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Advanced Cladding Materials for Accident Tolerant Fuels

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ABSTRACT

In terms of safety, nuclear fuel cladding is one of the main barriers in preventing the release of radioactive materials to the environment. The integrity of the fuel cladding is essential both in operation and in the long-term storage of spent nuclear fuel, not to mention accident conditions. In this paper, we review shortly relevant accident tolerant cladding materials from the Finnish point of view. Features compared to the traditional Zr-alloy cladding in the areas of oxidation, hydrogen pick-up, mechanical behaviour and fuel performance modelling are considered.

1 INTRODUCTION

The traditional UO_2-zirconium alloy fuel system has a well-proven safety and operational record that meet current safety requirements from the regulator. In addition, the performance of the fuel system, in current nuclear power plants (NPP) designs makes nuclear power a competitive clean energy alternative.

Projects related to accident tolerant fuels (ATF) and their development aim at improved operational safety of nuclear power plants. The recent OECD/NEA report on ATF fuels introduces metrics and evaluation approach for international use which are to be applied to the evaluation of new ATF concepts [1]. ATF designs should endure severe accident conditions longer than the traditional fuel and cladding systems, as well as offer the same or better fuel performance during normal operation [2]. However, as the material performance must be both evaluated and considered from the beginning of life through the final disposal of spent nuclear fuel, the design of new material concepts is slow.

This paper concentrates on some of the most promising and, from the Finnish point of view, interesting ATF cladding materials, based on the review in Ref. [1]. In addition, an overview of the general fuel performance, oxidation and hydrogen pick-up as well as mechanical behaviour of the cladding materials is provided and these topics are shortly reviewed in the following sections.

2 CANDITATE MATERIALS

ATF cladding includes numerous types of materials from metallic to ceramic. They all have very different neutron cross-sections and in-pile behaviour. The cladding of nuclear fuel has two major roles: (i) to confine fissile material, while maintaining a good neutron transparency and (ii) enable efficient thermal conductivity between the fuel and the coolant. Moreover, these include favourable corrosion and mechanical properties under both normal operation and accident conditions.

In accident performance analysis, to give measures for the problem and safety requirements, a design basis accident (DBA) is often applied. The DBA is defined as a postulated accident that a nuclear facility must be designed and built to withstand without loss to the systems, structures, and components necessary to ensure public health and safety. The regulatory criteria, which are designed with traditional fuel materials in mind, in a DBA scenario allow no incipient melting of the UO_2 fuel and the cladding temperature should not exceed 1200°C. A more general criterion is that the core should be able to accept emergency cooling.

2.1 Cr and Cr/Al coated zirconium alloys

When considering material properties under normal operating conditions, it is challenging to identify a cladding material that is superior to zirconium and its alloys. Therefore, it is reasonable to consider improving the Zr-alloy’s behaviour under accident conditions via coating the exposed cladding surface. The coating must be quite thin, less than 20 µm, to have minimal effect on the neutron absorption cross-section and fuel economics. The main advantage in coatings during operation conditions includes (i) reduced oxidation kinetics, especially for metallic coatings, (ii) reduced hydrogen pick-up fraction and (iii) increased wear resistance. Under normal operation conditions, the coated Zr-alloys have very similar mechanical behaviour, as compared to the uncoated claddings.

Under accident conditions, the coating can significantly reduce high-temperature steam oxidation and, therefore, heat and hydrogen production. The coating also improves post-quench ductility and reduces both creep and ballooning due to strengthening effects.

2.2 FeCrAl alloys

Iron-chromium-aluminium (FeCrAl) alloy clad fuel rods with UO_2 fuel provide an increased safety benefit during design-basis events and severe accident conditions. In addition, FeCrAl alloy cladding is fully compatible with both the current BWR and PWR coolant chemistries and shows excellent corrosion resistance [3]. However, there are two challenges for a FeCrAl cladding system. First, these alloys have a higher neutron cross-section compared to zirconium and its alloys. Secondly, it may introduce an increased tritium release into the reactor coolant.

A lack of available experimental data also applies to FeCrAl cladding. There needs to be more data to provide an increased understanding of the irradiated alloy properties and its in-pile behaviour.

2.3 42HNM

42HNM (in Cyrillic 42XHM) is a Ni-base alloy with Cr and Mo as primary alloying elements (41-43 wt.% Cr and 1-1.5 wt.% Mo). This alloy was developed in the late 90's in the A.A. Bochvar Research Institute of Inorganic Materials (SSC RF-VNIINM) as a radiation and corrosion resistant Ni-Cr alloy to replace austenitic stainless steels used as a cladding material for control rods [4, 5].

Its composition is very similar to alloy XHM-1, which is a chromium-enriched Ni-base alloy (44 wt.% Cr, 1–2 wt.% Mo) that has been suggested as a material for water-cooled fusion reactor components [5]. The biggest advantage of 42HNM, as a potential ATF cladding candidate, is its long history of application as a reactor internal material. Alloy 42HNM has been used as a cladding material for control rods in WWER-1000 type rectors, with a guaranteed operation time of 15 years. At the present time, this alloy is used as a reactor core structural material in nuclear icebreaker NPPs and in so called “special transport NPPs” with a typical service time of 30 years.

Alloy 42HNM outperforms many types of nuclear grade stainless steel as control rod cladding material in terms of mechanical properties; intergranular stress corrosion cracking (IGSCC) in chloride-containing environments and radiation effects, up to quite high doses (>30 dpa) [5]. Microstructural investigations performed on irradiated 42HNM have revealed that while irradiation induced dislocation loops are larger, the density of these loops is significantly smaller than in austenitic stainless steels irradiated up to the same dose. Based on this observation, it has been speculated that the formation of clear bands, and thus strain localization, which results in a loss of plasticity, does not occur to the same extent in 42HNM. Moreover, this presents good mechanical properties, in terms of plasticity and elongation after irradiation at 350°C, but at high temperatures above 550°C, these properties are significantly degraded [5].

Another disadvantage of this alloy is its high neuron capture cross-section. This neutron penalty would require compensation, though thinning of the fuel cladding's wall thickness and/or, possibly, enrichment of the fuel beyond the 5% limit. The necessity to compensate for the neutron penalty and for the more challenging handling of spent fuel rods, would impose significant increases in the associated costs. Alloy 42HNM represents only one of the several candidates in the Russian ATF development program, and presents both strong pros and cons. A more complete review of the Russian ATF program can be found in Refs [6, 7].

3 MODELLING AND MATERIAL BEHAVIOUR

3.1 Fuel performance overview

A standard evaluation of ATF performance consists of neutronics, thermal-hydraulics, fuel performance and a detailed system analysis. In addition, severe accident codes have been modified for ATF characteristics, but as there is little experimental data available at this point in time, the codes are mainly limited to qualitative estimates of these concepts. A large-scale analysis and code coupling is required to get the full picture of the reactor's safety and performance characteristics under accident conditions. For example, the set of codes employed in the USA for fuel and cladding concepts is described in Ref. [8].

In the last decade, efforts have been made to simulate accident scenarios, such as loss of coolant accidents (LOCA), with the fuel performance codes via coupling with the thermal-hydraulic and thermomechanical codes. For example, the fuel transient analysis code FRAPTRAN [9] has been coupled with TRACE [10] and GENFLO (a Finnish thermal-hydraulics code) [11] to simulate LOCA. FRAPTRAN itself is currently being developed for ATF modelling applications in several countries.

The concept-specific material properties in performance modelling have to be applied from validated models, experimental data and/or assumptions. In some cases, the behavioural models for ATF are similar enough to the UO_2 – Zr-alloy fuel to allow the same properties to be assumed, but in many cases material-specific models should be developed from scratch.

For ATF fuel performance modelling, two different approaches can be obtained. For homogeneous materials, like FeCrAl cladding, one can apply conventional fuel performance codes. Recently, the development and applications in the modelling of ATF cladding have been active with FRAPCON/FRAPTRAN, FUPAC [12], FALCON [13], and TRANSURANUS [14]. Candidates with more complex geometries, such as Cr and Cr/Al coated claddings, can be modelled with finite element (FEM) codes, such as BISON [15], COMSOL [16], ADINA [17], and ABAQUS [13], but models for traditional fuel performance codes have also been developed [29].

Finally, multi-scale modelling can play a significant role in the ATF design by reducing the need for in-pile testing and thereby accelerating the safety review process. For example, irradiation degradation has recently been evaluated with molecular dynamics and phase field methods for U-Mo fuel and SiC composite cladding [18, 19].

3.2 Oxidation and hydrogen pick-up

In traditional Zr-based nuclear fuel claddings one of the main lifetime limiting factors, in terms of thermal conductivity, is the thickness of the oxide layer. Over their lifetime, the fuel cladding is constantly exposed to a high temperature, high-pressure water environment, reactor type specific water chemistries and neutron irradiation. Exposure to this environment causes cladding oxidation.

One product of the oxidation reaction is hydrogen, and the cladding oxidation rate directly affects the hydrogen generation rate. Some hydrogen is then absorbed and diffuses through the oxide scale into the cladding bulk, while the remaining hydrogen is released as H_2-gas into the primary water. The hydrogen pick-up fraction is dependent on the material, microstructure, oxide layer thickness, water chemistry, temperature, and stress. Moreover, the hydrogen solubility in the matrix increases with temperature [20]. When hydrogen pick-up during oxidation is combined with stress, it leads to the formation of brittle hydrides, which reduce the cladding's ductility. As the oxide layer grows, the hydrogen content increases and thus hydride morphology and distribution evolves. This leads to local changes in the cladding's mechanical behaviour. These local changes, resulting from oxidation, are of particular concern, especially during accident conditions (e.g. LOCA, where cladding temperatures may rise above 900°C), see the next section for additional information on mechanical property considerations.

In an effort to improve oxidation behaviour, several candidate ATF cladding materials have been proposed [1, 21] , including, but not limited to, coated (Cr- or Cr/Al-) claddings and FeCrAl alloys, which have been previously discussed in this paper. ATF cladding concepts of Cr- and Cr/Al-coated Zr-alloys aim to improve oxidation, thanks to chromia-forming coating [22-25], corrosion resistance in nominal conditions and provide a significant enhancement of the resistance of the material to oxidation in steam at high temperatures (up to 1300°C), with a drastic decrease of hydrogen pick-up and/or release and equivalent, if not enhanced, mechanical properties under accident conditions. One specific concern with coated claddings is the possibility of coating spallation during high temperature transients, thus exposing the unprotected underlying material to extremely volatile oxidizing conditions. In addition, FeCrAl alloys present an improved resistance to corrosion [3]. Additional experimental data on the oxidation behaviour and hydrogen pick-up fraction in ATF cladding concepts is required to qualify these concepts and satisfy regulatory requirements.

3.3 Mechanical behaviour

Of the previously discussed ATF cladding candidate materials, oxide dispersion strengthened (ODS) FeCrAl steels exhibit excellent high temperature strength, creep resistance and improved corrosion resistance, as compared to Zr-alloys, which make FeCrAl claddings a safer option in a DBA scenario. Under the current DBA criteria, FeCrAl claddings should be able to respond better than the current zirconium alloy claddings. FeCrAl alloys melt in the vicinity of 1500°C, but FeCrAl materials are resistant to attack by steam to temperatures above 1200°C and up to the melting point.

Furthermore, the use of FeCrAl would eliminate or minimize the risk of ballooning and burst of cladding, experienced by some Zr-alloys at these temperatures. Temperature excursions may produce ballooning of Zr-alloy claddings, due to a pressure differential between the inner diameter and outer diameter of the cladding. The ballooning is caused by creep and oxidation that deteriorates locally the load bearing capability of the cladding wall. Burst processes are most prevalent in the temperature range 700-1000°C, where the contribution to bursting by oxidation of the cladding is significant for Zr-alloys, while for FeCrAl this contribution would be insignificant.

The drawback for ODS FeCrAl-alloys is that they can exhibit a strongly directional microstructure, leading to anisotropic mechanical properties, due to the fabrication process, especially when they are manufactured into thin-walled tubes. Another challenge originates from welding the ODS FeCrAl-alloys. Fusion welding causes agglomeration of fine oxide particles, which results in a loss of strength and creep properties of ODS joints. If these challenges related to the welding issues and the manufacturing process of ODS steels can be overcome, they form a promising group of alloys for nuclear fuel cladding applications in reactor concepts [26-28].

Regardless of the promising features in the coated ATF cladding concepts, open questions still remain. Although chromium is very ductile, cracking of the Cr-coating may occur at a certain level of strain during high temperature accidental conditions. The Cr-coating usually exhibits good adherence to zirconium substrate at normal operating conditions, but the maintaining of good adherence, i.e. avoiding delamination or spallation, at high temperature accidental conditions remains an open research question. Furthermore, results that validated the equivalent or better mechanical properties, such as strength and creep resistance under irradiation and in their irradiated state, as compared to traditional Zr-based claddings, remain scarce in open literature and call for further research.

4 CONCLUSIONS

Accident tolerant cladding material concepts Cr/Al coated cladding, FeCrAl alloy, and 42HNM show improved behaviour at higher temperatures compared to the traditional Zr-alloy. However, they have some drawbacks or additional limitations when applied in the operational conditions. This paper discusses features in fuel performance modelling, oxidation, hydrogen pick-up and mechanical behaviour that should be addressed when these materials are considered.

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