



Technical Inquiry 2018-3832

Developed by:
HDIAC
104 Union Valley Rd
Oak Ridge, TN 37830

HDIAC Contract Number:
FA8075-13-D-0001



HDIAC



Homeland Defense & Security
Information Analysis Center

Distribution A:
Approved for public
release. Distribution
unlimited.

This inquiry response is the result of four hours of research and analysis by HDIAC. This report is intended solely for informational purposes and is a cursory review and analysis of information available at the approved distribution level for each customer. This report is not to be construed as a comprehensive look at the topic in question. For more information on utilizing HDIAC for a more in-depth Core Analysis Task, visit www.hdiac.org.

Overview

A representative from the Y-12 National Security Complex requested information regarding protective coatings for nuclear fuel applications.

Findings

Within light water reactor environments, water radiolysis, gas stripping, and recirculation generate dangerously corrosive environments due to the many byproducts created through normal reactor operations (specifically hydrogen peroxide and hydroperoxyl) [1]. In addition to oxidation, the buildup of free hydrogen gas is of particular concern, especially after the Fukushima-Daiichi incident in 2011 where excess hydrogen ignited due to build up from a loss of coolant accident (LOCA).

Typically, nuclear fuel rods are wrapped in an exterior cladding of zirconium alloy, which forms an exterior layer of zirconium oxide (ZrO_2) [2]. Due to the radiation-heterogenic process that occurs at the alloy-coolant boundary, the ZrO_2 layer enhances the thermal and radiolysis decomposition processes of the coolant, leading to the generation of copious amounts of molecular hydrogen [2]. In the event of a LOCA, modeling shows that an exothermic reaction of zirconium-alloy cladding takes place in steam, particularly at temperatures of 1,200 degrees Celsius or higher [3]. Once exothermic oxidation begins, core destruction is nearly inevitable [3].

Coating and ATF Development

A researcher at Oak Ridge National Laboratory (ORNL) recently conducted a review of accident tolerant fuel (ATF) development efforts and the challenges associated with producing ATF cladding and coatings [4]. Report findings revealed that using conventional classes of protective oxides—namely chromia (Cr_2O_3), alumina (Al_2O_3), and silica (SiO_2)—protect against highly oxidative reactor environments [4].

As shown in Figure 1, Cr_2O_3 , Al_2O_3 , and SiO_2 exhibit significantly better resistance to high temperature oxidation than zirconium oxide (ZrO_2) [4]. As such, ORNL researchers posit that any ATF coating likely needs to contain, at a minimum, one of the elements Al, Cr, or Si [4]. The most promising coating technology for Zr alloy fuel rods are ones that form Cr_2O_3 , specifically Cr, CrAl, and CrN [4]. Likewise, Cr is the leading material for surface coatings of standard zirconium alloy claddings, as it provides a significant increase to corrosion protection across a wide range of temperatures without significant penalties from neutron absorption [3]. To date, most coatings forming Al_2O_3 or SiO_2 have been either FeCrAl or MAX-phase compounds such as Ti_2AlC , $TiAlN$, Ti_3SiC_2 , and Cr_2AlC [4].

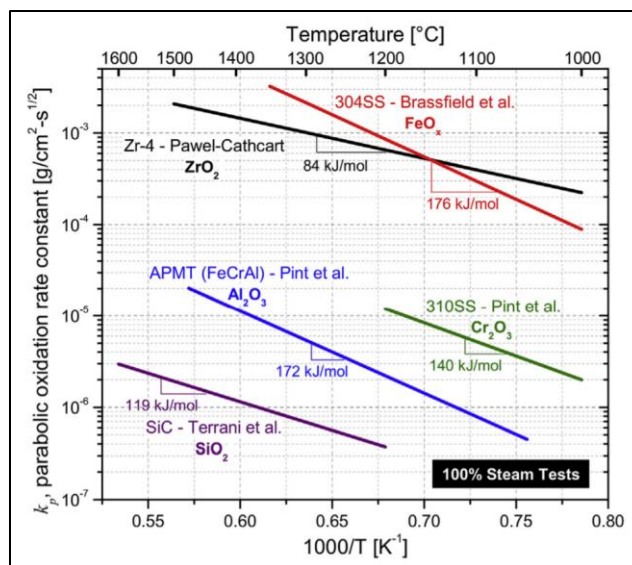


Figure 1: Parabolic oxidation rate for Cr_2O_3 , Al_2O_3 , and SiO_2 films produced from their parent materials typically used in ATF research [4].

Adding surface coatings to fuel cladding significantly increases fuel product safety and performance under normal operation and during LOCA [3]. Improving cladding performance by reducing the thickness of the oxide layer and hydriding of the cladding will increase economic efficiencies by increasing fuel available for burn-up [3].

FeCrAlY and FeCrAl-Mo

Westinghouse is investigating FeCrAlY as an alternative to chromium-based coatings [3]. In testing, FeCrAlY demonstrates considerable oxidation protective properties. However, iron forms a eutectic compound with zirconium at high temperatures [3]. In order to effectively implement FeCrAlY, these coatings would require an “interlayer of a diffusion barrier material [3].” Westinghouse and the University of Wisconsin are researching the development of these diffusion barrier materials [3].

Additionally, University of Wisconsin researchers are investigating the deposition and effectiveness of FeCrAl alloy and Mo onto Zr alloy using cold spray techniques [6]. Because the interaction of Fe with Zr at high temperatures forms a eutectic compound, researchers developed a Mo interlayer coating to facilitate a diffusion barrier between the two alloys [6]. Researchers demonstrate the successful development of a FeCrAl-Mo coating that improves high temperature oxidation resistance of fuel rods [6].

Ti₂AlC

The University of Wisconsin, together with Savannah River National Laboratory, is testing Ti₂AlC coatings for increased ATF [5]. Researchers demonstrate that the oxidation resistance of Ti₂AlC is significantly greater than that of uncoated zirconium alloy [5]. In addition to corrosion resistance, Ti₂AlC coatings add a high level of hardness and wear resistance to fuel rods, which can prevent damage during installation and normal operations [5].

Conclusion

HDIAC identified several promising solutions for the protection of nuclear fuel from corrosion and oxidation. Materials that contain or form the oxides Cr₂O₃, Al₂O₃, and SiO₂ exhibit significant resistance to high temperature oxidation. Namely, Cr, CrAl, CrN, Ti₂AlC, TiAlN, Ti₃SiC₂, Cr₂AlC, FeCrAlY, and FeCrAl-Mo facilitate high temperature oxidation resistance within the nuclear reactor environment and are the focus of many recent research projects.

We request your feedback on this Inquiry: <https://www.hdiac.org/new-inquiry-assessment-form/>

References

1. Chmielewski, A., & Szolucha, M. (2016). Radiation chemistry for modern nuclear energy development. *Radiation Physics and Chemistry*, 124. doi:10.1016/j.radphyschem.2016.01.002
2. Skotnicki, K., & Bobrowski, K. (2014). Molecular hydrogen formation during water radiolysis in the presence of zirconium dioxide. *Journal of Radioanalytical and Nuclear Chemistry*, 304. doi:10.1007/s10967-014-3856-9
3. Shah, H., Romero, J., Xu, P., Maier, B., Johnson, G., Walters, J., . . . Sridharan, K. (2017). Development of surface coatings for enhanced Accident Tolerant Fuel (ATF). In 2017 Water Reactor Fuel Performance Meeting. Retrieved from <https://www.researchgate.net/publication/320190475>
4. Terri, K. (2018). Accident tolerant fuel cladding development: Promise, status, and challenges. *Journal of Nuclear Materials*. doi:10.1016/j.jnucmat.2017.12.043
5. Maier, B., Garcia-Diaz, B., Hauch, B., Olson, L., Sindelar, R., & Sridharan, K. (2015). Cold spray deposition of Ti₂AlC coatings for improved nuclear fuel cladding. *Journal of Nuclear Materials*. doi:10.1016/j.jnucmat.2015.06.028
6. Yeom, H., Maier, B., Johnson, G., Dabney, T., & Walters, J. (2018). Development of cold spray process for oxidation-resistant FeCrAl and Mo diffusion barrier coatings on optimized ZIRLO™. *Journal of Nuclear Materials*. doi:10.1016/j.jnucmat.2018.05.014