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Current status of materials development of nuclear fuel cladding tubes for light water reactors

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ABSTRACT

Zirconium-based (Zr-based) alloys have been widely used as materials for the key components in light water reactors (LWRs), such as fuel claddings which suffer from waterside corrosion, hydrogen uptakes and strength loss at elevated temperature, especially during accident scenarios like the lost-of-coolant accident (LOCA). For the purpose of providing a safer, nuclear leakage resistant and economically viable LWRs, three general approaches have been proposed so far to develop the accident tolerant fuel (ATF) claddings: optimization of metallurgical composition and processing of Zr-based alloys, coatings on existing Zr-based alloys and replacement of current Zr-based alloys. In this manuscript, an attempt has been made to systematically present the historic development of Zr-based cladding, including the impacts of alloying elements on the material properties. Subsequently, the research investigations on coating layer on the surface of Zr-based claddings, mainly referring coating materials and fabrication methods, have been broadly reviewed. The last section of this review provides the introduction to alternative materials (Non-Zr) to Zr-based alloys for LWRs, such as advanced steels, Mo-based, and SiC-based materials.

Contents

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1. Introduction

Generally, fission products are confined primarily by means of four successive physical barriers: the fuel matrix, the fuel cladding, the boundary of the reactor coolant system and the containment system, of which integrities are protected against internal and external hazards by the implementation of the defence in depth (DiD) concept (IAEA, 1997). As the second security barrier, the selection of materials and corresponding fabrication process for the cladding is of importance. Due to combinations with a low thermal neutron capture cross section, excellent corrosion resistance in high-temperature water and adequate mechanical properties, Zr-based alloys are widely used as materials for all commercial LWR fuel cladding. The motivations to achieve acceptable safety margin and higher burnup are driving the evolution in the reliability of Zr-based cladding, thereby contributing to the birth of advanced Zr-based alloys like E635, ZIRLO, M5, MDA, X5A and J-alloys, etc., which strode a major step forward in improvement of corrosion resistance and mechanical performances. However, the well-known inherent demerits of Zr-based alloys such as rapid oxidation and hydrogen production at high-temperature steam, cannot be altered by optimizing the Zr-alloy chemical composition and/or their manufactory processes, which was highlighted further by the nuclear accident happened at the Fukushima Daiichi plants. As a result, the implementation of DiD concept has been reinforced and the term ATF has become popular (NEA/OECD, 2016). Comparison with the current UO_2/Zr system, the enhanced ATF, described by US Department of Energy (US DoE), can tolerate loss of active cooling in the core for a considerably longer time period while maintaining or improving the fuel performance during normal operations (Goldner, 2012). Therefore, the ATF cladding concept is inevitably a hot topic against the background of the deployment of the ATF rods, resulting in innovative designs, such as Zr alloys with coating, advanced steels, and ceramic cladding. Coating technology has been attempted to enhance the waterside corrosion resistance and wear resistance without modifying the existing Zr-based alloys, which enable this coated cladding to be put into commercial application in a short term; additionally, the alternative materials (Non-Zr) have been under development to substitute existing Zr-based claddings. Whereas, it is worth noting that the long time together with the high cost is needed to achieve a reasonable return due to the involvement of modifications on fuel enrichment, size of assemblies associated with the corresponding alternative materials.

In this paper, an introduction to over past 50-year evolution in Zr-based cladding for LWR (primarily BWR, PWR and SCPWR) fuel is presented, in which the impacts of alloying elements on the performances of Zr-based alloys are briefly described. Then, the up-to-date ATF claddings, coating technology and alternative materials, have been summarized successively in this review.

2. Development of Zr-based alloys

2.1. The characteristics of zirconium

Cladding tubes are the vital part of a nuclear reactor because they 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 (Alam et al., 2011). Hence, the material used for cladding tubes must have the important characteristics as follows; low thermal neutron capture cross-sections, high thermal conductivity, high strength, and high corrosion resistance.

From considerations of neutron economy, the suitability of materials for water-cooled thermal power reactors is restricted

Table 1
Calculated effective neutron absorption cross section (neutron absorption cross section per unit of yield strength) for pure elements in comparison to Zr (Azevedo, 2011).

Elements	Neutron absorption cross section (Barns)	Yield strength (MPa)	Relative effective neutron absorption cross section in relation to Zr
Be	0.009	200–350	0.04
C	0.004	24–28	0.2
Mg	0.063	65–100	1
Si	0.16	165–180	1
Zr	0.185	135–310	1
Al	0.231	30–40	8
Mo	2.48	170–350	10
Cr	3.05	185–280	15
Nb	1.15	75–95	15
Fe	2.55	110–165	20
Ni	4.43	80–280	30
V	5.04	125–180	40
Sn	0.63	7–15	70

to metals like aluminum (Al), magnesium (Mg), and Zr or their alloys (see Table 1 (Azevedo, 2011)). However, severe blistering and accelerated corrosion precluded application of aluminum for power reactors at the temperature above 150 °C. Beryllium (Be), manganese (Mn) and their alloys also were not considered for cladding due to their failure to meet the mechanical properties and corrosion resistance at water-cooled reactor operating temperature (200–300 °C) (IAEA, 1987). At normal operating reactor temperature (300 °C), Zr is an exceptional substance and has been employed as fuel cladding tubes since the early 1950's because it has much lower neutron absorption cross section as well as adequate mechanical properties than the other commercially available structural materials (Baczynski, 2014). It was initially thought that the poor corrosion resistance of the unalloyed zirconium produced by the van Arkel process was a result of stray impurities, but it was found that improving the purity by Kroll process could not eliminate the problems. The oxidation rate of pure Zr decides the oxide grain orientation in the oxide films, which led to severe growth stresses and cracking of the oxide and oxide spalling (Cox, 2005). Therefore, to get additional properties for better operation in water cooled reactors, Zr was alloyed with constituents which have low nuclear impacts like tin (Sn), oxygen (O), and niobium (Nb), while other transition metals (iron (Fe), chromium (Cr), nickel (Ni), etc.) can be accepted up to limited concentrations (below 0.5 wt% total). It is worthwhile to discuss the effects of the alloying elements and impurities on the properties of Zr-based alloys.

2.2. Effects of alloying elements on Zr-based alloys

Pure Zr has two distinct type of crystal structure, hexagonal close-packed (HCP) in α-phase, and body-centered cubic (BCC) in β-phase and the α-to-β transformation temperature is 850 °C (Alam et al., 2011) (other different value, 865 °C, also was reported (Banerjee and Banerjee, 2016; Lemaignan, 2012)). Further, a two-phase (α + β) regime exists in the temperature range of 850–950 °C in Zr-based alloys. Zr-based alloys remain in α phase at the normal plant operation temperature (below 400 °C) (Azevedo, 2011). However, it is possible that phase transient conditions occur in the accident and deteriorates the accident continuously. The alloying elements could be divided into α stabilizer and β stabilizer. O and Sn, having high solubility in α phase and stabilize it at high temperature (Fig. 1), are able to raise the α-to-β transformation temperature, while β phase is stabilized by the addition of Fe, Cr, Ni and Nb (Fig. 2) (Lemaignan, 2012).

As an alloying element, O was added to Zr-based alloys to increase the yield strength by solution strengthening and its level in the range of 0.11–0.16% (all wt% in this review) is recommended