactor information system – PRIS. Available at http://www.iaea.org/pris/ - Access on September 29 2013.

[2] J. Lamarsh, A. Baratta, Introduction to Nuclear Engineering, Prentice Hall, New York, 2002.

[3] De Boeck B., Introduction to severe accidents. Especially the containment behaviour. AIB VINCOTTE NUCLEAR report AVN 97/013.

[4] International Atomic Energy Agency: Nuclear Emergency Response Headquarters – Government of Japan: Report of Japanese Government to the IAEA Ministerial Conference on Nuclear Safety. IAEA, Vienna, June 2011.

[5] B. Sehgal, Nuclear safety in light water reactors. Severe accident phenomenology, 1st Edition, Elsevier Academic Press, 2012.

[6] B. E, A. F., Generic approach for designing and implementing a passive autocatalytic recombiner par system in nuclear power plant containments., Nuclear Engineering and Design 275 (2002) 1–15.

[7] International Atomic Energy Agency: Mitigation of hydrogen hazards in water cooled power reactors. IAEA-TECDoc 1196, Vienna, 2001.

[8] T. Bury, Analysis of thermal and flow processes within containments of water nuclear reactors during loss-of-coolant accidents, Ph.D. thesis, Institute of Thermal Technology, Silesian University of Technology, Gliwice (2005).

[9] A. Fic, J. Skorek, Mathematical model of transient thermal and flow processes in containment of a pwr nuclear reactor, Archiwum Energetyki 1-2 (1993) 19–32.

[10] J. Skorek, J. Składzień, Thermal analysis of the loss-of-coolant accident within the containment of the wwer-440 and wwer-1000 nuclear reactors, Computer Assisted Mechanics and Engineering Sciences 1 (3/4) (1994) 217–231.

[11] T. Bury, Influence of the accident localization system construction on the course of the primary circuit rupture accident in vver 440 units, Zeszyty Naukowe Politechniki Warszawskiej, seria Konferencje 1 (2002) 169–176, (in Polish).

[12] Z. Techy, G. Lajtha, R. Taubner, Accident loads for vver-440/213 containment, Nuclear Engineering and Design 157 (1995) 375–385.
[13] AREVA: AREVA passive autocatalytic recombiner. Document G-008-V1PB-2011-ENG, AREVA, Paris, 2011.
[14] Kostka P., Techy Z. and Sienicki J., Hydrogen mixing analyses for a VVER containment. Proceedings of 10th International Conference on Nuclear Engineering, Arlington, 2002, paper ICONE10-22206.