Scripta Materialia 141 (2017) 99–106

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

Viewpoint Set

# The development of cladding materials for the accident tolerant fuel system from the Materials Genome Initiative

Zhen Liu, Yifan Li, Diwei Shi, Yaolin Guo, Mian Li, Xiaobing Zhou, Qing Huang, Shiyu Du *

*Engineering Laboratory of Specialty Fibers and Nuclear Energy Materials, Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo, Zhejiang 315201, China*

ARTICLE INFO

*Article history:*
Received 30 January 2017
Received in revised form 26 July 2017
Accepted 26 July 2017
Available online xxxx

*Keywords:*
Materials Genome Initiative
ATF cladding
FeCrAl
SiC/SiC composites
Multiscale simulation

ABSTRACT

The 2011 Fukushima Daiichi disaster raises the requirement of accident tolerant fuel (ATF) cladding. As promising candidates, the FeCrAl ternary alloy and SiC fiber reinforced SiC matrix (SiC/SiC) composites are discussed with details for recent development, as well as the issues of their in-core application and fabrication. Herein, the design of materials based on the Materials Genome Initiative (MGI) is introduced as the effective scheme in accelerating the development for ATF claddings. Correspondingly, the potential technical routes are provided and some examples of preliminary modelling studies as the preparation steps for MGI are presented.


Nuclear energy plays an increasingly important role in modern industry. Safety is undoubtedly the topmost issue for the operation of any nuclear reactor. The Fukushima Daiichi disaster in 2011 [1] has been regarded as the alarm of the security for all reactors worldwide, which is caused by the failure in oxidation resistance of Zircaloy cladding materials during the loss of coolant accident (LOCA) scenarios. Therefore, there has been a strong incentive to develop a new set of light water reactor (LWR) fuel-cladding system which can reduce the probability of core degradation during accidents. The accident tolerant fuel (ATF) is considered as one of the most attractive concepts as the solution for improved safety, defined as the fuel-cladding system that can maintain its performance and functions for a considerable duration compared with the standard $UO_2$-Zircaloy system during accident scenarios [2–4]. The United States department of energy (USDOE) has supported the pioneer research and development of the ATF system since 2013 and set an enterprising schedule of irradiation test for lead rods in a commercial reactor by 2022 [5]. In fact, there are yet a large number of fundamental questions to be addressed.

To improve the accident tolerance, the development of ATFs would mainly follow these three potential schemes [6,7]: (a) reduce the cladding oxidation rate with high-temperature steam to suppress the oxidative heat release and hydrogen production; (b) improve the cladding thermo-mechanical properties to postpone or eliminate the cladding burst and failure; (c) design new fuel-cladding systems with lower operation temperatures. Considering that revolutionary designs of a fuel-cladding system are difficult to operate on current serving reactors, it is more urgent to develop ATF claddings compatible to current fuel pellets with enhanced high-temperature performance. Since the microstructural modification on existing zirconium claddings seems difficult to meet the demand of accident tolerance [6], new cladding materials has been considered as a promising option.

The potential candidate materials for ATF claddings include Fe-based alloys, such as Fe~18Cr~10Ni austenitic stainless steels (ASS), nickel-rich ASS (Fe~20Cr~20Ni), Fe-Cr and FeCrAl [8,9] ferritic stainless steels (FSS), as well as the ceramic materials SiC formed by nano-infiltrated transient eutectoid (NITE) [10] or Chemical vapor deposition (CVD) method and MAX phase materials [11] or coatings. A series of experiments have been performed to compare the oxidation resistance of these candidates by placing them in 1200 °C steam condition for 8 h [12–15]. Only Fe-25Cr, Fe-20Cr-5Al and CVD-SiC could form protective oxide layer, as shown in Fig. 1. From the thicknesses of oxide layers, the FeCrAl and CVD-SiC have much lower oxidation rate than the Fe-Cr sample. Furthermore, with consideration on the irradiation behavior, the composition of nickel needs to be controlled with caution because nickel has strong thermal-neutron absorbing ability and even produces radioactive cobalt via the $^{58}$Ni (n, p) $^{58}$Co reaction [16] and helium via the (n, $\alpha$) reactions of $^{58}$Ni and $^{59}$Ni isotopes [17]; high Cr composition would lead to serious problems on radiation induced hardening [18–20], meaning difficulty persists for the FeCrNi and Fe-Cr alloys to be suitable for nuclear application. Therefore, the FeCrAl ternary alloy and SiC ceramics are the two attractive candidates for ATF cladding materials.

In this paper, the recent reports on FeCrAl and SiC for ATF claddings are firstly reviewed and then the recently raised scheme for material design on the basis of Materials Genome Initiative (MGI) is introduced to

* Corresponding author.
*E-mail address:* dushiyu@nimte.ac.cn (S. Du).

http://dx.doi.org/10.1016/j.scriptamat.2017.07.030
1359-6462/

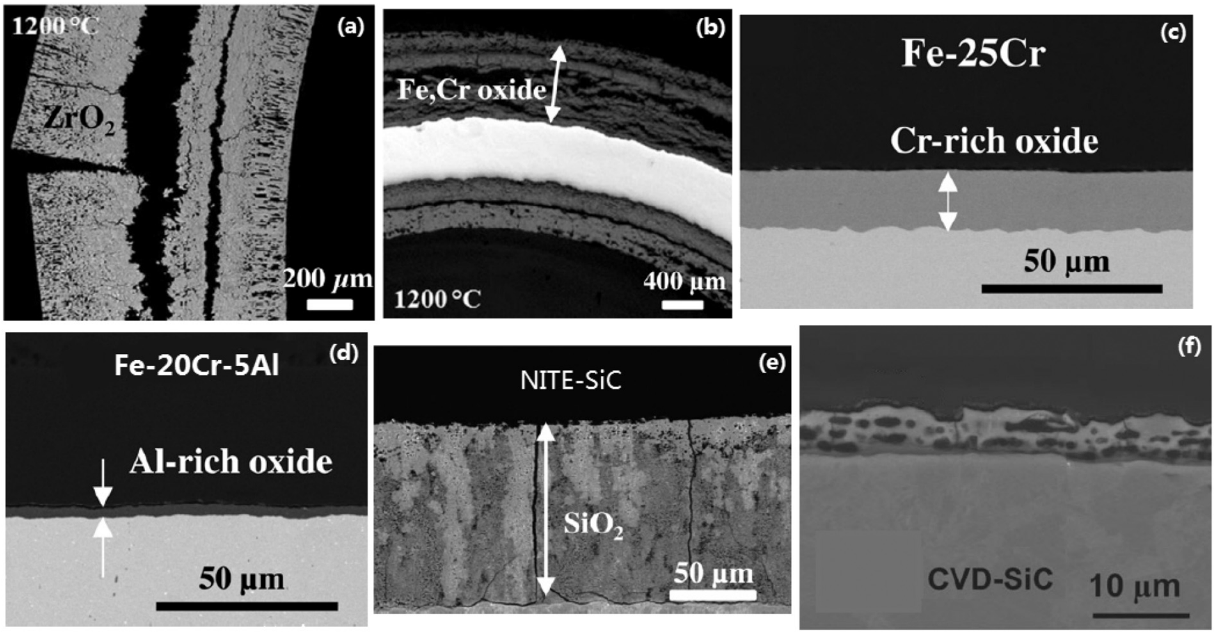


**Fig. 1.** Backscattered electron image of the oxide layer cross section micrographs after exposure in 1200 °C steam for 8 h: (a) Zircaloy-2 [14] (b) FeCrNi 317L [14] (c) Fe-25Cr [14] (d) Fe-19Cr-5Al PM2000 [14] (e) NITE-SiC [14] (f) CVD-SiC [16].

accelerate the development of these promising candidates. The content of this article is organized as follows. Section 2 reviews the recent progresses on the development of FeCrAl alloy and SiC fiber reinforced SiC matrix composites (SiC/SiC) composites as well as the currently unsolved issues for their LWR in-core application and fabrication. Section 3 mainly describes the suggestion on the protocol for MGI application on ATF cladding design as the major point of view for this paper as well as the general methodology of multiscale modelling for ATF claddings. In Section 4, some preliminary results of theoretical calculations on FeCrAl alloys and SiC/SiC composites are provided.

Several groups have been devoted to the development of the nuclear grade FeCrAl alloy for LWR ATF cladding application [21,22]. The production of FeCrAl alloys can be classified by generation I and II (Gen I & II). The design of Gen I FeCrAl alloys was focused on the optimization of composition including additive agents in order to satisfy the demands for nuclear utility, such as irradiation and oxidation resistance, weldability and fabricability. The in-situ tube burst tests [23,24] have proved that the Gen I FeCrAl cladding exhibits improved strength and oxidation resistance and demonstrated its 10% higher temperature and time to burst compared with the reference Zircaloy cladding. The Gen II ATF FeCrAl alloys are the screening result based on the Gen I candidates and targeted for further strengthening and oxidation resistance at elevated temperatures, as well as a good fabricability for potential commercialization.

It is believed that the higher chromium (Cr) composition would be better for corrosion resistance due to its support for the stability of the scale over a wide temperature range. However, the irradiation induced precipitates dominate the hardening response in neutron-irradiated Fe-Cr-Al alloys; the irradiation tests [18–20] have shown that the alloys with high Cr fraction is suffered from the radiation-induced embrittlement causing reduction in ductility and fracture toughness due to the formation of Cr-rich $\alpha'$ precipitates. For this reason, it is necessary to control the Cr composition to attenuate the irradiation hardening without loss in oxidation resistance. Further oxidation tests [25,26] indicate the oxidation resistance strongly depends on the composition of Cr and Al and higher Al content is needed when the Cr fraction decreases. However, the high Al may cause difficulties in welding and fabrication [27,28], which means caution on Al composition should also be exercised for alloying. Integrating the results from several experiments [29–31], the alloy of Fe-12~13Cr-4.5~5Al can form protective oxide layer and withstand a high temperature up to 1475 °C. Thus, it can be considered as the possible composition range for Gen II development, though further delicate sampling from various FeCrAl structures is still necessary to discover the optimal compositions. The raw FeCrAl alloys are well known for poor thermal mechanical strength at >600 °C, but the recent work has shown that this encumbrance to application can be solved by second phase strengthening and grain size control [30]. The minor additive elements of the alloy include Mo, Nb, Si, Zr, C, etc. and the second phase are designed to be thermally stable compound under heat treatment, such as $M_{23}C_6$ (M: mainly Cr), MC (M: mainly Nb), and Laves-$Fe_2M$ (M: mainly Mo and Nb) [21]. The oxide dispersion strengthened (ODS) FeCrAl alloys via $Y_2O_3$, $TiO_2$, $HfO_2$ and $ZrO_2$ nanoparticles are also of great interest for their capability of assistance in enhancing mechanical strength and radiation tolerance. Yamamoto et al. [31] has investigated the additive agents and procedures of thermal treatment for wrought FeCrAl to control the grain size aiming to obtain good mechanical properties. A Fe-13Cr-4.5Al alloy with Mo, Nb additives showed non-recrystalized fine grain microstructure after hot-rolling and annealing at 800 °C. Pint et al. [32–34] have developed the ODS FeCrAl alloys to balance the strength and oxidation tolerance with relatively low Cr component. Fe-12Cr-5Al alloys obtained by ball milling FeCrAl powder with $Y_2O_3$ and $ZrO_2$ additives shows both good thermal-tensile strength and oxidation resistance at 1200 °C but low ductility up to 800 °C. The enhanced intrinsic strength would allow the reduction of cladding thickness, down to ~300 μm for wrought FeCrAl alloy and ~250 μm for ODS FeCrAl, which decreases the neutron penalty caused by replacement of Zr-based alloys with Fe-based alloys [35,36]. The database for irradiation behaviors of Gen I &II FeCrAl candidates has been recently constructed [37] which may provide foundation for in-depth investigation towards the nuclear application of these materials.

In addition, the welding behaviors before and after irradiation are also studied as well as the fabricability [38–41]. As expected, the welded FeCrAl specimens present a 10–35% decrease of overall strength due to the microstructure degradation [39]. The post-irradiation examinations (PIE) [40] also show that the welded specimens exhibit more pronounced radiation-induced embrittlement than the non-welded ones. In this regard, more studies are required for understanding the