# INFLUENCE OF OXIDATION ON FUEL COOLANT INTERACTION

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#### **ABSTRACT**

When during a severe reactor accident the molten core comes into contact with the coolant water a steam explosion may occur. A steam explosion is an energetic fuel coolant interaction process during which the energy of the molten corium is transferred to the coolant water on a timescale smaller than the timescale for system pressure relief and so induces dynamic loading of surrounding structures. Steam explosions are an important nuclear safety issue because they can potentially jeopardize the primary system and the containment integrity of the nuclear power plant.

If non-oxidized zirconium is present in the corium melt the oxidation of zirconium may significantly influence the fuel coolant interaction process, as observed in experiments. In the ZREX experiments the steam explosion strength was largely increased by the oxidation zirconium, whereas in the recently performed OECD SERENA project KROTOS and TROI experiments it seems that the oxidation had an inhibiting effect. To find out the reasons for this qualitatively different behavior, the experimental results were investigated in comparison and computer simulations were performed.

In the paper, the performed analysis of the influence of oxidation on the steam explosion energetics is presented and discussed. Based on the experimental findings, the hydrogen film hypothesis is proposed, claiming that only a limiting amount of zirconium may be oxidized during the premixing phase in subcooled conditions and that the remaining non-oxidized zirconium is available for the oxidation in the explosion phase. Various computer simulations were performed with the MC3D code to support the hypothesis and to get additional insight. It may be concluded that the proposed hypothesis reasonably well explains the observed experimental findings.

**KEYWORDS** 

Fuel coolant interaction, zirconium oxidation, hydrogen production, steam explosion simulation

#### 1. INTRODUCTION

The importance of severe accident research has been again seen after the March 2011 Fukushima nuclear reactor accident in Japan. As the accident analysis showed, it seems that in the Daichi Unit 3 a steam explosion occurred [1]. A steam explosion, in the frame of nuclear reactor safety, is an energetic fuel coolant interaction (FCI) process, which may occur during a severe accident when the hot reactor core melt (corium) comes into contact with the coolant water [2] [3]. During the steam explosion the energy of the molten corium is transferred to the coolant water in a timescale smaller than the timescale for system pressure relief and so induces dynamic loading of surrounding structures. Steam explosions are an important nuclear safety issue because they can potentially jeopardize the primary system and the containment integrity of the nuclear power plant [4] [5].

Steam explosion experiments have revealed that there is an important influence of the material properties on the FCI [6] [7]. During a severe reactor accident the corium melt interacting with the water may have various compositions, which depend on the severe accident scenario, and thus it may have different physical and chemical properties. One of the key phenomena, which is not well understood, is the oxidation of the corium melt during the FCI in case of the presence of non-oxidized zirconium. During the oxidation of zirconium heat is released and hydrogen is formed. The influence of oxidation on the FCI is very complex [8]. The released heat has a promoting effect on the steam explosion energetics due to the increased energy content of the melt and due to the delayed solidification. But at the same time it has also an inhibiting effect due to the increased void build-up. High void reduces the steam explosion energetics due to lack of water, which is essential for the development of a steam explosion, and due to compressibility effects. Also the hydrogen production has at the same time a promoting and an inhibiting effect on the steam explosion energetics. As hydrogen is a non-condensable gas the expansion process is not diminished due to condensation, as in case of vapor, which increases the energetics. But on the other hand, hydrogen increases the stability of the vapor film and increases the void fraction, which reduces the triggerability of the explosion and the energetics. Namely, for steam explosion triggering the melt has to come in direct contact with liquid water, which is hindered if hydrogen is present in the vapor film around the melt droplets because hydrogen cannot condense. There are also indirect consequences of the oxidation on the energetics. It seems that the oxidation promotes the jet-breakup; therefore the mass of melt droplets is larger, which has a promoting effect. But if the droplets at triggering time are older (increased solidification) or if smaller droplets are formed (increased solidification and void build-up) this reduces the energetics. The oxidation issue was recognized for a while as given for example in [9], however the understanding of the phenomena is far than being well understood.

The influence of the oxidation on the FCI was investigated in the ZREX premixing and explosion tests [10], performed at the Argonne National Laboratory in the USA, and in the OECD SERENA project explosion tests [11], performed in the KROTOS (CEA, France) and TROI (KAERI, Korea) facilities. The main purpose of the ZREX tests was to investigate the possible chemical augmentation of the FCI energetics due to the zirconium-water reaction, whereas in SERENA the influence of the corium composition on the FCI was analyzed, which included also the influence of the oxidation process for a sub-stoichiometric melt (U,Zr)O<sub>2-x</sub>. In the ZREX experiments performed with simulant ZrO<sub>2</sub>/Zr and SS/Zr (stainless steel-zirconium) melts the explosion energetics were significantly augmented by the zirconium-water interaction, suggesting that the extent of chemical augmentation is approximately proportional to the zirconium content in the melt. The only experiments addressing this important oxidation issue with prototypic melt, i.e. with nuclear fuel UO<sub>2</sub>, were performed recently in the frame of the OECD SERENA project in the KROTOS and TROI facilities. In each facility one experiment was performed with corium UO<sub>2</sub>/ZrO<sub>2</sub>/Zr melt, whereas the other experiments were performed with oxidic corium. Both experiments with added zirconium resulted in the least energetic FCI of the whole test series (six experiments in each facility). Thus at first glance it seems that the influence of oxidation on the steam explosion energetics is different for prototypic melt, as used in the SERENA experiments, than for simulant melt, as used in the ZREX experiments. To find out the reasons for this qualitatively different behavior the experimental results were investigated in comparison and various computer simulations were performed to get additional insight.

In the paper, first the comparative analysis of SERENA and ZREX experimental results is described. Next the hydrogen film hypothesis is proposed, which might reasonably well explain the observed experimental findings. Then the parametric oxidation influence study performed with the MC3D code [12] [13] is presented and the results are discussed.

#### 2. EXPERIMENTAL OBSERVATIONS

#### 2.1. ZREX and SERENA Oxidation Tests

Steam explosion experiments are carried out by releasing the melt into a test tube filled with water, where the dynamic FCI takes place. An external trigger is mounted at the bottom of the interaction section to initiate the steam explosion. Several sensors are installed to measure and observe the temperature, global pressure, dynamic pressures, dynamic force, mixing of the melt in the water and the amount of generated hydrogen. The dynamic pressures during the FCI are measured using piezoelectric transducers mounted on the test section wall. The ZREX experimental facility has the smallest test tube among the considered facilities, but a relatively large containment vessel free volume. The test sections of ZREX and KROTOS are of similar one-dimensional tube geometry. Because of the small radial dimension of the test section with rigid walls, the explosion propagation and expansion is essentially one-dimensional, maximizing the axial output of the mechanical energy release. The TROI facility was designed for testing the FCI behavior in more reactor-like conditions by having more mass and multidimensional melt water interaction geometry. Two series of ZREX experiments were performed with simulant melt varying the Zr content [10]:

- ZREX series: 1.0 kg of Zr/ZrO<sub>2</sub> mixtures, Zr content from 60 wt.% to 100 wt.%,
- ZRSS series: 1.2 kg of Zr/SS mixtures, Zr content from 0 wt.% to 100 wt.%.

In the SERENA test series Zr was added in the prototypic oxidic corium material in the fifth tests [11]:

- KROTOS test KS-5: 2.4 kg of UO<sub>2</sub>/ZrO<sub>2</sub>/Zr mixture, Zr content 8.5 wt.%,
- TROI test TS-5: 17.9 kg of UO<sub>2</sub>/ZrO<sub>2</sub>/Zr mixture, Zr content 5 wt.%.

The released mechanical energy during the steam explosion was estimated from the measured dynamic pressures. Fig. 1 shows the explosion strength results, depicting the conversion ratios between the mechanical energy and the total available energy (stored thermal energy plus chemical energy) plotted as a function of the Zr content for all oxidation tests, performed in similar sub-cooled conditions. In the left diagram the increasing explosion strength may be observed for the ZREX experiments with increasing the Zr content. The right diagram presents the expanded view of the low Zr percentage range with SERENA results. The KS-5 result is missing due to experimental data acquisition problems, but there are indications that the FCI was weak. The two vertical braces present the range of conversion ratios for the whole KROTOS and the whole TROI series (six experiments in each facility with various corium compositions, with Zr added only in the fifth tests). The SERENA results are presented only qualitatively without numbers because they are still restricted only to SERENA project members. The decreasing (TROI) and the increasing (ZREX) trends of the explosion strength are schematically marked with the grey dashed lines. The dashed line for TROI was plotted through a typical value of the conversion ratio for the oxidic corium melt without added Zr and the TS-5 result. It seems that the results exhibit reduced explosion strength at low Zr content (up to about 10%) and gradually increasing explosion strength for Zr content larger than about 30%.

It should be mentioned that the observed reduced explosion strength in SERENA experiments with added Zr is not necessarily a direct consequence of Zr oxidation, but could occur due to various other reasons, thus the results are not conclusive. In KS-5 the explosion was triggered later than in other experiments, resulting probably in increased melt solidification at explosion triggering (due to longer quenching times), which reduces the explosion strength. In a steam explosion only effectively liquid melt droplets may efficiently participate because effectively solid droplets may not undergo fine fragmentation. And in TS-5 maybe a small spontaneous steam explosion occurred before the planned artificially triggered one, resulting possibly in highly voided pre-mixture regions at the time of the planned triggering, which could inhibit the development of a strong explosion. In the following we will assume that in TS-5 the explosion strength was reduced due to oxidation effects.

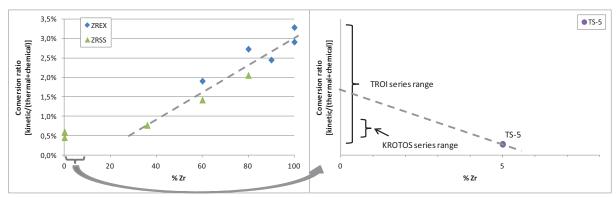


Figure 1. ZREX conversion ratios (left) and SERENA TS-5 conversion ratio with estimation of KROTOS and TROI conversion ratio ranges (right). SERENA results are presented without numbers because they are still restricted only to SERENA project members.

# 2.2. Hydrogen Film Hypothesis

## 2.2.1. Produced Hydrogen

In ZREX the oxidation of Zr was more or less complete for many explosion tests. An interesting characteristic is observed in ZREX non-triggered tests (premixing tests without the succeeding explosion) examining the absolute amount of produced hydrogen. Fig. 2 presents the measured hydrogen amount plotted against the Zr content (left diagram) and against the water sub-cooling (right diagram) [10]. All experiments produced approximately 1 to 2 moles of hydrogen for melt compositions with 0% to 100% of Zr, with two exceptions: the ZREX-23 and ZREX-27 tests, which produced significantly more hydrogen. The reason seems to be the water sub-cooling as depicted in the right diagram. All experiments were carried out with 75 K to 80 K of water sub-cooling, except ZREX-23 (30 K) and ZREX-27 (10 K). At 0% of Zr the result presents the hydrogen produced by the oxidation of stainless steel metals because there is no Zr available for oxidation.

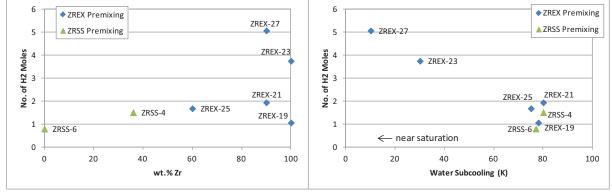


Figure 2. ZREX non-triggered tests: number of moles of hydrogen produced as a function of zirconium content (left) and water sub-cooling (right).

## 2.2.2. Premixing Phase

The results in Fig. 2 suggest that the oxidation in terms of the absolute amount of produced hydrogen during premixing in sub-cooled conditions is rather independent on the amount of Zr in the melt, presuming that a minimum amount of oxidizable (metal) material is present in the melt. It seems that sub-cooled conditions allow only a limited extent of oxidation, whereas in saturated conditions the capacity for oxidation increases. Our simplified explanation for this behavior is that the absolute capacity for oxidation is limited by the amount of vapor that is available in the gaseous film initially formed around the melt droplets. After the melt droplets are formed by the melt jet fragmentation, the gaseous film gets filled with hydrogen which is produced on the surface of the droplet by the oxidation reaction with the vapor, as depicted in Fig. 3. When the gas film is mainly filled with hydrogen the oxidation process stops or it is significantly reduced (reductive conditions). The absolute capacity for oxidation is therefore dependent on the total surface area of the melt droplets formed during the premixing. In saturated conditions the vapor does not condense on the water-vapor interface and so the hydrogen gas flows away from the droplet together with the generated vapor. The inflow of fresh vapor to the droplet surface is therefore enabled and thus the capacity for oxidation during premixing is increased.

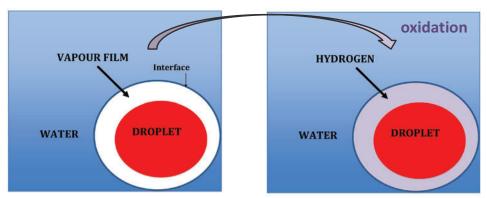


Figure 3. Filling of gas film around droplet with hydrogen due to oxidation for sub-cooled conditions.

The limited capacity for oxidation during premixing implies that in a given melt composition only a limited absolute amount of Zr may be oxidized during the premixing in sub-cooled conditions. It means that if the amount of Zr is equal or smaller than the limiting amount (threshold), the oxidation in premixing is complete (100%), and if the amount of Zr is larger than the threshold, the remaining non-oxidized Zr is available for oxidation during the explosion phase.

#### 2.2.3. Explosion Phase

During the explosion phase the melt droplets undergo fine fragmentation and the fine fragments disperse in the water. Here the amount of vapor around each fine fragment should be sufficient to fully oxidize the residual Zr. The threshold Zr amount for typical sub-cooled conditions is estimated to be around 10 wt.% of the melt [14]. If the Zr content in the melt is equal or smaller than the threshold (then the oxidation in premixing is complete) the explosion strength will depend solely on the competing promoting and inhibiting effects of oxidation during the premixing, and if the Zr content is larger than the threshold, the explosion strength will also depend on the amount of the remaining Zr that is fully oxidized during the explosion phase. In the TS-5 and KS-5 tests the Zr content was lower than the estimated threshold, thus the oxidation in premixing should be complete, as is suggested also by the performed extensive KS-5

debris analysis [15]. It seems that the oxidation during the premixing has an inhibiting effect on the explosion energetics according to the reduced explosion strength in TS-5 and KS-5 (but as stated before the results are not conclusive). The Zr content in the ZREX tests was larger than the estimated threshold, suggesting that any oxidation during the explosion phase significantly contributes to the explosion energetics. For late triggering the oxidation during the premixing has a promoting effect on the steam explosion energetics because the released heat during the oxidation reaction delays the solidification and thus more liquid melt is available at explosion triggering.

It should be stressed that the provided explanation is only a very simplified picture. In reality the situation is much more complex because only the hot melt in contact with the vapor may oxidize. Thus the oxidation during premixing depends also on the quenching time scale, and during the explosion only melt droplets which undergo fine fragmentation may oxidize. To get a better insight in the effect of oxidation, parametric simulations were carried out with the MC3D computer code.

#### 3. COMPUTER SIMULATIONS

# 3.1. Modelling and Calculation Conditions

The simulations were performed with the MC3D v3.8 computer code, which is being developed by IRSN, France [12] [13]. It is a multi-dimensional Eulerian code devoted to study multi-phase and multi-constituent flows in the field of nuclear safety. The steam explosion simulation is being carried out in two steps. First the premixing phase is simulated and then the succeeding explosion phase, using the premixing simulation results as initial conditions and applying an explosion trigger. MC3D is one of the few codes that can model oxidation and handle the presence of non-condensable gases. The reason for the lack of oxidation models in FCI codes is the complex influence of oxidation on FCI, which is not yet well understood, and the associated uncertainties and modelling difficulties. The performed study is one of the first tries to simulate the important oxidation influence.

The simulations were carried out in the KROTOS geometry in the 2D axisymmetric cylindrical coordinate system on a mesh with 20 x 120 cells and with the calculation domain size 0.356 m x 2.6 m (Fig. 4). Standard calculation models and adequate modelling parameters (from detailed simulation analysis of KS-5 test [16]) were chosen. The experimental conditions and material properties in the KS-5 test were applied in all calculations (Table I), only the oxidation parameters were adequately varied. The prescribed characteristic time for the oxidation in the premixing phase was 0.1 s and 0.001 s in the explosion phase. The explosion calculations were carried out for two triggering times: at the moment when the melt penetrated 0.3 m into the water (similar to ZREX experiments) – early triggering at 0.47 s, and at the moment when the melt arrived near the bottom of the test tube (similar to KROTOS and TROI triggering strategy) – later triggering at 0.9 s. In the calculations approximately 25% of the melt was in the water at the early triggering time, and all the melt was in the water at the later triggering time. The calculation time was 2 s for the premixing phase and 10 ms for the explosion phase.

Firstly, the parametric analysis was carried out by scanning both, the premixing and the explosion phase with varying the oxidizing Zr content from 0% to 100%. Then the hydrogen film hypothesis was tested by choosing the threshold amount of Zr at 10%.

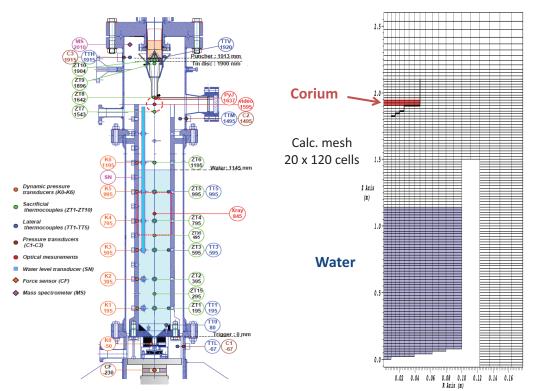


Figure 4. The KROTOS test tube (left) and the applied MC3D calculation mesh (right).

Table I. Experimental conditions in KS-5 test as applied in all calculations

	Property	Value
Melt properties	Delivered melt mass	2.428 kg
	Initial melt temperature	2860 K
	Liquidus temperature	2800 K
	Overheating	60 K
	Pressure	2.1 bar
Test section	Water temperature	327 K
	Initial subcooling	67 K
	Water depth	1.145 m
	Calculation release height	1.900 m
	Conical nozzle	1.820 m

## 3.2. Oxidation Influence in Premixing Phase

The oxidation influence in the premixing phase was examined by varying the oxidizing Zr content from 0% to 100% in the premixing calculation. The oxidation in the explosion calculation was set to 0%. In Fig. 5 the basic premixing integral results are summarized for all calculation cases at the early and later triggering times. Each diagram includes the total liquid droplet mass (red markers), the active droplet mass (violet markers) and the void fraction under the water level (light blue markers). The active droplets are defined as liquid droplets in regions with less than 60% of void and present the droplets which may efficiently participate in the explosion. The active droplets thus represent the premixing anticipation of the

explosion strength. The points in the diagrams are connected so that the development of the presented quantities may be followed more easily. It may be seen that in both triggering time series the void fraction is increasing with increased oxidizing Zr content, and that the active droplet mass is typically decreasing. An initial distinction may be observed at later triggering (right diagram) between the 0% Zr and 1% Zr case, where the total liquid and active droplet mass significantly increase. It seems that already a small amount of oxidation (compared to no oxidation) importantly reduces the solidification due to the increased droplets energy. For larger Zr content the total liquid droplet mass is approximately steady and the inhibiting influence of the increased void fraction prevails. For later triggering the dynamics of the void build up and premixture venting is quite turbulent, thus the active mass is fluctuating in accordance with the actual phases distribution at triggering time.

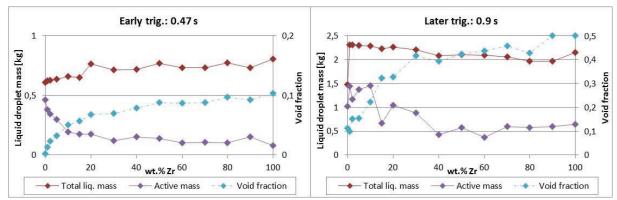


Figure 5. Liquid and active droplet mass plotted together with void fraction (right vertical axis) at early (left diagram) and later (right diagram) triggering times.

The explosion phase was calculated starting from the premixing results. The explosion strength is presented in Fig. 6 by summarizing the maximum pressures and maximum pressure impulses (time integral of pressure) at the pressure transducer locations. It may be observed that the pressure loads in general follow the active droplet mass development (violet curve in Fig. 5) for each triggering time series. After an eventual initial jump or plateau, the pressure loads are in general decreasing. For higher Zr content the pressure impulses are fluctuating due to the varying high void distributions and the increased melt thermal energy.

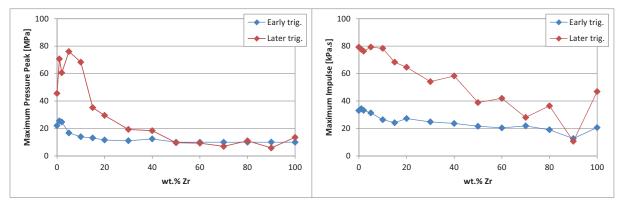


Figure 6. Trend of maximum pressures (left) and maximum impulses (right) with increasing oxidizing zirconium content. Blue markers depict the early triggering time (0.47 s) and red ones the later triggering time (0.9 s) cases.

## 3.3. Oxidation Influence in Explosion Phase

Here the oxidation in the premixing calculation was set to 0% and the oxidizing Zr content in the explosion phase was varied from 0% to 100%. Fig. 7 summarizes the maximum pressures and maximum pressure impulses reached in the simulations. The explosion simulations at larger Zr content stopped before the specified end time because the large released oxidation energy resulted in temperatures outside the range of the models. Therefore only those pressure and impulse values are depicted that could be deduced from the calculation results. It may be observed that the explosion strength is clearly increasing with the increase of the Zr content. The explosion strength increases due to the increased release of chemical energy and the increased production of hydrogen.

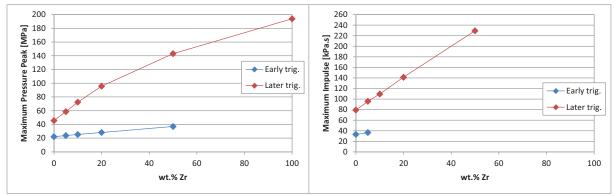


Figure 7. Trend of maximum pressures (left) and maximum impulses (right) with increasing the oxidizing zirconium content. Blue markers depict the early triggering time (0.47 s) and red ones the later triggering time (0.9 s) cases.

### 3.4. Oxidation Influence with Hydrogen Film Hypothesis

The hydrogen film hypothesis was checked by choosing the threshold amount of Zr at 10% in the premixing phase. The oxidizing Zr content was varied from 0% to 100%. In the premixing phase the oxidation is complete for the Zr content ranging from 0% to 10%. For higher Zr content the same fixed threshold amount of Zr is being oxidized during premixing (i.e. 10% of the melt mass) and the remaining Zr is oxidized during the explosion phase. Fig. 8 helps to visualize the oxidized amount of Zr in each phase as a function of the total oxidizing Zr content.

Fig. 9 summarizes the maximum pressures and maximum pressure impulses reached in the simulations plotted as a function of the oxidizing Zr content. Despite some calculations at larger Zr content stopped due to too high temperatures a clear trend is evident. Except the initial increase in the maximum pressure, the pressure loads are decreasing (maximum pressure) or being quite constant (maximum pressure impulse) until the chosen threshold Zr amount of 10%. After that the explosion strength is increasing with increasing the Zr content. The calculated behavior of the explosion strength is in reasonable agreement with the experimental findings in Fig. 1 and may potentially confirm the proposed hydrogen film hypothesis.

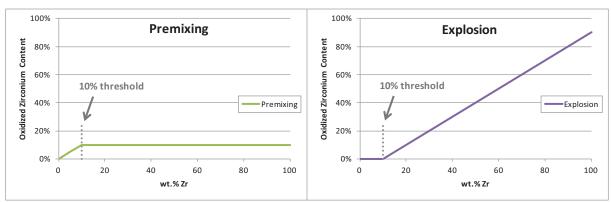


Figure 8. Chosen oxidized zirconium content (%) during the premixing (left) and explosion (right) phase as a function of the total oxidizing zirconium content.

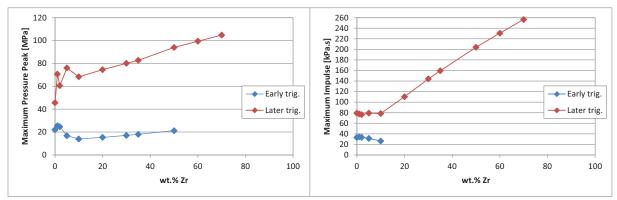


Figure 9. Trend of maximum pressures (left) and maximum impulses (right) with increasing the oxidizing zirconium content. Blue markers depict the early triggering time (0.47 s) and red ones the later triggering time (0.9 s) cases.

The initial increase in the maximum pressure, which was not observed in the experiments, indicates that some additional premixing processes may have to be taken into account. Indeed, the video camera recordings in the KS-5 and TS-5 experiments exhibit a distinctive turbulent reaction between the water and the melt jet immediately after the entrance into the water that was not observed in other experiments. It may be attributed to oxidation processes that contribute to faster jet fragmentation and potentially also to the formation of smaller droplets. Due to the earlier droplets generation more droplets are solidified before the triggering time, and potentially smaller droplets are quenched faster, which also increases the void production. All these processes decrease the explosion strength. If faster jet fragmentation and smaller melt droplets were properly considered in the simulations, it would likely compensate the effect of the increased droplet energy on the delayed droplet solidification for low Zr content and would thus probably eliminate the initial increase in the maximum pressure. The simulations would then probably exhibit a gradual reduction in the explosion strength also for very low Zr content, as indicated by the experiments. An additional explanation for the observed discrepancy at very low Zr content could be that in the experiments some oxidation of the melt occurs also without added Zr due to the stainless steel oxidation (ZREX) or due to the generation of over-stoichiometric melt (SERENA). Thus the calculations without any oxidation are not relevant and consequently as the starting point the calculations with at least some oxidizing Zr content should be considered.

#### 4. CONCLUSIONS

The influence of oxidation on the steam explosion energetics was analyzed by the comparison of SERENA and ZREX experimental results and by performing various computer simulations. In the ZREX experiments the steam explosion strength was largely increased by the oxidation of Zr, whereas in the recently performed SERENA experiments it seems that the oxidation had an inhibiting effect (the results are not conclusive). To explain this qualitatively different behavior the hydrogen film hypothesis is proposed, which states that the vapor film around the melt droplets gets filled with the produced hydrogen that reduces or even stops further oxidation. It implies that only a limiting amount of Zr may be oxidized during the premixing phase in sub-cooled conditions and that the remaining non-oxidized Zr is available for the oxidation in the explosion phase. This hypothesis may reasonably well explain the observed differences of the influence of the Zr content on the explosion strength in the ZREX and SERENA oxidation tests.

To get a better insight in the effect of oxidation and to check the proposed hydrogen film hypothesis a parametric oxidation influence study was performed with the MC3D code. The simulation results showed that the oxidation during the premixing in general reduces the explosion strength and that the oxidation during the explosion increases it. The results applying the hydrogen film hypothesis are in reasonable agreement with the experimental observations. The explosion strength decreases or is quite constant till the limiting amount of Zr, which may be oxidized during the premixing phase, and then it increases with increased Zr content. The only discrepancy occurs for very low Zr content, where in the calculations the explosion strength may initially first increase. This is attributed to the experimentally observed early oxidation processes during the premixing that promote the jet fragmentation and potentially result also in the formation of smaller droplets. These early oxidation processes, which reduce the explosion strength, were not considered in the simulations. An additional explanation for the observed discrepancy at very low Zr content could be that in the experiments some oxidation of the melt occurs also without added Zr. Thus the calculations without any oxidation are not relevant and consequently as the starting point the calculations with at least some oxidizing Zr content should be considered.

Together with the analytical work the need for further experimental investigations arises. It would be beneficial to carry out SERENA like premixing tests with prototypic oxidic corium with added metallic Zr, focusing especially on the measurement of produced hydrogen and the oxidation extent of the melt droplets, and the jet fragmentation and the droplet size distribution dependency on the Zr content. Further explosion experiments would also be needed within the lower Zr percentage range of 0% to 30%, where there is lack of experimental data.

### **ACKNOWLEDGMENTS**

The authors acknowledge the financial support of the Slovenian Research Agency within the research program P2-0026 and the cooperative CEA-JSI research project (contract number 1000-13-0106).

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