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DOUBLE COATED CLADDING FOR ADVANCED ACCIDENT TOLERANT FUEL - OVERWIEW

1. Introduction

With the increasing world's energy demand, the urgent need to reduce the use of fossil fuel, and shifting towards clean and sustainable energy sources, nuclear energy represents a promising solution. Many years of research and development generated a quite safe and reliable technology. the emphasis of advanced LWR fuel development was on improving nuclear fuel performance in terms of increased burnup for waste minimization, increased power density for power upgrades, and increased fuel reliability [1] however; Following the earthquake and tsunami in Japan in 2011, and the subsequent damage to the Fukushima Daiichi nuclear power plant complex, the accident highlighted some undesirable performance characteristics of the standard fuel system during severe events including accelerated hydrogen production under certain circumstances [1] that compromise the safety of nuclear power plants.. Thus, fuel system behavior under design-basis accident and severe-accident conditions became the primary focus for advanced fuels, and as a result; the post Fukushima research focus shifted towards the Accident tolerant Fuel (ATF)

[2,3,4,5]. When compared to conventional UO_2/zirconium-alloy fuel, ATF concepts are expected to provide enhanced performance, reliability, and safety characteristics during off-normal conditions while maintaining or improving performance during normal conditions. Using protective coating on the current in use zirconium alloys is considered as a near-term ATF that could provide the enhanced protection and safety while reducing the higher risk and costs associated with developing new fuel materials.

2. Cladding Materials Requirements

Clad tubes or claddings are a cylindrical tubes used to provide air-tight enclosure for the radioactive nuclear fuel such that the heat produced due to fission can safely be transferred to the coolant without contamination [6]. Clad tubes are vital part of reactors as they are not only provide an enclosure to the highly radioactive fuel but also remain in direct contact with the coolant during reactor operation which makes it vulnerable to corrosion. Its integrity becomes very important during accidental scenario when most of the heat emitted due to fission reaction accumulates inside the claddings [6]. There are multiple design constraints for the selection of the fuel cladding material. These constraints includes the neutron absorption cross section, the maximum service temperature, the creep resistance, the mechanical strength, the toughness, the neutron radiation resistance, the thermal expansion, the thermal conductivity and the chemical compatibility with fissile products and coolant, moderator and fuel materials. Additionally, the cladding material should present an acceptable service temperature to increase the thermal efficiency of the reactor, leading to a trade-off between service temperature and neutron transparency. Many materials combining Be, C, Mg, Zr, Si, and O (including metallic and ceramic systems) might be considered as potential new materials for nuclear fuel cladding in terms of neutron economy [7]. The fuel cladding material should also be radiation resistant and poses stable microstructure under irradiation. Finally, the cladding material must be corrosion/oxidation resistant to (coolant/moderator/fuel/fission products) and should present high thermal conductivity to increase the energy efficiency of the reactor; and low thermal expansion coefficient to minimize the thermal stresses in the cladding/pellet interface [8].

3. Zirconium Alloys

Zirconium alloys as fuel cladding materials for light water reactors play an important role in the nuclear industry due to an optimized set of corrosion, mechanical properties and a low thermal neutron absorption cross-section. The current in use Zr alloys differ considerably by radiation growth, radiation creep, corrosion resistance, high-temperature strength, hydrogenation and other characteristics that guarantee reliability. Despite the high number of studies around the world on zirconium alloys, including that containing Niobium, it is recognized that only five alloys are well studied and prepared for operation in PWRs, namely American Zircaloy-4 and ZIRLO, Russian E110 and E635 and French M5 [9]. The characteristic feature of zirconium alloys of Russian production is the presence of Niobium. Niobium is the main alloying element for binary and for multi components alloys. Base zirconium alloys of Zircaloy-2 and Zircaloy-4 are alloyed by tin, iron, chromium and nickel. The most crucial conditions for the current fuel claddings can occur in the case of a loss of coolant accident (LOCA). LOCA events may arise from a breakup in the primary cooling system that results pressure loss in nuclear core and vaporization of the coolant. Under these conditions, the fuel temperature rises, and the cladding temperature also increases abruptly. When Zr-based claddings interact with water steam at a high temperature, it causes oxidation and embrittlement by releasing additional heat due to the exothermic reaction [10]:

$$Zr + 2H_2O = ZrO_2 + 2H_2\uparrow,\ \Delta H = -584.5 \text{ kJ/mol at } 1200\ ^\circ C$$

4. The Fukushima Accident

The accident at Fukushima Daiichi Nuclear Power Station in Japan on March 11, 2011, has put safety concerns front and center of the ever-contentious debate about nuclear energy. Following the loss of external and internal power by the Tsunami, the reactors overheated and the fuel melted, highly flammable hydrogen was generated (mostly by a reaction between steam and zirconium “cladding” that surrounds the fuel), It built up in the reactor buildings of units 1 and 3 of power plant, before eventually exploding. Hydrogen may also have caused an explosion in unit 4 after it migrated there from unit 3 along their common venting system. With large quantities of radioactivity released into the environment, over three hundred thousand residents evacuated from the vicinity of the plants, and a cleanup operation that will take decades and cost tens, if not hundreds of billions of dollars, critics have argued that nuclear power is too dangerous to be acceptable. But are they right? Can nuclear power be made significantly safer? The answer depends in no small part on whether nuclear power plants are inherently susceptible to uncommon but extreme external events or whether it is possible to predict such hazards and defend against them.

Fig. 1. Using fire engines to remove the decay heat at Fukushima Diiachi

5. Accident Tolerant Fuel Programs

As a response to the design flaws exposed by Fukushima Nuclear Accident in 2011, a global shift has been occurred in nuclear fuel research, towards a fuel with specific ability to tolerate accidents. The concept of accident-tolerant fuel (ATF) has emerged as a unique fuel design expected to improve the safety of nuclear reactors. The underlying principle behind the ATF concept is to enable a reactor core to remain intact for a rather long time without the need for swift operator interventions in cases of severe events, when the external cooling supply is lost. Among the core tasks of ATF, is the design of cladding materials with outstanding performance. Although ATF is being developed primarily to give an advantage in high temperature oxidation scenarios that occur following an accident, there are a general set of requirements that are placed on nuclear fuel cladding. These requirements are that the cladding retains shape, retain all pellets and fission products, in addition to effectively transfer heat to coolant. In the framework of this concept, one of the economically advantageous and feasible in a relatively short time (<10 years) is the deposition of oxidation-resistant coatings on the existing zirconium alloys.

Several organizations have initiated studies on coatings on the Zr-based alloys over the last five years with the goal of enhanced accident tolerance. The coatings studied thus far broadly fall within two categories [11]:

• Metallic coatings:

- Pure Cr (AREVA/CEA/EDF, the Korea Atomic Energy Research Institute [KAERI], and University of Illinois Urbana-Champaign [UIUC]);

- Cr alloys: Cr-Al binary alloy (KAERI and UIUC);

- FeCrAl and Cr/FeCrAl multi-layer (KAERI and UIUC). For FeCrAl or iron-based alloys, a barrier layer is needed at the coating/substrate interface to prevent the formation of Zr-Fe eutectic at around 900°C. In the KAERI concept, a barrier layer of Cr or Cr-Al alloy is considered

• Ceramic coatings:

- Nitrides: CrN, TiN, TiAlN, CrAlN or multi-layers of different nitrides (IFE/Halden, The Pennsylvania State University [PSU]);
- MAX phases: Ti_2AlC, Cr_2AlC, Zr_2AlC, Zr_2SiC (KIT, AREVA).

Nitride ceramic coatings are used to harden materials and improve their wear behavior, especially TiN and TiAlN. Additionally, CrN is also used for corrosion protection. The Mn+1AXn (MAX) phases, where M is an early transition metal, A is a group 13 – 16 element and X is C and/or N, represent a family of layered ternary carbides and nitrides, which have attracted a great deal of attention in recent decades because of their unique combination of metallic and ceramic properties. Additionally, surface treatments are also investigated in Korea at KAERI as a complement to coating deposition, with the objective of increasing the high-temperature strength of the zirconium substrate by the formation of oxide dispersion strengthened (ODS) surface layer containing Y_2O_3 nano-particles. The KAERI concept is therefore more complex than other concepts (which feature only one external coating layer) because of the adoption of two surface treatments: an ODS surface layer in the zirconium substrate for High Temperature (HT) strength and the deposition of a second coating on top for the HT steam oxidation behavior [11].

6. Cladding Inner Side Protection

The same attribute that makes surface coatings on Zr-based alloys the most viable near-term ATF cladding technology presents their biggest performance challenge: the ~25-40 tones of Zr metal remains in the LWR core. For a LOCA, even a design basis LOCA, rod ballooning and burst occurs at temperatures as low as 700°C [12, 13, 14].

A realistic scenario that steam from the outer environment enters the cladding tube, through a rupture after ballooning and bursts, under loss of coolant accident (LOCA) conditions, or through cracks arise as a consequence to stress corrosion cracking (SCC) phenomenon, led by the projected stresses on the deformed fuel and the presence of accumulated fission products (FPs) in the pellet-clad interface (PCI). This exposes at least some part of the cladding's internal surface to the oxidizing environment, even though the outer surface may be protected by the coating. A recent and ongoing effort aims to tackle this issue by adding an inner surface coating [15, 16]. In fact, this concept is not new, internal surface coatings were proposed in the 1970s as a solution for the problem of SCC in power reactors. A graphite film successfully applied in CANDU reactor claddings due to its relatively short length (about 0.5 m), however; obstacles such as the practicality of applying thin films on the internal wall of a 4.5 m long tube has hindered the wide implementation of this solution [17]. Since the emergence of ATF and the acompined researches on coatings on zirconuim alloys, the problem of the potential inner uncoated side degradation have been stressed, leading to the emerging efforts at the level of deposition technologies to overcome this drawback.

7. Deposition Technologies

There is a continuous Efforts to homogeneously coat the inside of a fuel cladding tube , several Cr processes, including electro-deposition, Cold Spray, PVD-type, 3D-laser and vacuum-arc cladding have been already considered [16]. CEA collaborated with some French universities to develop a special metal–organic chemical vapor deposition (MOCVD) process. This method can provide a coating of a Zr alloy matrix at temperatures lower than the re-crystallization temperatures of the Zr alloy. A Cr-based coating is successfully deposited on the inner surface of a cladding tube. However, many grain boundary gaps, as well as impurities or meta-stable phases, formed in the polycrystalline Cr coating prepared by this method. the high-temperature oxidation resistance of the Cr-based coating failed to match that of the CrxCy amorphous coating prepared by the same MOCVD method because the amorphous material did not possess grain boundaries in the microstructure and with that, no channels for the rapid in-diffusion of oxidants through the coating [18].

DLI-MOCVD process is used to deposit CrCx in the inner side of fuel clad and its Scaled up for the internal treatment of a batch of 16 nuclear fuel cladding segments with a CrCx protective coating [19]. The working principle of DLI-MOCVD process can be considered relatively simple. A liquid solution containing the precursor diluted in toluene is injected into a flash vaporization chamber to generate a reactive vapor, transported by a carrier gas to the CVD reactor; monitored pulsed injection system divides the liquid solution into a cloud of small droplets, similar to a spraying system. It allows a more efficient vaporization step. This reactive vapor is thermally decomposed inside the reactor using a tubular oven [19]. However, more research to generate information about the protective nature of this CrCx is needed.

8. Conclusions

Zirconium has served the industry well over a 50-year time period and a remarkable evolution in fuel performance and reliability of such fuel cladding have been witnessed. Nuclear industry has optimized the current Zr-UO_2 which represents a large financial investment. Protective coatings represent a promising near term solution to overcome the Zr-UO_2 flows in severe accidents events, building up on the already existing operational experience. An inner-side coating will fill the gaps in terms of potential risks accompanies the use of protective coatings on the external surface of fuel cladding as ATF, and further enhance safety of NPPs.

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COMPARISON OF COOLED LIQUID METALLIC REACTORS (SODIUM AND LEAD)

A breeder reactor is a nuclear reactor that generates more fissionable material than it consumes. This unique reactor is meant to increase the amount of nuclear fuel available for electric power generation. A breeder reactor uses either uranium-238 or thorium, both of which are abundant. A conventional nuclear reactor can only use the readily fissionable but scarce isotope uranium-235 for fuel [1].

Types