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Original Article

AREVA NP's enhanced accident-tolerant fuel developments: Focus on Cr-coated M5 cladding

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A B S T R A C T

AREVA NP (Courbevoie, Paris, France) is actively developing several enhanced accident-tolerant fuels cladding concepts ranging from near-term evolutionary (Cr-coated zirconium alloy cladding) to long-term revolutionary (SiC/SiC composite cladding) solutions, relying on its worldwide teams and partnerships, with programs and irradiations planned both in Europe and the United States.

The most advanced and mature solution is a dense, adherent chromium coating on zirconium alloy cladding, which was initially developed along with the CEA and EDF in the French joint nuclear R&D program. The evaluation of the out-of-pile behavior of the Cr-coated cladding showed excellent results, suggesting enhanced reliability, enhanced operational flexibility, and improved economics in normal operating conditions. For example, because chromium is harder than zirconium, the Cr coating provides the cladding with a significantly improved wear resistance. Furthermore, Cr-coated samples exhibit extremely low corrosion kinetics in autoclave and prevents accelerated corrosion in harsh environments such as in water with 70 ppm Li leading to improved operational flexibility.

Finally, AREVA NP has fabricated a physical vapor deposition prototype machine to coat full-length cladding tubes. This machine will be used for the manufacturing of full-length lead test rods in commercial reactors by 2019.

1. Introduction

The Fukushima Daiichi accident triggered a worldwide research and development effort in the nuclear fuel industry to increase fuel margins in severe accidents, which has led to the development of enhanced accident-tolerant fuels (EATF) [1,2]. In this context, AREVA NP is actively developing several concepts ranging from short-term evolutionary (Cr-coated zirconium alloy cladding and Cr_2O_3-doped UO_2 fuel) to long-term revolutionary (SiC/SiC composite cladding) solutions, relying on its worldwide teams and partnerships, with programs and irradiations planned both in Europe and the United States. The AREVA NP EATF developments are fueled by three programs: the United States Department of Energy (DOE) accident-tolerant fuels (ATF) program, which is currently beginning phase 2, the French Joint Research program (with CEA and EDF), and the irradiation of EATF concepts in the Gösgen reactor. These programs enable AREVA NP to use its forces

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in all regions in a joint effort to accelerate the implementation of its concepts.

In the French Joint Research program with CEA and EDF, AREVA NP has been developing Cr-coated zirconium alloy and SiC/SiC composite cladding, and these solutions are currently being irradiated in the Gösgen reactor in Switzerland, thus linking those two programs in a broader European effort. The ongoing developments in Europe will support and feed the developments and implementation in the United States as part of the DOE ATF program, which will focus on the characterizations necessary for licensing and value assessment of the ATF concepts. The main goal in the DOE program is to insert AREVA NP's evolutionary fuel rod solution, consisting of Cr-coated M5 cladding and Cr_2O_3-doped UO_2 fuel, in a commercial reactor in the United States by 2019. As part of this effort, irradiation testing of this solution is planned to start in 2018 in representative Pressurized Water Reactor (PWR) conditions in the Advanced Test Reactor (ATR) in Idaho National Laboratory.

AREVA NP therefore focuses primarily on its near-term fuel rod solution consisting of Cr-coated M5 cladding combined with Cr_2O_3-doped UO_2 fuel but continues to investigate other potential cladding solutions that may exhibit even better performance under accident conditions but which currently have major challenges for implementation, such as SiC/SiC composite cladding. In this case, AREVA NP recognizes that the developments will take time and aims at resolving the main technical challenges in the upcoming years.

This article focuses on the development program of the Cr-coated M5 cladding, which is organized into several parts as described in Fig. 1: Cr coating manufacturing, material characterization, material irradiation, and fuel performance modeling. The first three parts are used for the fabrication and justification necessary to introduce lead test rods in commercial reactors, which is AREVA NP's near-term goal. This article therefore gives a general status update on these developments.

2. Cr-coated M5 cladding developments

The most advanced and mature cladding solution for AREVA NP is the Cr-coated M5 cladding, which consists of a 15-μm thick dense Cr coating layer deposited on the surface of an M5 cladding tube. In the same way, several other institutions worldwide are developing coatings, and especially Cr coatings, as a near-term EATF solution [3–7]. The main difference between the different ongoing international developments on coatings is the fabrication method and sometimes the coating material. For example, KAERI is developing Cr–Al alloy coatings using 3-D laser deposition [8,9]. This article focuses on the ongoing coating developments at AREVA NP.

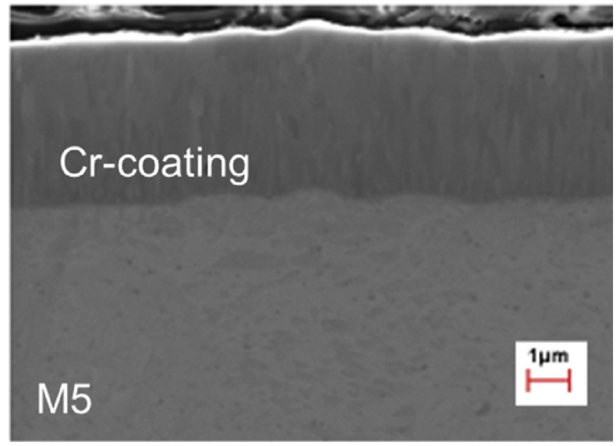

Fig. 2. Cross-section SEM micrograph of a Cr-coated M5 sample.
SEM, scanning electron microscope.

The AREVA NP coating is deposited using a physical vapor deposition (PVD) technique that does not modify the microstructure of the underlying zirconium substrate and forms a dense layer with no porosity at the Cr–Zr interface. The coating thus fabricated is very adherent and is therefore very protective as was shown in previous articles [10,11]. Fig. 2 shows a scanning electron microscope cross section of the Cr coating layer deposited on an M5 substrate, showing a dense and homogeneous Cr layer. This solution shows improved high temperature (HT) steam oxidation resistance and exhibits some benefits in HT creep performance, which makes it an ideal short-term EATF solution [12,13]. This section reviews the development updates concerning manufacturing, out-of-pile characterizations, and material irradiation projects.

2.1. Manufacturing

2.1.1. Full-length prototype fabrication

All the previous characterizations have demonstrated the benefit of Cr-coated cladding in both nominal and accidental

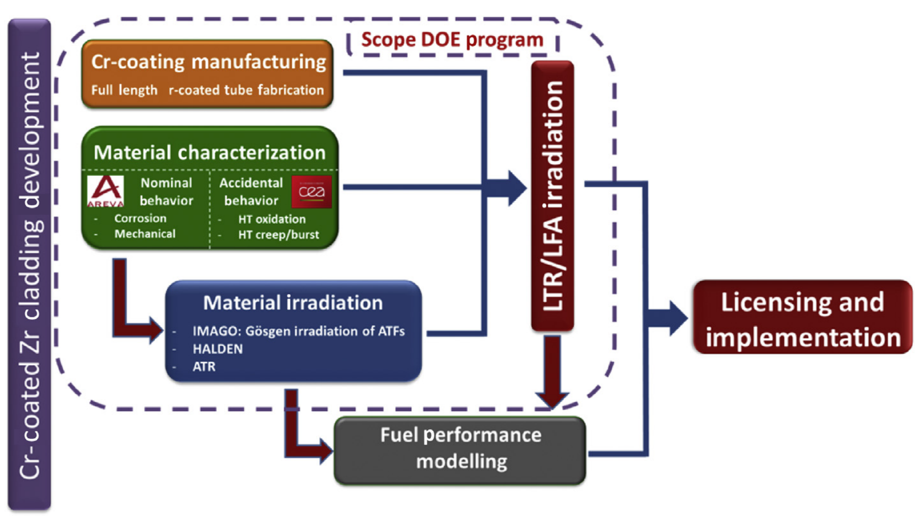

Fig. 1. Schematic representation of the Cr-coated zirconium alloy cladding development program.
ATF, accident-tolerant fuels; ATR, Advanced Test Reactor; DOE, Department of Energy; HT, high temperature. LFA, Lead Fuel Assembly; LTR, Lead Test Rod.

conditions [10–13], and therefore, AREVA NP decided to launch the industrialization of this ATF solution to be able to fabricate full-length cladding tubes for lead fuel rod (LFR) irradiations within the next couple of years.

To produce Cr-coated cladding samples, pure Cr is deposited on the surface of the samples using a special PVD process. This technique forms a very dense coating with neither cracks in the Cr layer nor porosity at the Cr–Zr interface, thus providing excellent adherence, as described in a previous article [14]. Consequently, in 2016, the design of a PVD reactor for full-length cladding tubes was performed, and the fabrication of this prototype machine was launched. The goal of the prototype is to demonstrate the feasibility of full-length Cr-coated tube fabrication using PVD and to use this facility to manufacture the first full-length Cr-coated cladding tubes for insertion in commercial reactors as LFRs.

The prototype machine was finalized in the summer of 2017 and is currently performing start-up optimization and production of the first full-length Cr-coated cladding tubes. This schedule is in line for the manufacturing of LFRs for irradiation in a commercial reactor by 2019. Fig. 3 shows the state of the prototype fabrication as of mid-April 2017.

2.1.2. Welding of Cr-coated M5 tube

A first feasibility study of the welding of Cr-coated tubes was performed last year using AREVA NP's "Upset-Shape Welding" (USW) process. This is the current resistance welding process used in AREVA's fuel manufacturing plants, and therefore, the goal was to evaluate whether the Cr coating impacted the fuel fabrication procedures. The results of the feasibility study were extremely positive, showing that no modification of the welding parameters was needed to adapt to the fabrication of the Cr-coated fuel rods. Consequently, at the end of 2016–beginning of 2017, qualification of the USW process for Cr-coated fuel rod fabrication was performed at the fuel assembly manufacturing plant of Romans. This qualification was successful with once again no change in the welding parameters. Fig. 4 shows the visual examination of the weld on a Cr-coated M5 tube with no flaws detected. The weld samples were also tested through ATSM G2 corrosion tests in 360°C water for 24 hours, and through burst tests, which revealed that the burst occurred outside the weld, thus certifying the good quality of the weld. These tests were standard tests for the weld process assessment and were performed on uncoated material as manufacturing tests in fuel fabrication facilities. The leak tightness of the fuel rod will therefore be guaranteed with the same level of quality as for current fuel rods. Consequently, these results show that the Cr coating does not modify the current welding process and therefore has no impact on the overall fuel rod manufacturing process.

Fig. 3. Picture of the current status of fabrication of the full-length Cr coating deposition prototype.

2.2. Out-of-pile characterizations

This section focuses on the experimental analyses performed at the AREVA NP facilities. The primary aim of these investigations is to evaluate the normal operating condition behavior of the Cr-coated cladding, while the CEA focuses on the HT conditions and accidental behavior, which will be presented in another article [15]. The previous results obtained at the AREVA NP Research and Technical Centers focused mainly on obtaining data necessary for irradiation justification to prepare for LFR justification [10,11]. The results presented in this article are intended to evaluate potential-added benefits from the use of Cr-coated cladding in normal operating conditions, including an investigation of the behavior of Cr-coated M5 beyond the Cr–Zr eutectic point.

2.2.1. Corrosion in 360°C water with 70 ppm of Li

Concerning the nominal behavior, Cr-coated zirconium alloys have shown excellent corrosion behavior in a static autoclave with representative PWR water chemistry ([B] = 650 ppm; [Li] = 2 ppm; no added dissolved O_2 or H_2) [10,11]. In a static autoclave, it is impossible to control the dissolved hydrogen content, so no dissolved hydrogen was added, and this also has the advantage to accelerate the corrosion in these tests. This is in agreement with what is conventionally done in static autoclaves to evaluate the behavior of cladding out-of-pile. In these tests, no delamination or dissolution of Cr in the water was ever observed, and the coating remained very adherent.

To analyze the behavior of this EATF solution in degraded water chemistry and evaluate the susceptibility of Cr-coated cladding to harsh environments, a corrosion test in water containing 70 ppm of Li was performed at the AREVA NP Research Center of Ugine. This test is a typical test performed for all zirconium alloys to evaluate their performance in very harsh environments, where zirconium alloys exhibit breakaway corrosion after about 100 days of exposure [16]. Fig. 5 shows the current results after 168 days of exposure. The uncoated M5 exhibited breakaway corrosion after 140 days as expected, whereas the Cr-coated tube (coated only on the external surface), where the inner uncoated surface was exposed to water, still had not experienced breakaway. Once again no weight loss was observed for the Cr-coated samples, and no dissolution of Cr was measured. This suggests that the Cr coating reduces the susceptibility of zirconium alloys to corrosion in lithiated environments, which may increase operational flexibility concerning water chemistry constraints for utilities by modifying some of the current water chemistry constraints. The tests will be continued to determine the extent of the added benefit of using Cr-coated M5.

2.2.2. Wear tests

In the same way, additional benefits for utilities may be obtained by using Cr-coated cladding due to the increased hardness of chromium, which improves wear resistance of the Cr-coated M5 cladding. Parametric tests were performed in the AURORE loop at the AREVA NP Technical Center of Le Creusot under much harsher conditions than what is encountered in a reactor. The goal of these tests was not to be representative but to produce significant wear in a short time and therefore clearly reveal the differences in behavior of the two cladding types.

In this test, Cr-coated and uncoated cladding tube samples were placed in a grid cell where all the dimples and springs were removed except for one spring when testing spring/clad behavior and for one dimple when testing the dimple/clad behavior. The

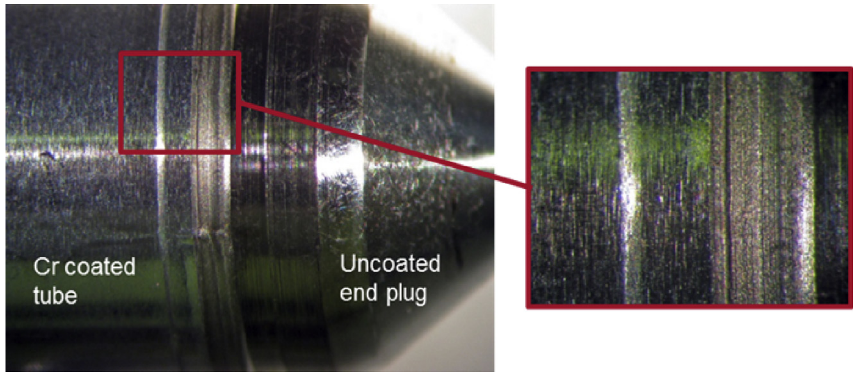

Fig. 4. Visual examination of a weld using AREVA's USW process showing no flaws of the weld. USW, upset-shape welding.

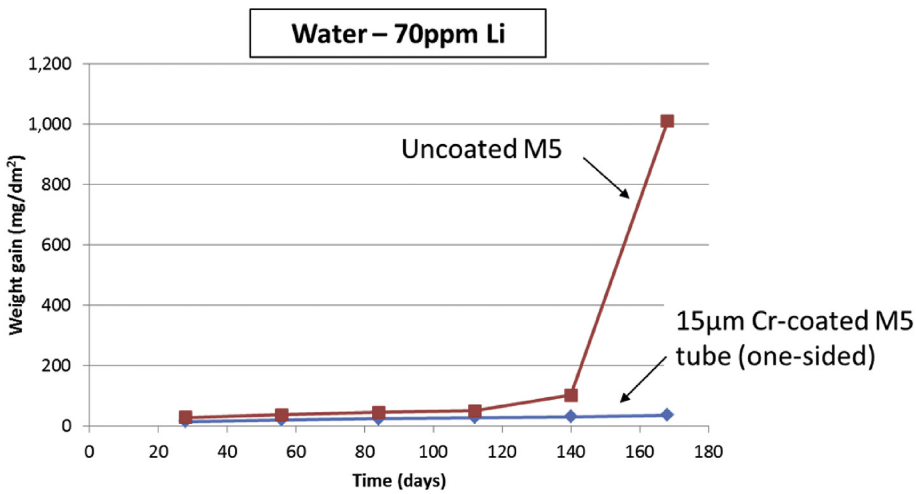

Fig. 5. Corrosion kinetics of uncoated and one-sided Cr-coated M5 tubes in 360°C water with 70 ppm Li.

cladding sample was then submitted to linear displacement of ± 200 μm with a normal load of 1.5N and a frequency of 20 Hz. The tests were performed for a total of 100 hours at 300°C in PWR water conditions ([B] = 1000 ppm; [Li] = 2 ppm) to be representative of the temperature and corrosion conditions during operating conditions.

In the dimple/cladding test, the cladding wear volume was reduced by close to 98% for the Cr-coated cladding compared with the uncoated cladding with almost no observable wear on the cladding. The dimple on the other side exhibited more wear and gradually took the shape of the cladding, which corresponded to an ideal configuration concerning wear and led to saturation of the wear behavior of the cladding/dimple couple.

Fig. 6 gives a schematic representation of the experimental setup for the parametric wear tests and the plot of the total wear depth as a function of friction energy for the clad/spring case. The results show a significant wear resistance of the Cr-coated cladding compared with the uncoated cladding. In the Cr-coated tube, there was negligible wear on both the cladding and the spring, whereas in the uncoated case, it was mainly the cladding that exhibited wear. By using Cr-coated cladding, the total wear volume was reduced by two orders of magnitude!

Consequently, the parametric wear tests performed in the AURORE loop at the Le Creusot Technical Center demonstrated significant improvement in the wear behavior for the Cr-coated cladding compared with the uncoated cladding where the Cr coating serves as a protection for the cladding due to the hardness of chromium and the excellent adherence of the coating developed by AREVA NP. The use of Cr-coated cladding is therefore expected to reduce the number of fuel failures by significantly reducing grid-to-rod fretting failures and potentially reducing debris fretting failures, thus providing economic benefits for utilities.

2.2.3. *HT ramp tests beyond the Cr–Zr eutectic point*

One of the main aspects in evaluating EATF applications is the HT behavior, and especially for coating or multilayer solutions, it is important to investigate the behavior beyond the eutectic point. The Cr–Zr eutectic occurs around 1330°C; therefore, high speed ramp tests reaching temperatures above 1400°C were performed in an argon atmosphere. The ramp speed was 25°C/s, and the maximum temperature reached was around 1500°C, thus much beyond the eutectic formation. After the test, the sample retained its integrity and geometry, suggesting no significant degradation of the sample. In addition, metallographic analysis showed that the chromium diffused inside the zirconium substrate and formed a eutectic-like microstructure over 100 μm from the surface. This result confirmed that the 15 μm coating thickness was small enough to prevent any detrimental effect on the cladding integrity and that the Cr-coated cladding could survive even beyond the eutectic formation. Fig. 7 shows the temperature profile for the test and the visual appearance of the Cr-coated sample before and after the ramp test. Complementary tests are planned in a steam atmosphere to investigate a more representative behavior where there is competition between the diffusion of chromium in

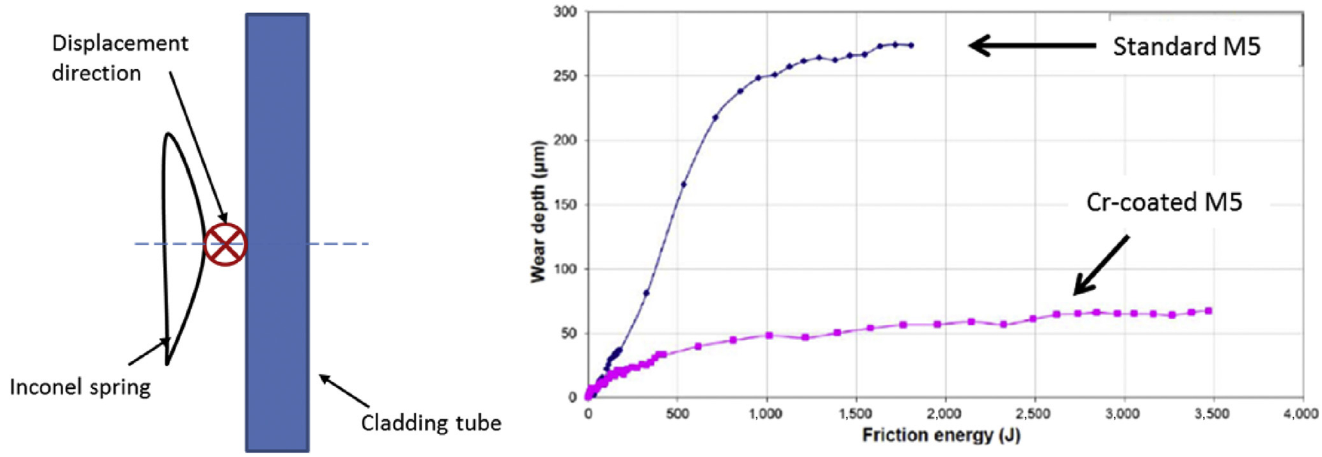

Fig. 6. Schematic representation of the experimental setup for the parametric wear tests and the plot of the total wear depth as a function of friction energy for the clad/spring case.

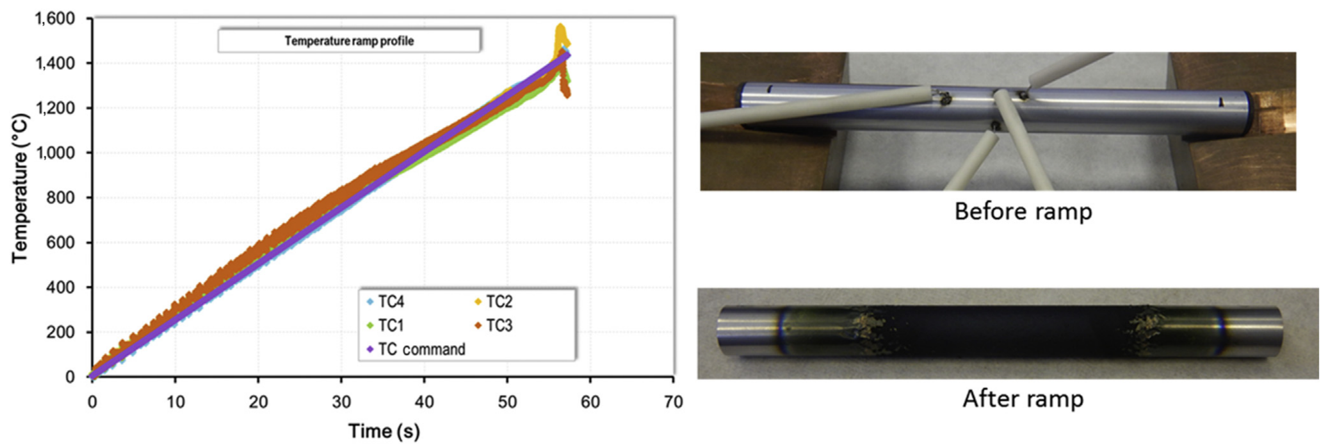

Fig. 7. Temperature ramp profile and visual aspect of the Cr-coated sample before and after temperature ramp up to 1500°C at a speed of 25°C/s. TC, thermo-couple.

the zirconium substrate and the formation of chromium oxide. In this case, the behavior may be slightly different and therefore should be assessed.

2.3. Irradiations

To confirm the excellent behavior of the Cr-coated zirconium alloy cladding observed out-of-pile, an irradiation program was launched in 2016 in the Gösgen reactor in Switzerland, which was the first irradiation of EATF concepts in a commercial reactor. This irradiation project was called IMAGO, which stands for Irradiation of Materials for Accident-tolerant fuels in the GÖsgen reactor. The goal of IMAGO is to verify the behavior of EATF concepts in representative PWR conditions, mainly the corrosion behavior, the microstructural evolution under irradiation, and some mechanical properties. The data obtained will serve as input for the justification of future fuel rod irradiations. Both Cr-coated zirconium alloy and SiC/SiC composite cladding samples were inserted in mid-2016 in the form of material test rods placed within the guide tubes of some fuel assemblies. Some material test rods will be extracted after the first and other subsequent cycles for hot cell postirradiation examinations.

Additional irradiations of Cr-coated cladding are planned in research reactors to evaluate the behavior of fuel rods and the interaction of fuel pellets with the Cr-coated cladding:

As part of the French Joint Research program with CEA and EDF, the irradiation of UO_2-—Cr-coated cladding rodlets (containing both Zy4 and M5 substrates) in the HALDEN reactor started in July 2017. Several pellet diameters will be tested to investigate the impact of pellet—cladding interaction on the Cr-coated cladding behavior.

As part of phase 2 of the DOE ATF program, an irradiation test of AREVA NP's short-term EATF concept (Cr_2O_3-doped UO_2 fuel with Cr-coated M5 cladding) is planned for very early 2018 in the ATR at Idaho National Laboratory in the United States as part of the ATF-2 irradiation. This will be the first irradiation test of the AREVA NP's complete EATF fuel rod concept.

These irradiations will provide complementary information to the IMAGO irradiation for the justification of future LFR irradiations with data more representative of the fuel rod by investigating the overall pellet—cladding interaction in axial and diametral conditions.

3. Conclusion

AREVA NP has set up a worldwide EATF program with work being performed in the United States as part of the DOE project and within the French Nuclear Collaborative program with CEA and EDF. Two cladding solutions are being developed in Europe and have been irradiated in the Gösgen reactor since mid-2016: Cr-coated zirconium alloys and SiC/SiC composite sandwich cladding. Concerning the development of Cr-coated M5, previous results demonstrated its excellent behavior in nominal conditions and the significantly improved behavior in accident conditions. Consequently, the industrialization of the concept was initiated, and the fabrication of a full-length Cr coating PVD deposition prototype is ongoing. The goal is to demonstrate the feasibility of the full-length Cr-coated tube fabrication using PVD and to fabricate in a very short term the first tubes for upcoming LFR irradiations in commercial reactors.

In addition, to support LFR insertion, AREVA NP has launched several irradiation projects in material test reactors to obtain in-pile data to support the justification of fuel rod irradiations. The IMAGO project in the Gösgen reactor started irradiation in June 2016 and is the first irradiation of EATF solutions in a commercial reactor. Other fuel rodlet irradiations are planned in research reactors (HALDEN and ATR) in 2017 and 2018 to investigate the overall behavior of Cr-coated fueled rods.

Finally, the out-of-pile characterizations were performed and confirmed that Cr-coated M5 cladding can provide significant benefits in normal operating conditions in terms of susceptibility to corrosion in harsh environments such as high lithiated water chemistry and in terms of wear resistance. Consequently, the Cr-coated M5 cladding will likely provide utilities with additional plant operational margins and flexibility as well as economic benefits due to reduced fuel rod failures from grid-to-rod or debris fretting, which is a major cause of fuel failures in current PWRs.

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