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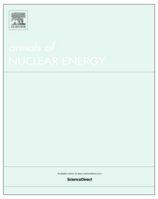

Experiments on silver-indium-cadmium control rod failure during severe nuclear accidents

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ABSTRACT

A large-scale bundle experiment with silver-indium-cadmium absorber rod in the QUENCH facility as well as a series of small-scale single absorber rod tests with prototypical materials have been conducted. A variety of parameters like temperature history, initial contact between cladding and guide tube and possibility of inner oxidation of the guide tube was investigated in the single-rod tests. Different failure modes from local melt-through to explosive failure were observed depending on the sample type. No interaction between Ag-In-Cd melt and stainless steel was seen up to 1750 K, near to the melting temperature of the steel. The cladding did not balloon before failure. Metallographic post-test analysis identified various phases formed during the high-temperature interactions between the involved materials. Furthermore, solidus and liquidus temperatures of the Ag-In-Cd alloy were determined accurately by DSC.

1. Introduction

Silver-indium-cadmium alloy with the composition 80 wt% Ag, 15 wt% In, and 5 wt% Cd is used as neutron absorber material in control rods (CR) of many Pressurized Water Reactors (PWR). It is the material with the lowest melting temperature (approx. 1100 K) among all metallic and ceramic materials applied in nuclear reactors, and hence will melt first during design basis and beyond design basis nuclear accidents. In most reactors, the Ag-In-Cd alloy is encased in stainless steel (SS) cladding tubes surrounded by guide tubes made of Zircaloy. Depending on the reactor size 2500–5000 kg of Ag-In-Cd absorber alloy is used in a PWR.

The general failure mechanism of Ag-In-Cd control rods during high-temperature transients has been already described in the overview papers by Petti (1989), Hofmann (1999), Lewis et al. (2008), and De Luze et al. (2013). The absorber rod alloy is thermodynamically stable with its stainless steel cladding, even in the liquid state. All binary phase diagrams between the main stainless steel components (Fe, Cr, Ni) and Ag, In, and Cd show neither formation of compounds nor any eutectic interaction, and only very limited mutual solubility of the elements at higher temperatures (Massalski et al., 1990). However, the absorber rod guide tube made from Zircaloy chemically interacts with the stainless steel (SS) cladding of the absorber rod. A solid-state contact between stainless steel and Zircaloy by bending or ballooning (Lewis et al., 2008; De Luze et al., 2013) results in chemical interactions with the formation of liquid phases around 1400 K. According to the binary phase diagrams the lowest eutectic temperatures between Zr and Fe, Cr, and Ni are at 1201, 1109, and 1233 K, respectively (Massalski et al., 1990). After failure of the absorber rod cladding, the molten Ag-In-Cd alloy comes into contact with the Zircaloy guide tube and chemically dissolves it without formation of a solid interaction layer (Hofmann and Markiewicz, 1994). This molten Zr-Ag-In can attack and chemically dissolve stainless steel, Zircaloy cladding and even UO_2 of the adjacent fuel rods well below the melting points of Zircaloy and urania. The relocating Ag-In-Cd alloy is therefore able to propagate and accelerate the core-melt progression at rather low temperatures. Furthermore, silver, indium, and cadmium are among the main contributors to aerosol release in the reactor cooling system and may strongly influence nature and transport of fission products in the primary circuit and later on in the containment (Petti, 1989; Dubourg et al., 2010).

The bundle experiment QUENCH-13 with prototypical Ag-In-Cd control rod as well as a series of 11 single-rod tests with 10-cm long control rod segments were performed at Karlsruhe Institute of Technology (KIT) in order to improve the data base on Ag-In-Cd control rod degradation and aerosol release. The general results of the bundle experiments (Sepold et al., 2009) as well as the aerosol behavior (Lind et al., 2010) have been already described elsewhere. This paper concentrates on the degradation and failure mechanisms of Ag-In-Cd control rods as well as on the interaction

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between silver-indium-cadmium absorber melt with other core components. Furthermore, an accurate melting temperature measurement by DSC of the Ag-In-Cd alloy used in the experiments as well as in German PWRs is presented.

2. Experimental details

The following materials were used in the QUENCH-13 bundle tests as well as in the single-rod experiments for the construction of prototypical control rods: The silver-indium-cadmium rod with a diameter of 8.87 mm was provided by Advanced Nuclear Fuels GmbH with a composition of 80 wt% Ag, 15 wt% In, and 5 wt% Cd. The cladding tube was made of stainless steel DIN 1.4541/AISI 321 (X6CrNiTi18-10) with outer and inner diameters of 10.2 and 8.96 mm, respectively; and the guide tube was made of Zircaloy-4 (Sn: 1.3 wt%, Cr: 0.12 wt%, Fe: 0.23 wt%, Zr balance) with 13.8/12.4 mm external and internal diameter. Hence, there is only a little gap between Ag-In-Cd and stainless steel (0.045 mm) and a considerable one between stainless steel cladding and Zircaloy-4 guide tubes (1.1 mm).

The QUENCH facility and the special design of the QUENCH-13 bundle are described in detail in Sepold et al. (2009). The QUENCH test bundle consisted of 21 fuel rod simulators with a total length of approximately 2.5 m. The central fuel rod simulator was replaced by a prototypical PWR control rod (Fig. 1). The control rod was filled with helium to a prototypic pressure of 1.2 bar. Steam access into the gap between cladding and guide tube was allowed by several holes at bottom and top of the control rod simulator. This is prototypical for PWR control rods where the access of water and steam into this gap is foreseen. The surrounding 20 fuel rod simulators were electrically heated by tungsten heaters of 1 m length. The cladding material of these rod simulators was standard Zircaloy-4. Fuel was represented by the ZrO_2 pellets. The bundle was extensively instrumented with about 60 thermocouples distributed along 17 axial positions. A mass spectrometer and various aerosol measurements systems were installed at the off-gas pipe.

The single-rod experiments with segments of Ag-In-Cd control rods were conducted in the QUENCH-SR test rig with inductive heating of the specimens (Hofmann et al., 1997) as it is shown in Fig. 2. The test rig was coupled with a quadrupole mass spectrometer (Balzers GAM 300). Temperatures were controlled and measured by a two-color pyrometer and additionally measured by a thermocouple attached to the surface of the rod segment at the mid axial position. Additionally, the pressure in the quartz tube was recorded. Two video systems were installed to observe the failure mode and melt release.

10-cm long specimens with different designs regarding the initial contact between stainless steel and Zircaloy-4 tube (symmetric, asymmetric) as well as the possibility of inner Zircaloy-4 oxidation (with and w/o 4-mm holes in the Zircaloy-4 guide tube) were investigated, see Fig. 3. Inner oxidation of the Zircaloy-4 guide as well as the local contact between the two tubes are expected to be prototypical for PWR control rod assemblies with a length of >4 m and the gap filled with water. The rods were filled with air at atmospheric pressure. All tests were performed in an oxidizing steam-argon (50 g/h steam, 50 L/h Ar) atmosphere. The first five tests were conducted before the QUENCH13 bundle experiment in order to support the definition of the conduct of the bundle tests. The samples were pre-oxidized in the steam-argon mixture for 5000 s at 1250 K (SIC-01,-03,-04,-05) and 1423 K (SIC-02), respectively. The subsequent transient phase was run with a heating rate of 0.1 K/s until failure. The tests were finished by switching-off the inductive heating and the steam flow. The Zircaloy-4 guide tube collapsed at around 1470 K during test SIC-01 because of the consumption of the air in the gap between stainless steel cladding and Zircaloy-4 guide tube. Therefore, starting with test SIC-02 a hole was drilled into the closed guide tubes, large enough for allowing pressure equalization but small enough to avoid steam access to the inner Zircaloy surface. The QUENCH-13 bundle test was defined based on the results of this test series. Especially, the preconditioning temperature in the bundle test was set to a temperature below expected control rod failure which should take place during the transient phase.

A second series consisting of six tests was performed after QUENCH-13 retracing the temperature histories at the two bundle elevations 950 mm (4000 s pre-oxidation at 1253 K, SIC-06,-07,-08,-09) and 750 mm (1063 K, SIC-10) (Sepold et al., 2009). The heating rate during the transient phase in this series was slightly higher (0.13 K/s). The final test SIC-11 was conducted without Zircaloy-4 guide tube to observe ballooning behavior of the stainless steel cladding before its failure. Details on test conditions are summarized in Table 1. Fig. 5 reveals a typical test conduct with

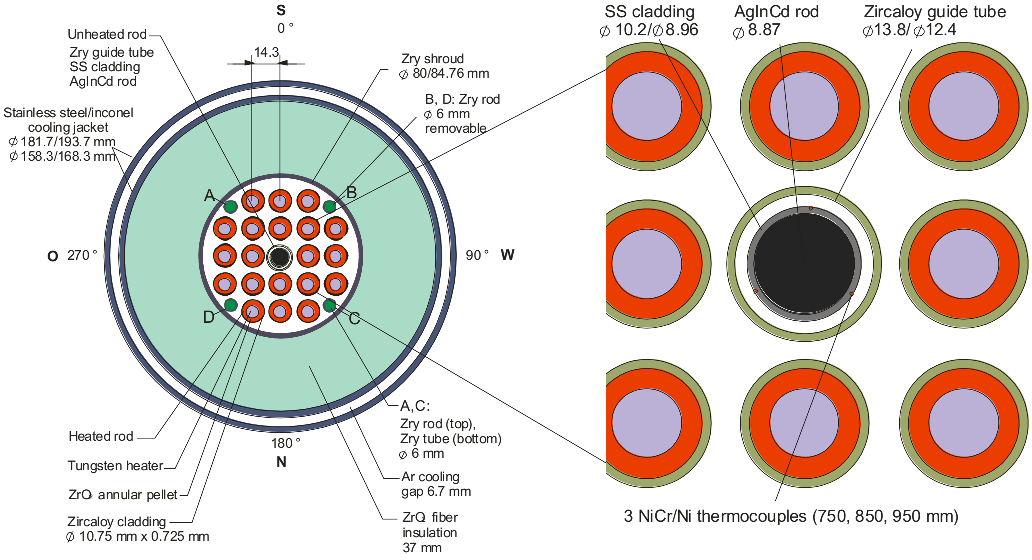

Fig. 1. QUENCH-13 fuel rod simulator bundle (cross section).