Analysis of oxidation influence on steam explosion energetics

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ABSTRACT

When during a severe reactor accident the corium melt comes into contact with the coolant water a steam explosion may occur. If non-oxidized zirconium is present in the corium melt the oxidation of zirconium may significantly affect the steam explosion energetics, as observed in experiments. In the ZREX experiments the steam explosion strength was largely increased by the oxidation of 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 article, 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 sub-cooled water 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 qualitatively explains the observed experimental findings.

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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 (Tanabe, 2012). 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 (Corradini et al., 1988; Berthoud, 2000). 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 (Theoufanous, 1995; Leskovar and Uršič, 2009).

The steam explosion process is commonly divided into four phases, i.e. premixing, triggering, propagation and expansion (Corradini et al., 1988; Berthoud, 2000). In the premixing phase the molten jet breaks up and a coarsely mixed region of molten corium and coolant water is formed. A vapor film separates the

melt and water, so the heat transfer between the melt and water is relatively low. The system may remain in this meta-stable state till the melt is quenched or, if the conditions are appropriate, till a steam explosion is triggered. The timescale of the premixing process is in the range of seconds, and the length scale of the melt particles is in the range of millimeters. In the triggering phase the steam explosion is triggered. The triggering event is a disturbance, which destabilizes the vapor film around a melt particle allowing liquid–liquid contact, leading to locally enhanced heat transfer, pressurization and local fine fragmentation. Many reasons can cause vapor film destabilization, including pressure pulses resulting from different impacts (in experiments interactions are often triggered when the melt reaches the bottom of the test vessel), transition from film boiling to nucleate boiling, and water entrapment at melt-structure contact. During the propagation phase, an escalation process takes place resulting from the coupling between pressure wave propagation, fine fragmentation, and heat transfer after the triggering event. The fine fragmentation process rapidly increases the melt surface area, vaporizing more coolant and increasing the local vapor pressure. The pressure wave spatially propagates throughout the melt–coolant mixture, fine fragmenting the melt and causing the whole region to become pressurized by the coolant vapor. During the propagation phase the thermal energy of the melt is converted into the thermal energy of the coolant. The timescale of the explosion propagation process is in the

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range of milliseconds, and the length scale of the fine fragmented particles is in the range of 10 μm. During the expansion phase the thermal energy of the coolant is converted into mechanical energy. The expansion of the high-pressure mixture against the inertial constraints imposed by the surroundings determines the damage potential of the steam explosion. The damage might be caused through pressure shock waves at the beginning of the process and afterwards through the kinetic energy transmitted to the surrounding structures.

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. According to the results of the TMI-2 core examination, layers of mainly oxidic material (UO_2 and ZrO_2) and layers of mainly metallic material (steel, Zr) are the main expected forms of materials during accident progression in the lower plenum of the reactor vessel (Sehgal, 2012). As revealed by the OECD MASCA project experiments, the melt pool forming in the lower plenum of the reactor vessel may gradually stratify in three layers of different melt composition, i.e. a molten oxidic pool with a light metal layer on top and a heavy metal layer below (Seiler et al., 2007). Thus, the failure of the reactor vessel could result, depending on the failure location, in ex-vessel FCI with purely metallic melt, purely oxidic melt or with combination of both melts. Furthermore, the composition of the oxidic melt may not be stoichiometric. Steam explosion experiments have already revealed that there is an important influence of the material properties on FCI (Corradini, 1991; Huhtiniemi et al., 1999). One of the key phenomenon, which is recognized but not well understood, is the oxidation of the corium melt during FCI in case that the melt contains a metallic component that is chemically reactive with water (Meignen et al., 2014, 2005). The purpose of the article is to examine the influence of the metallic material oxidation on the steam explosion energetics.

For a reactive metal component such as zirconium, the potential chemical reaction energy could be about seven times greater than the stored thermal energy of the melt (Cho et al., 1998). Thus, if a significant fraction of the chemical energy could be released during an explosive interaction, the explosion energy might be strongly augmented by the chemical reaction. Zirconium oxidizes in water vapor according to the following reaction, during which heat is released and hydrogen is formed:

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2 + energy \ (\sim 6.5 \ MJ/kg \ Zr).$$
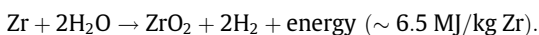
The influence of oxidation on the FCI is very complex. The released heat has a promoting effect on the steam explosion energetics due to the increased energy content of the melt and due to the delayed solidification. But at the same time it has also an inhibiting effect due to the increased void build-up. 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.

Oxidation during quenching of molten reactor core materials in water vapor was already a subject of past researches (Baker and Just, 1962; Baker, 1983). The experimental results with molten zircaloy in water have shown that the reaction strongly depends on the particle size and the water temperature. The reaction was limited to less than ∼20% of the melt if the mean particle diameter was above ∼0.8 mm and ∼2 mm for sub-cooled and near saturation conditions, respectively. The experimental results were well explained with a sphere quenching model that considered two main mechanisms of oxidation, i.e. solid-state and gas-phase diffusion. The gas-phase diffusion is important at the initial phase of oxidation whereas the solid-state diffusion dominates after the creation of the oxide layer. The presence of the oxidation layer decreases the oxidation rate resulting in faster quenching that finally limits the oxidation. The experimental results with molten stainless steel in water have shown that the reaction depends on the particle size and the water temperature. These results have also suggested that higher local concentration of hydrogen limits the oxidation.

The effect of oxidation on FCI was observed in several FCI experiments. In the FITS experiments conducted at the Sandia National Laboratories (SNL), USA, also the hydrogen production during interaction of melt (55 wt% Fe–45 wt% Al_2O_3) with water was measured (Marshall, 1988). The tests of the D series with benign interaction showed that for highly sub-cooled water the iron oxidation was in the range of 16–30%. In the D series tests with saturated or near saturated water the iron oxidation was in the range of 19–31% for near ambient pressure (non-explosive interaction) and in the range of 67–97% at elevated pressure (benign interaction). In the C series tests 4–8% or 18–33% of the available metal was oxidized in the benign interaction or explosive tests, respectively. The hydrogen production during FCI was investigated also at the Argonne National Laboratories (ANL), USA (Marshall, 1988; Spencer et al., 1994). In the non-explosive CCM tests 2.1–12.8 kg of corium melt (60 wt% UO_2–16 wt% ZrO_2–24 wt% SS) was poured into saturated or sub-cooled water. The metal oxidation was up to 35% for the saturated conditions. For sub-cooled conditions the oxidation rate was lower. In the CWTI experiments 1.3 kg and 2.4 kg of corium melt (60 wt% UO_2–16 wt% ZrO_2–24 wt% SS) was poured into water. High oxidation rate of 58% to 76% of iron was observed for saturated water conditions and 10% for sub-cooled conditions. In the non-explosive FARO L-11 test (JRC Ispra, EU), 151 kg of corium melt (76.7 wt% UO_2–19.2 wt% ZrO_2–4.1 wt% Zr) was poured into saturated water (Magallon and Hohmann, 1997). The results of this non-explosive experiment are suggesting that zirconium was probably completely oxidized at the end of the melt fall phase. To summarize, all experiments have demonstrated that the oxidation rate increases with the coolant temperature and that it is higher in explosion events.

Systematically the influence of the oxidation on FCI was investigated in the ZREX premixing and explosion tests (Cho et al., 1998) (ANL, USA). Recently it was investigated in the OECD SERENA project explosion tests (Hong et al., 2013), performed in the KROTOS (CEA, France) and TROI (KAERI, Korea) facilities. The main purpose of the ZREX tests was to explore 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 $(U,Zr)O_{2-x}$. In the ZREX experiments performed with simulant ZrO_2/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