Accident Tolerant Fuel Cladding Development: Promise, Status, and Challenges[*]

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Abstract

The motivation for transitioning away from zirconium-based fuel cladding in light water reactors to significantly more oxidation-resistant materials, thereby enhancing safety margins during severe accidents, is laid out. A review of the development status for three accident tolerant fuel cladding technologies, namely coated zirconium-based cladding, ferritic alumina-forming alloy cladding, and silicon carbide fiber–reinforced silicon carbide matrix composite cladding, is offered. Technical challenges and data gaps for each of these cladding technologies are highlighted. Full development towards commercial deployment of these technologies is identified as a high priority for the nuclear industry.

1. Introduction

1.1. Origins of Zr-based fuel cladding

Within 18 months of Admiral Hyman Rickover's initial visit to Clinton Engineer Works (Oak Ridge, TN) in June 1946 to discuss the possibility of nuclear-powered naval propulsion, he had made the decision to proceed with zirconium as the fuel cladding in these systems [1]. This decision was facilitated early on by the discovery of the low thermal neutron absorption cross section in Zr once hafnium was separated [2]. A few years later his decision was vindicated through the unintentional, and yet serendipitous, contamination of a batch of Zr-based alloy with stainless steel residue, later named Zircaloy-2, that resolved the poor high temperature water corrosion of this alloy system [3].

After adoption in the 1950s of the light water reactor (LWR) as the main industrial design for commercial electricity production, Zr-based alloys were widely used as the cladding for urania fuel pellets. Various austenitic stainless steels were also sporadically used for this purpose into

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the mid-1980s [4], after which they were discontinued because they offered lower uranium resource utilization and exhibited similar failure rates as the Zr-based alloys [5]. In the 1990s the nuclear power industry systematically drove up its capacity factors to achieve an unprecedented near 90% by the end of that decade, a feat that remains unmatched by any other electricity generation source to this day [6,7]. This was achieved by implementing an excellent operational culture and increasing the fuel burnup [8] while greatly reducing the failure rates of Zr-based fuel cladding [9,10]. In the first decade of the 21st century, fuel reliability programs widely adopted by the nuclear industry achieved impressive results [11,12]. New Zr-based alloys with better performance were also adopted during the past quarter century [13], particularly in pressurized water reactors (PWRs). Most of the Zr-based alloys utilized today are the result of a combination of historic Western (Fe, Cr, Sn addition [14]) and Eastern (Nb addition [15]) alloying strategies with impressive performance during normal operation when compared to their predecessors [16].

1.2. Severe LWR accidents and their impact

Given the awesome power density of LWR cores, ~150 W/cm^3, coupled with the potential for reactivity insertion, or loss of cooling in these complex systems, the engineers who designed these plants recognized the need for safety systems early on. To devise a mitigation strategy, two extreme bounding accidents were postulated and prescriptively defined as a reactivity insertion accident (RIA) and a loss-of-coolant accident (LOCA). The active safety system (emergency core cooling system, ECCS) was then designed specifically to respond to these postulated or design basis accidents (DBAs) by maintaining a controllable and coolable core in case either occurred. In other words, these prescriptively defined accident scenarios were the basis of the ECCS design. Upon review of the regulatory guidance on the requirements for ECCS [17], it quickly becomes clear that they were defined to avoid severe Zr-based cladding degradation under these DBAs [18].

Taleb defines black swan events as those that are deemed highly improbable but, when they occur, are of major consequence [19]. One may point to “beyond DBAs” as an example of black swan events. Such accident scenarios, also referred to as severe accidents, were deemed too improbable (with an assessed probability of <10^{-5} reactor-year [20]) and were not considered in the basis of design for the ECCS. Yet they occurred at the Three Mile Island (1979) and the Fukushima Daiichi (2011) nuclear power stations, totaling to an occurrence rate of >2 × 10^{-4} reactor-year. Note that a large-break LOCA or RIA, the so-called DBA events, has yet to occur in Western LWRs.

The heat equation in its simple form (Equation (1)) is sufficient to grasp the nature of accident progression inside a reactor core. The left-hand side of the equation describes the temperature (T) variation as a function of time (t) that is dampened by the heat capacity (ρC_p) of the materials in the core. The first term on the right-hand side generically represents, in this simplistic form, heat transfer processes (whether conduction [thermal conductivity, k], convection, or radiation) to remove heat from the core. The second term on the right-hand side is the heat generation rate (Q).

$$\rho C_p \frac{\partial T}{\partial t} = k\nabla^2 T + Q \tag{1}$$

During severe accidents, core cooling is often interrupted and the decay heat in the fuel (Q in Equation (1)) drives the core temperature upward [21]. As the core becomes uncovered (water level decreases), the heat transfer processes become less efficient, temperatures rise further, and the fuel elements start to experience physical and chemical degradation. Physical degradation occurs first (700–1000°C) and involves ballooning and burst of the thin-walled cladding tube; its extent and consequences have been extensively studied [22–25]. Chemical degradation is dominated by steam oxidation of Zr metal, which is highly exothermic. For instance, the ~125 kg of Zr metal in each PWR fuel assembly produces >820 MJ of heat and >2700 mol of hydrogen gas after it undergoes oxidation. Depending on the LWR design, the ~25–40 tonnes of this metal present in the core, if fully oxidized, would produce an excessive amount of heat (adding to Q in Equation (1)), in turn exacerbating the course of accident progression and furthering the burden on ECCS, Figure 1.

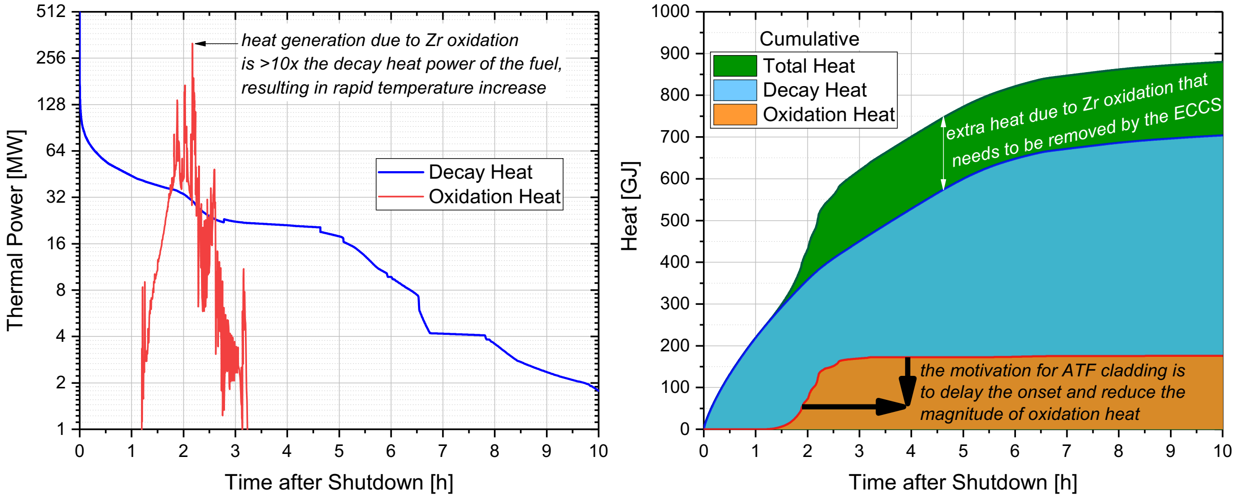

Figure 1. Thermal power and cumulative energy due to radionuclide decay heat and Zr-based cladding oxidation heat during a short-term station blackout [26] *(courtesy of Kevin Robb).*

1.3. Motivation for ATF cladding

The motivation for transitioning away from Zr-based cladding to an accident tolerant fuel (ATF) cladding alternative is straightforward: to reduce the burden on the ECCS during severe accidents by decreasing the rate and total amount of heat generated from cladding oxidation in high temperature steam. This reduces the Q in Equation (1), in turn lowering the rate of temperature rise on the left-hand side of the equation. The slower temperature rise delays core degradation processes, provides additional coping time, and reduces the threshold on the cooling required to mitigate accident progression. These effects are now well-studied and documented [26–29].

Since reducing the rate and total amount of heat generated as a result of steam oxidation is the basis behind development of ATF cladding materials, it is worthwhile to ask whether there exist materials with significantly higher steam oxidation resistance. The answer to this question was not readily clear after the 2011 Fukushima Daiichi accident. Simply put, few technological areas assign research interest to the oxidation behavior of materials in a high-partial-pressure steam environment at $T > 1200°C$. However, soon after this accident, various research and development (R&D) programs were launched across the globe to address this question [30–33]. Early studies showed that although high temperature steam is a far more aggressive environment than dry O_2 [34,35], the three conventional classes of protective oxide films—namely, chromia, alumina, and silica—may be utilized to protect the underlying materials [36]. This is shown in Figure 2, the parabolic oxidation rate constant, for various cladding materials and their resulting oxide films when they are exposed to steam at elevated temperatures.

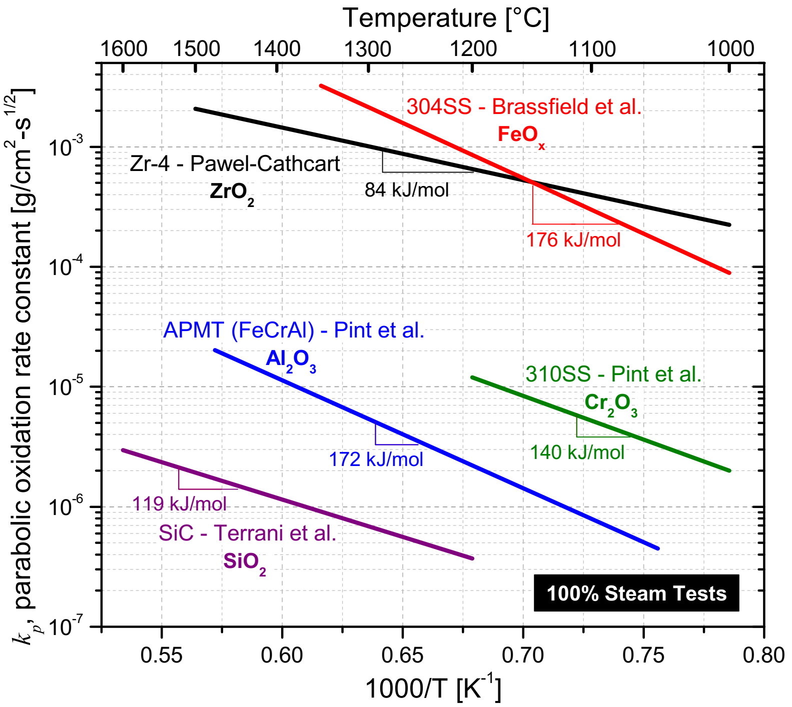

Figure 2. Parabolic oxidation rate for various cladding materials and their resulting oxide in steam as a function of temperature [36–39].

To resist high temperature steam oxidation, the oxide films need to be physically and chemically stable in steam while also acting as effective barriers against the transport of the oxidizing species (i.e., by limiting solid state diffusion of dissolved O, OH, H_2O) to reach the underlying material. Although zirconia exhibits exceptional thermodynamic stability in steam and excellent adherence to the underlying metal at $T > 1100°C$ [40], it is a fast conductor of oxygen and does not protect the underlying Zr metal. Chromia, alumina, and silica, on the other hand, exhibit acceptable stability in steam (note that they may react with steam and volatilize at elevated temperatures [41–43], although slowly) while acting as effective barriers for diffusion of oxidizing species and reaction products. As shown in Figure 2, the parabolic oxidation rate for the materials that form and are in turn protected by these oxide films is roughly two orders of magnitude lower. This reduction in oxidation rate directly translates to the reduction in the rate of heat and hydrogen generation in a LWR core during the course of a severe accident.

The three main ATF cladding technologies under active development across various international programs consist of materials that form one of these three protective oxides. This paper offers a review of these ATF cladding technologies. First, the design philosophy and the desirable properties of a generic LWR fuel cladding material are discussed. Then the focus shifts to specific concepts, development progress, and challenges. For the latter, it is intended that fundamental material system challenges be discussed, rather than engineering and development issues to optimize production of these technologies.

2. LWR cladding design philosophy

Nuclear fuel cladding is the first full barrier for retention of radionuclides, followed by the reactor pressure vessel and the containment building in a typical LWR plant design. This function, and its reliability, define the LWR fuel cladding. Fuel cladding may not be thought of as a structural material with its associated functional requirements. These distinctions, which may appear subtle at first glance, are profound. Although excessive dimensional change in the cladding during the fuel lifetime is undesirable (e.g., excessive irradiation growth [44] and assembly bowing [45]), unlike structural materials, a good degree of flexibility for deformation is highly desirable in LWR fuel cladding.

2.1. Typical evolution

Figure 3 depicts a simplified progression of diametral evolution in LWR fuel cladding and pellet as a function of burnup. As the fuel pin powers up, the pellet and cladding experience immediate thermal expansion, adjusting the thickness of the initial He-filled gas gap between them. Since the He pressure inside the rod is always lower than the coolant pressure in the system, compressive hoop stress results in creepdown of the cladding onto the fuel pellet. The pellet, containing ~5% porosity in the as-fabricated state, initially undergoes irradiation-induced densification [46]. However, after reaching low burnups (<5 MWd/kgU), it experiences swelling due to generation of fission products that manifests linearly with burnup at a rate of 0.7–1% per 10 MWd/kgU [47,48]. Depending on the fuel pin's power history, the gap between the pellet and the cladding disappears at ~20 MWd/kgU due to the concomitant swelling of the former and creepdown of the latter. After this point, the stress state in the cladding is dominated by its mechanical interaction with the fuel pellet [49,50]. The ability of the cladding to exhibit ductility after this point and accommodate pellet expansion is key to its integrity. Rapid pellet expansion occurs at different rates during operational power ramps (up to 10^{-5} s^{-1} [51]) or sudden RIAs (up to 5 s^{-1} [52]). Although urania itself exhibits substantial creep, >10^{-10} s^{-1} [53–55], sufficient creep and ductility in the LWR cladding are necessary to avoid failure. Note that while radiation damage reduces the ductility of Zr-based alloys [56,57] like most other metals [58], the most significant loss in ductility and ability to accommodate strains in Zr-based fuel cladding occurs due to hydrogen pickup as a result of its waterside corrosion [16,24,59–62].

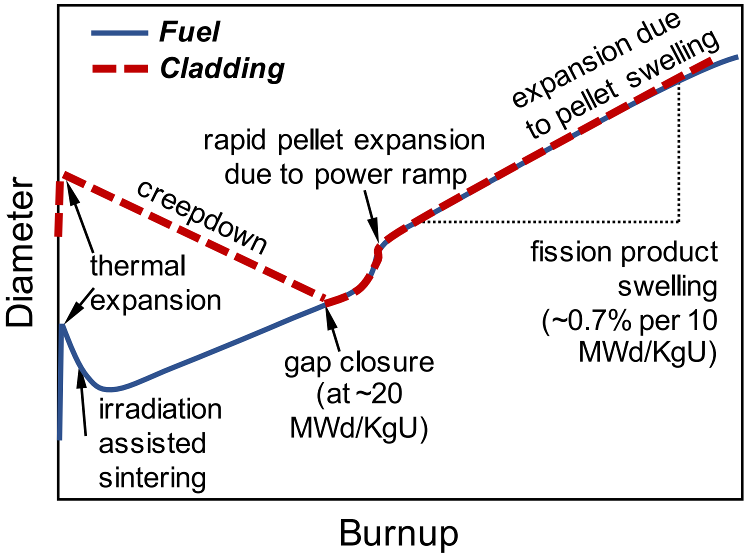

Figure 3. A simplified progression of diametral evolution in LWR fuel cladding and pellet as a function of burnup.

The typical behavior described above is different from what most engineers are taught to deem as superior properties for structural materials [63], where significant emphasis is placed on strength and creep resistance. Even for sodium-cooled fast reactor cladding, the functional requirements differ: a malleable fuel and cladding with high strength and creep resistance are coveted [64] since any small changes in core geometry directly and significantly affect core physics.

2.2. Desirable properties

Table 1 attempts to organize the desirable properties of LWR fuel cladding and pellet under various normal and off-normal conditions. During normal operation, high thermal conductivity in the pellet and cladding are desired to limit fuel centerline temperature [65]. Adequate, but not excessive, creep in both components and high cladding strength are desired to avoid and withstand stress buildup in the cladding. High critical heat flux for cladding heat transfer to the coolant is desirable to avoid departure from nucleate boiling and dryout in PWRs and boiling water reactors (BWRs), respectively [66]. Minimization of cladding corrosion to avoid thickness loss in the load-bearing cladding tube is desirable. As noted earlier, since Zr-alloy corrosion is accompanied by concomitant H pickup [67], reducing its rate is of further importance.

Under power ramps, high thermal conductivity and creep rates along with a low coefficient of thermal expansion are desirable in the fuel pellet to minimize pellet–cladding mechanical interaction (PCMI). In the cladding, high strength along with the ability to accommodate pellet expansion via creep is highly desirable.

For a design basis LOCA, high thermal conductivity is desirable in the fuel and the cladding to reduce the thermal energy stored in the pin. At the same time, high heat capacity is also desirable since it will reduce the rate of temperature rise as a result of decay heat. Note that for specific LOCA sequences in a BWR, the impact of improved fuel thermal conductivity is negligible [68]. Of great importance during this scenario is reduction of cladding oxidation rate to limit its degradation during accident progression and loss of post-quench ductility [24,69]. Cladding creep resistance is highly desirable to delay fuel rod burst and ballooning [70].

Under an RIA scenario, as explained by Fuchs and Nordheim [71,72] and in direct contrast to a LOCA scenario, low heat capacity in the fuel is desirable to limit the extent of energy deposition in the core. Since the fuel pellet's coefficient of thermal expansion directly dictates the extent of PCMI, a smaller value for this material property is desirable. A cladding with high heat transfer characteristics and ductility is ideal under an RIA scenario [73,74].

Finally, for a beyond DBA scenario such as a station blackout (SBO), where core temperatures far in excess of 1200°C are possible [75], high temperature steam oxidation resistance, as well as overall chemical and physical stability, is of most importance [76].

Table 1. Desirable properties of LWR fuel cladding and pellet under normal operation (NO), power ramps (PR), loss-of-coolant accident (LOCA), reactivity insertion accident (RIA), and station blackout (SBO). Upward and downward arrows indicate that higher and lower magnitudes are desirable for a given property, while a dash indicates insensitivity.

	Cladding					Fuel Pellet				
	NO	PR	LOCA	RIA	SBO	NO	PR	LOCA	RIA	SBO
Thermal conductivity	↑	↑	↑	-	-	↑	↑	↑	-	-
Heat capacity	-	-	↑	-	↑	-	-	↑	↓	↑
Oxidation rate	↓	↓	↓	↓	↓	-	-	↓	-	↓
Coefficient of thermal expansion	-	-	-	-	-	-	↓	-	↓	-
Creep rate	↑	↑	↓	-	↓	↑	↑	-	-	-
Strength	↑	↑	↑	↑	↑	-	↓	-	↓	-
Critical heat flux (CHF)	↑	↑	-	↑	-	N/A				

Note that although under different scenarios lower or higher magnitudes for some of the material properties are desired in the cladding and the fuel, slow oxidation rate and high strength are always desirable in the cladding.

3. Coated Zr-based cladding

An immediately obvious and evolutionary approach to ATF cladding is the adoption of a protective coating on the surface of Zr-based alloys [77]. Thin coatings are expected to have a minimal effect on the thermomechanical behavior of Zr-based cladding, assuming sufficient creep and limited strain mismatch are engineered into the coating [78]. Thin coatings do not notably change the core physics in LWRs [79] and have the potential to enhance the heat transfer characteristics of the cladding [80]. It is necessary that the coating be adherent to and chemically stable with the Zr-based cladding substrate during normal operation and off-normal conditions, protecting it from rapid oxidation during beyond DBAs. Coatings on the surface of Zr-based alloys were previously considered to enhance their corrosion resistance and alleviate susceptibility to grid-to-rod fretting failure [81,82]. As noted in Section 1.3, materials that are capable of exhibiting high temperature steam oxidation resistance are chromia, alumina, and/or silica formers. Therefore, any ATF coating technology needs to contain at least one of the elements Cr, Al, or Si.

3.1. Development status

The most widely explored coating technologies on Zr-based alloys to date are the ones that form chromia. Specifically, Cr metal [83–85], CrAl [86], and CrN [87,88] coatings have been studied. In the case of a metallic Cr coating with a thickness of a few to tens of micrometers, Figure 4, the resulting chromia that forms under both aqueous or high temperature steam conditions protects the underlying Zr metal [89,90]. Furthermore, reduced cladding ballooning during LOCA testing and resistance to cladding post-quench ductility loss have been reported for these coatings [91,92]. Multiple in-pile experiments to further evaluate the performance of this technology are currently ongoing, with preliminary ion irradiation data indicating adequate behavior [93]. As a BCC (body-centered cubic) metal, it is expected that Cr metal will exhibit dimensional stability during neutron irradiation at LWR-relevant temperatures [94].

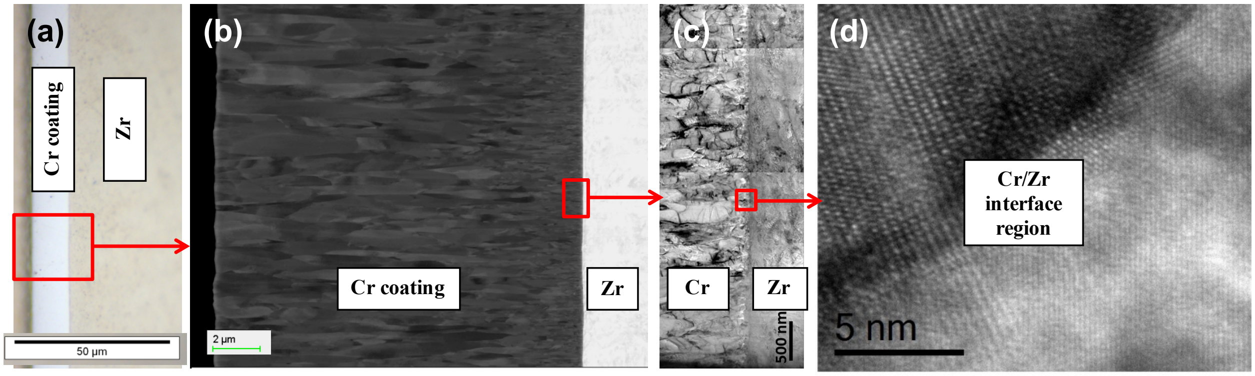

Figure 4. Metallic Cr coating of thickness 12–15 µm deposited on the surface of Zr-based cladding and characterized by a) optical microscopy, b) backscattered scanning electron microscopy, c) bright field, and d) high resolution transmission electron microscopy. The coating appears to be fully dense and homogeneous while the Zr–Cr interface shows good metallurgical bonding without indications of cracks or voids (courtesy of Jean-Christoph Brachet and colleagues at CEA, SRMA, Paris-Saclay University, France).

Excellent stability of a thin (<5 µm) CrN coating on the surface of Zr-based cladding under prototypical fuel irradiation conditions has been demonstrated [95]. Integral LOCA testing of unirradiated CrN-coated cladding exhibited excellent coating adherence even after burst testing but showed no improvement in oxidation or burst behavior vs. uncoated cladding, Figure 5. Although the adverse effects of N during air oxidation of Zr-based alloys are well understood [96], the small quantity of this element in the thin coating is not expected to cause large degradation of the cladding during high temperature steam oxidation.

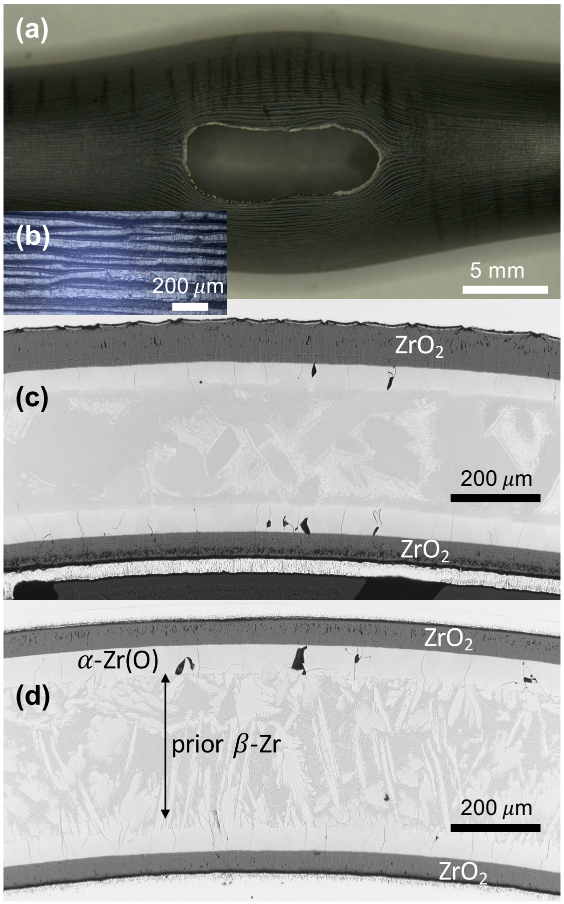

Figure 5. a) CrN-coated Zircaloy-4 cladding after pressurization to 8.3 MPa and LOCA burst testing. b) High magnification optical image of the CrN coating on the surface. Metallographic cross sections of the c) CrN-coated and d) uncoated Zircaloy-4 after the LOCA test sequence conducted at ORNL's Severe Accident Test Station [97] *with 5°C/s ramp to 1200°C in flowing steam (courtesy of Rudi Van Nieuwenhove and Bruce Pint).*

Coatings that are meant to form alumina or silica have predominantly manifested as MAX-phase compounds [98] or FeCrAl in the case of the former, with other variants discussed in [77]. Ti_2AlC [99,100], TiAlN [101,102], Ti_3SiC_2 [103], and Cr_2AlC [104] as MAX-phase coatings have been examined, although none of these examinations to date have produced a complete assessment of coating performance under normal operation, DBA, and beyond DBA conditions. The FeCrAl coating, although adequate for normal operating conditions, forms a eutectic with Zr at temperatures <1200°C and is not deemed a useful ATF coating [105]. Other coating materials [77] that do not contain Cr, Al, or Si and therefore cannot produce a protective oxide film are not considered in this review. Although these coatings may be beneficial from a normal operational standpoint (e.g., TiN [101]), they may not be considered as ATF cladding technologies.

It is worthwhile to review basic properties of the various proposed coatings, to decipher which technologies among the myriad of candidates are worth further development as coated Zr-based ATF cladding concepts. The coatings, assuming they can be reliably deposited and remain adherent to the surface of Zr-based cladding, need to exhibit the following: corrosion resistance in LWR coolant environments, neutron irradiation stability, and oxidation resistance to high temperature steam. Table 2 provides a summary of these properties for select coatings. Note that if only one of these performance characteristics is not met, the coating technology is deemed not useful for the ATF cladding application.

Table 2. Summary of coating performance characteristics of select ATF cladding technologies. Y, N, and U denote yes, no, and unknown, respectively.

	Cr	CrN	CrAlN	TiAlN	TiN/ TiAlN	Ti_2AlC	Ti_3SiC_2	CrAlC
Resistant to corrosion in LWR coolant	Y [106]	Y [95]	N [107]	N [107]	Y [102]	U	U	N [104]
Stable under neutron irradiation (260–400°C)	Y [94]	Y [95]	U	U	U	N [108]	N [109]	U
Increased resistant to high temperature steam oxidation	Y [92]	N [110]	U	Y [111]	U	Y [36]	N [36]	U

A glance at Table 2 implies that Cr metal coating is the most promising technology for further development. The same protective oxide films that protect Cr-, Al-, and Si-bearing coatings at high temperatures also form in the aqueous environment of the LWR coolant [77]. Unfortunately, only chromia is stable in this environment, while silica and alumina tend to dissolve rapidly as silicic acid, H_4SiO_4, and aluminum oxy-hydroxide, AlO(OH) [112]. Incorporation of Ti, which forms a stable oxide (much like Zr), into these coatings can mitigate dissolution (e.g., TiN/TiAlN [102]); however (again much like Zr), Ti undergoes rapid oxidation at elevated temperatures and its prevalence in the coating will likely compromise the protective nature of alumina/silica (e.g., protective silica does not form during high temperature oxidation of Ti_3SiC_2 [36]).

3.2. Challenges and research needs

3.2.1. Zr remains in the core

The same attribute that makes surface coatings on Zr-based alloys the most viable near-term ATF cladding technology presents their biggest performance challenge: the ~25-40 tonnes of Zr metal remains in the LWR core. For a coolant-limited accident, even a design basis LOCA, rod ballooning and burst occurs at temperatures as low as 700°C [113]. This exposes at least some fraction of the cladding's internal surface to the oxidizing environment, even though the outer surface may be protected by the coating. A recent and ongoing effort aims to tackle this issue by adding an inner surface coating [114].

3.2.2. Need for elucidation of beyond DBA behavior

Except for a few studies [106,115], none of the research groups to date have exceeded the temperature limit of the design basis LOCA scenario (1204°C [116]) during their steam oxidation tests when examining the coatings discussed in Table 2. These environments are indeed demanding on the coating, which needs to withstand steam oxidation on one side and chemical reaction with the highly reactive Zr metal on the other [105].

Ultimately, owing to the R&D programs discussed above, several emerging coating technologies hold abundant promise for improving fuel performance during normal operation (Cr, CrN, and TiN) and should be pursued to elevate their technology readiness level towards ultimate deployment in commercial reactors. The reader is reminded that Zr-based cladding technology is deemed safe and adequate for normal, anticipated operational occurrence (AOO), and DBA scenarios. However, beyond DBA testing must be conducted on ATF cladding technologies to showcase their improvements in cladding performance; such instances are glaringly lacking for almost all coating-based ATF technologies under development to date.

Brachet [89] provides a sound explanation of how Cr-coated Zr-based cladding improves the peak cladding temperature limit of the design basis LOCA (by ~100°C) and extends the time at elevated temperatures before post-quench ductility is lost (hours for coated cladding instead of minutes for uncoated cladding). The impact of these additional safety margins on the beyond DBA behavior of LWRs should be assessed through use of severe accident analysis codes. Without significantly more experimental data, an assessment of this and other coating technologies is not possible.

4. FeCrAl cladding

Fe-based alloys have been used as nuclear fuel cladding since 1951, when the Experimental Breeder Reactor I (EBR-I) first went critical with austenitic stainless steel–clad Mark-I fuel assemblies [117]. Naturally, the use of these austenitic alloys was extended into commercial LWRs [4] for decades, even though the first LWR power station in Shippingport, Pennsylvania, started its operation with Zr-based clad fuel provided by Admiral Rickover’s navy [118]. Austenitic stainless steels (types 304, 316, and 347) used as cladding in BWRs were eventually replaced with Zr-based cladding due to the stress corrosion cracking (SCC) failure experienced in high-oxygen-activity coolant environments of the pre-1990s era without water chemistry control [5]. This failure mode is distinct from a previously common mode of SCC failure in Zr-based alloys: corrosive fission products initiated failure in the inner surface of the cladding [119]. Although austenitic steel–clad fuel operated reliably in PWRs [5], the drive to achieve higher burnups, and by extension better economics, also facilitated their eventual replacement by Zr-based cladding.

Ferritic steels, having a BCC structure as opposed to the Ni-stabilized FCC (face-centered cubic) structure of austenitic Fe-based steels, are known to exhibit better SCC resistance [120] but were never adopted for use in commercial LWRs, although they were used in sodium-cooled fast reactors [64]. Ferritic alloys have also been developed for other potential commercial nuclear power applications such as the development program by General Electric in the 1960s, which aimed to take advantage of the high temperature oxidation resistance of FeCr, FeAl, and FeCrAl ferritic alloys, for high temperature reactor applications [121–127].

After survey tests examining a variety of candidate Fe-based alloys [34,35], reexamination of oxidation-resistant Fe-based alloys for LWR application was proposed [5]. Once again, alloys capable of forming protective chromia, alumina, or silica were to be examined. To form protective chromia films, ferritic steels with Cr content >22 wt% were deemed necessary [35,128], far surpassing the Cr content above which α' phase separation and resulting embrittlement occurs in the Fe-Cr binary system at LWR-relevant temperatures [129,130]. For austenitic alloys capable of forming a protective chromia at 1200°C (e.g., 310 SS [35]), similar Cr contents had to be accompanied by high Ni additions to preserve the FCC structure. This in turn results in high thermal neutron absorption in the cladding (~10× higher than Zr-based cladding [79,131]) and is deemed impractical. Additions of 2 wt% Si to Fe-12Cr did significantly improve steam oxidation resistance, but protective scale formation in steam was limited to <1100°C [132]. While alumina-forming austenitic alloys [133] are not oxidation resistant under these same conditions, exceptional steam oxidation resistance was observed for ferritic FeCrAl alloys up to near their melting point (~1500°C) [134–136].

4.1. Development status

Dedicated R&D programs in the United States [137–139] and Japan [140–142] are pursuing FeCrAl cladding as an ATF cladding technology. Although the main focus of the former program is on developing wrought oxidation-resistant alloy variants [143], the Japanese effort intends to also greatly improve on the strength by pursuing oxide dispersion strengthened (ODS) FeCrAl alloys [144].

To avoid embrittlement of FeCrAl cladding as a result of the α' precipitation [129,130,145–148] that occurs after irradiation at ~300–400°C (LWR-relevant temperatures), lean (≤13 wt% Cr, ≤4 wt% Al) commercial FeCrAl alloys were evaluated for this application. However, these lean compositions, although resistant to oxidation in air, were discovered to have poor resistance to high temperature steam [35]. Accordingly, systematic studies of the critical Al and Cr contents in the alloy system were performed to identify the necessary combination for adequate steam oxidation resistance up to ~1500°C [134,136,141,149,150] while minimizing the potential for α'-induced embrittlement or weld-initiated cracking [151]. Furthermore, the effects of alloy composition on its melting point [152], oxidation beyond melting point [135], and compatibility with other fuel assembly constituents have been examined [132,140,153].

An up-to-date review of various properties and behavioral aspects of FeCrAl alloys for the ATF cladding application is available in [154]. Production of wrought [143,155,156] and ODS [157,158] bar stock and thin-walled cladding has been described and demonstrated, Figure 6. As-fabricated strength and ductility in FeCrAl alloys may be tuned by controlling alloy composition and microstructure to achieve values comparable to or higher than Zr-based alloys [143,158]. Mechanical properties after neutron irradiation, for dose regimes relevant to LWR fuel cladding (Table 3), have been quantified [145,148]. It is shown that although the strength increases and uniform elongation approaches nil at <7 dpa, the irradiated material retains sufficient ductility as total elongation on the order of ~10% up to at least ~15 dpa. A variety of recent welding studies [151,159,160] and limited neutron irradiation data [161] on these welds are now available, indicating robust fusion-based welds are possible through strict control of the process and alloy composition. Corrosion behavior of these alloys in LWR coolant environments has been examined, and resistance to uniform corrosion via formation of Cr-rich oxides (wholly distinct from alumina formation in high temperature steam) without a notable dependence on alloy composition has been reported [140,162–165], Figure 7. Owing to their BCC structure, ferritic alloys exhibit roughly 100× better SCC resistance in aqueous environments when compared to austenitic stainless steels [120,166]. This, combined with the strict water chemistry control of today’s LWRs, assuages the historical concern with Fe-based clad fuel pin failure.

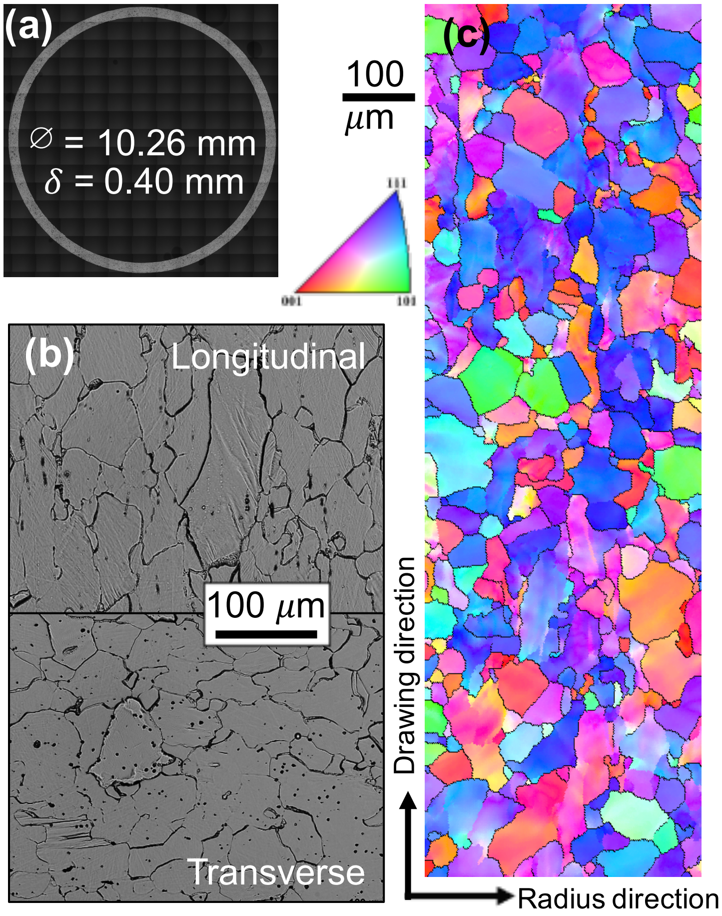

Figure 6. a) Commercially fabricated, thin-walled Fe-12Cr-6Al-1Mo cladding in BWR geometry; b) optical micrograph of the grain structure in the longitudinal and transverse directions, showing slightly elongated grains of ~61 µm; c) inverse pole figure map of grain orientation in the radial direction after a tube drawing process in which deformation is accommodated heterogeneously [156] *(courtesy of Yukinori Yamamoto).*

Table 3. Displacements per atom (dpa) per effective full-power year (EFPY) for various cladding materials in LWRs. Calculation used the SPECTER [167] *code and lifetime average neutron flux spectra tallied within the LWR cladding (courtesy of Nicholas Brown).*

Cladding Material	E_d* (eV)	PWR (dpa/EFPY)	BWR (dpa/EFPY)
Zr	40	3.9	3.1
Cr	40	4.1	3.3
Fe-12Cr-6Al	40, 40, 27	4.0	3.3
SiC	35, 20	5.7	4.6

*E_d is the displacement energy in the NRT model [168] associated with each element.

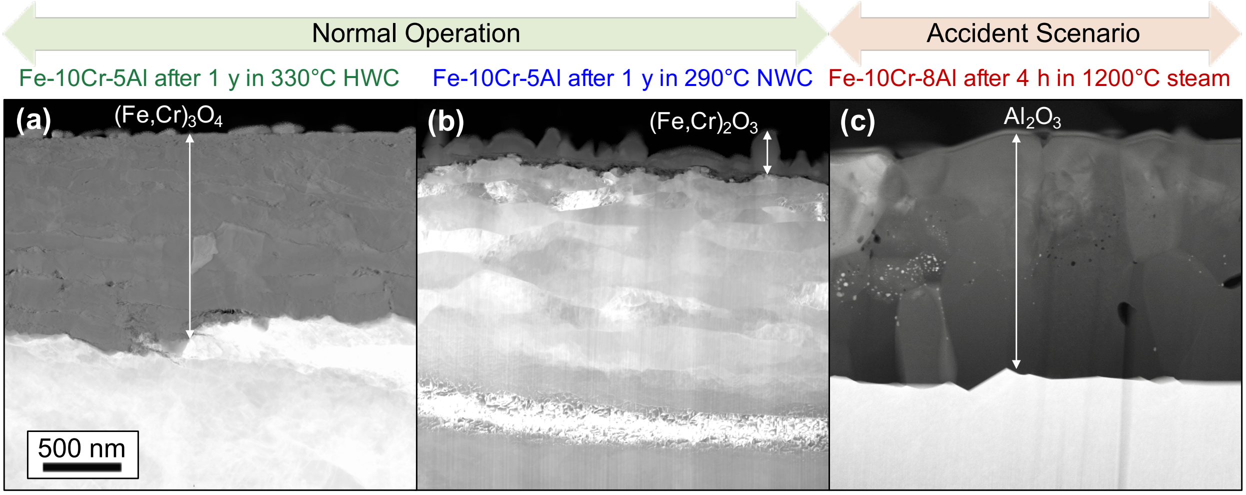

Figure 7. Depending on the environment, various protective films form on the surface of FeCrAl alloys. a,b) In low and high oxygen activity aqueous environments, and largely insensitive to the Cr content in the alloy within a 10–18 wt% Cr window, protective Cr-rich spinel or hematite films form [162]*. c) Under high temperature steam oxidation, if a critical and sufficient combination of Cr and Al is present in the alloy* [134]*, protective alumina forms. High angle annular dark field (HAADF) transmission electron images are courtesy of Kinga Unocic.*

Fuel performance and off-normal behavior of FeCrAl-clad UO_2 pellets, in their optimal thin-walled geometry, have been assessed [169–171], indicating reliable performance. Normal operation and AOO behavior of the FeCrAl cladding is expected to be superior to that of Zr-based cladding, given the enhanced oxidation resistance during post-CHF events and indications of higher CHF [172]. LOCA simulation tests on low-strength wrought alloys indicate comparable burst behavior to Zr-based cladding [173], while no loss of post-quench ductility was observed [174,175]. Adoption of ODS FeCrAl variants with higher strength and high temperature creep resistance [157,158], owing to the dispersion of fine oxide particles [176], is expected to greatly enhance burst margins while preserving the oxidation resistance to near melting. Much like unirradiated Zr-based cladding, FeCrAl cladding exhibits abundant ductility during rapid PCMI tests [177], simulating RIA events. Since the loss of ductility after neutron irradiation is not exacerbated by hydride precipitate formation in these alloys, better PCMI performance is also expected at high burnups.

4.2. Challenges

4.2.1. Poor thermal neutron utilization factor

The composition of nuclear-grade FeCrAl alloy has been optimized (10–13 wt% Cr, 5.5–6 wt% Al, 2 wt% Mo for solid solution strengthening [143], and <0.1 wt% Y to exploit the reactive element effect [178]) to alleviate embrittlement concerns raised early on [5] while exhibiting robust environmental stability. However, other challenges remain with this cladding system that are inherent to the material. The first is the magnitude of the neutron absorption cross section of natural Fe and Cr (with known uncertainties [179]), resulting in roughly ~4–6% of thermal neutrons being absorbed in the cladding vs. ~1% absorbed in the Zr-based cladding [79]. To compensate for this absorption, the pellet enrichment may be increased and/or cladding thickness reduced and pellet dimeter extended, allowing for cycle lengths comparable to the current fuel system in PWRs [131,180] and BWRs [181]. In both cases, this is accompanied by extra cost (i.e., cost of enrichment or cost of more LEU mass), resulting in a ~15–25% increase in fuel bundle cost [5]. This economic impact is further scrutinized in Section 6.

4.2.2. Potential for increased tritium release

Another challenge that requires further understanding and resolution is the permeability of BCC ferritic alloys to H isotopes [182], specifically tritium that forms as a result of ternary fission in the fuel. In PWRs a substantial inventory of tritium is produced from the dissolved boric acid in the coolant via the $^{10}B(n, 2\alpha)^{3}H$ reaction, which is roughly equal to the extent produced in the fuel and is contained within the primary loop [183]. In BWRs, in the absence of dissolved B in the coolant, the main source of tritium production is the fuel, and since the reactor system injects steam directly from the core to the turbines, the increased ^{3}H release from the fuel is impactful. In the current fuel system, the strong chemical affinity of Zr for H [184] traps the tritium as it diffuses away from the fuel towards the coolant. In the case of ferritic alloys, permeability roughly two orders of magnitude higher than that of Zr-based alloys and twice that of austenitic Fe alloys has been reported [183]. The full impact of increased release needs to be understood, as it previously occurred in PWR and BWR cores with austenitic stainless steel–clad fuel pins [4]. Permeability to H may be retarded by oxide films [185–187] on the surface of the cladding, which could be engineered during fabrication or grown in situ during operation. Since alumina dissolves in high temperature water [112], potential candidates are the outer (Fe,Cr)-rich oxide that forms during uniform corrosion [162] or an inner alumina that may form, as suggested in [183], as a result of fuel–cladding chemical interaction. Evidence that the former is an effective barrier has recently been made available [141]. Relevant to formation of an alumina barrier in the cladding's inner surface, a recent study of FeCrAl oxidation in air reported formation of only 10–50 nm oxides after 100 h exposures at 300–600°C, with crystalline alumina only forming at the highest temperature [188].

4.2.3. Need for elucidation of beyond DBA behavior

A final consideration regarding the performance of this cladding material under beyond DBA conditions is worthwhile. Although the UO_2-FeCrAl system has received the most complete severe accident analysis assessment of any ATF fuel candidate to date [27,189], with the aim of quantifying the gains in margins of safety and coping time during such events, additional work is necessary. While alumina formation, inherent to the bulk material, offers remarkable oxidation resistance up to ~1500°C, rapid oxidation occurs at temperatures near the melting point of the alloy [135]. The integral behavior of the FeCrAl-clad pin and fuel assembly beyond this point is not well understood. Integral fuel bundle oxidation tests up to and beyond this point, such as the ones described in [190], are necessary to gain additional information.

The results of the previously referenced severe accident simulations show that although significantly less H_2 and CO are produced, additional coping time for short-term, long-term, and partially mitigated SBO scenarios is on the order of 1–5 h, increasing with delays in the accident's onset after core shutdown. The usefulness of this additional coping time, although deemed superior to the coated cladding ATF concepts, needs to be fully assessed by reactor operators and regulators.

5. SiC/SiC cladding

SiC fiber reinforced SiC matrix composite (SiC/SiC) technology has yielded engineering materials used today in highly demanding applications, such as components in commercial jet engines [191]. Remarkably, the technology originated from nuclear-energy-related R&D in 1970s, when Yajima first produced high strength SiC fibers [192,193]. Application of bulk SiC in fission energy systems dates back even further and persists to this day, with its use as a constituent of coated fuel particles in high temperature gas-cooled reactors [194].

Today, new generations of SiC fiber and methods of composite production that yield nuclear-grade SiC/SiC are available [195]. Nuclear-grade SiC/SiC is defined here as composites utilizing Generation III SiC fibers [196] with chemical vapor infiltrated (CVI) [195] or nano-infiltration transient eutectic phase (NITE) [197] SiC matrices. CVI consists of chemical vapor deposition of SiC using methyltrichlorosilane (MTS) or ethyltrichlorosilane (ETS) onto the SiC fiber skeleton to form a dense composite [198]. The NITE process forms a sintered SiC matrix with high crystallinity from nanopowder feedstock to avoid damaging the fibers during forming [199]. Other SiC-matrix-forming methods such as melt infiltration (or liquid silicon infiltration) [200], reaction sintering [201], or polymer impregnation and pyrolysis [202], yielding large fractions of secondary phases or amorphous pockets in the material, are not deemed radiation stable [203].

Owing to their exceptional oxidation resistance [204] and high temperature strength [205,206], far surpassing Cr-coated Zr-based or FeCrAl cladding materials, SiC/SiC composites are deemed the ideal ATF cladding material.

5.1. Development status

Sauder provides a complete summary of SiC/SiC development and properties for nuclear applications [207], and many other useful reviews focusing on application of these materials in fusion energy systems are also available [208–210]. With the high temperature strength and oxidation resistance of SiC-based materials recognized, their application as LWR fuel cladding was proposed early on [211–213].

Two distinct production routes for SiC/SiC cladding are in use today, conforming to the two radiation-stable methods described earlier. The isothermal CVI methodology is the most common approach for cladding production used in the United States [214,215], France [216], South Korea [217], and Japan [218]. This method results in a highly pure and crystalline, and therefore highly radiation-stable [219], composite of relatively low density (with 10–25%

porosity) [195]. Note that other viable and potentially attractive CVI techniques (e.g., forced-flow, thermal-gradient CVI [220]) yielding higher density are not in use today for SiC/SiC cladding production. The second route for production of SiC/SiC cladding, primarily pursued in Japan [221], involves the NITE process, with utilization of hot pressing for production of cladding also demonstrated elsewhere [222]. This methodology delivers a dense material with improved mechanical properties [223]. A combination of the CVI and NITE techniques for nuclear-grade SiC/SiC production has also been suggested [224].

Besides establishing a robust production methodology, the focus of the past decade's development efforts has been fixated on understanding and quantifying the mechanical behavior of SiC/SiC composite tube structures [217,225,226], developing radiation-stable joining methods [227], and neutron irradiation testing of these composites [228,229]. Multitechnique mechanical characterization studies with application of digital image correlation [230] and orchestration of round robin tests [231] have provided good understanding regarding mechanical response of SiC/SiC cladding structures. A variety of joints were demonstrated [232] and irradiated under LWR-relevant temperatures, resulting in identification of SiC-based joints as the most appropriate [233]. Neutron irradiation tests at LWR-relevant temperatures showed that the mechanical behavior of SiC/SiC does not degrade within the dose regimes applicable for LWR cladding applications (Table 3) [234,235]. Fuel–cladding chemical compatibility tests indicate no notable interaction (at least in the absence of irradiation) between SiC and urania below 1200°C [236,237].

Finally, within the same timeframe, a number of high temperature steam oxidation and quench tests were conducted on SiC-based materials to evaluate their performance under beyond DBA scenarios [39,238–240]. An example is presented in Figure 8, showing chemical vapor deposition (CVD-SiC) and NITE-SiC specimens after 1700°C steam oxidation tests. Even though the silica film on the surface is molten at this temperature, oxidation progresses far slower than in metallic cladding materials. All these tests clearly indicate that owing to its superior performance under these extreme conditions, if successfully deployed, SiC-based cladding is the most impactful ATF technology under beyond DBA scenarios.

It is worth noting that for roughly as long as SiC/SiC has been considered for LWR fuel cladding technology, it has also been deemed a potential replacement for the Zr channelbox in BWR fuel assemblies [241]. Since thermal neutron absorption is even lower in SiC than in Zr, such a transition is accompanied by an immediate advantage towards better fuel utilization in LWRs [181]. This author deems the channelbox application as the appropriate immediate deployment target for SiC/SiC in LWRs to precede SiC-based cladding deployment.

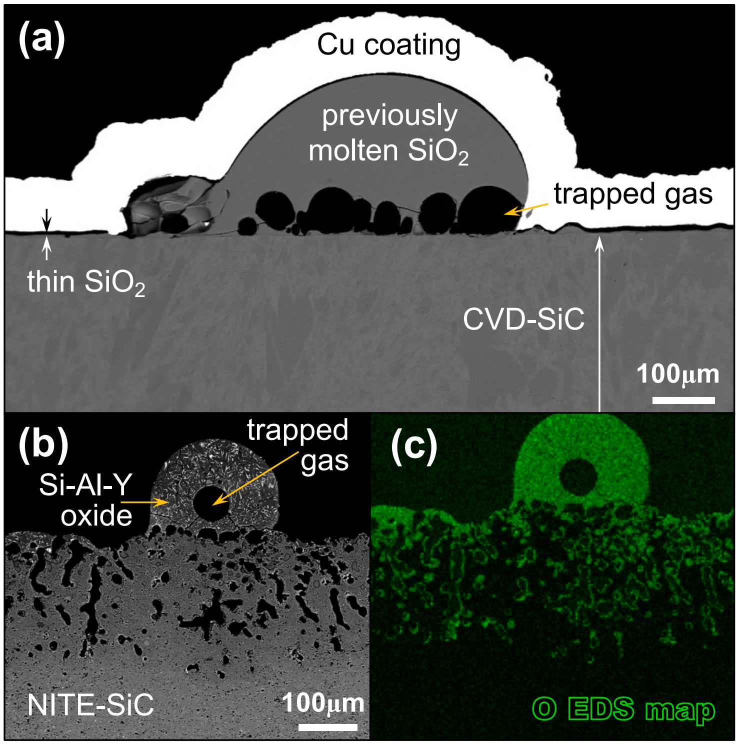

Figure 8. Scanning electron microscopy image from cross sections of a) CVD-SiC and b) NITE-SiC oxidized in 1700°C steam for 2 and 0.5 h, respectively, showing previously molten silica on the surface containing trapped reaction product gas bubbles. c) Characteristic X-ray map of O from the NITE-SiC specimen showing that unlike CVD-SiC, corrosion progresses nonuniformly and faster. Tests were conducted in flowing steam at ORNL's Severe Accident Test Station [97]. *Although not shown here, if the NITE-SiC specimens are coated with a thin layer of CVD-SiC, they experience slower oxidation and behave in a manner similar to bulk CVD-SiC* [242].

5.2. Challenges

Critical examinations of SiC/SiC technology for LWR cladding application have been codified in [243,244]. Two critical feasibility issues, namely aqueous corrosion and fuel cladding failure due to microcracking during normal operating conditions, were identified as key areas requiring further examination.

5.2.1. SiC dissolution in aqueous environments

Corrosion of SiC-based materials in high-temperature high-pressure water, henceforth referred to as hydrothermal corrosion, received attention as early as the 1980s [245], when mass loss as a function of time was reported. Studies specific to LWR coolant chemistry have emerged since the 2000s, indicating the superior behavior of high purity SiC (i.e., CVD) [246,247]. Similar to high temperature steam environments, the corrosion process is initiated with oxidation of Si in SiC to form silica while the C, except for a narrow window of conditions [248], volatilizes as CO_2, CO, or CH_4, depending on the oxygen activity in the system [249]. However, unlike the flowing steam environments where SiO_2 undergoes slow (mass-transport limited [204]) volatilization as $Si(OH)_{4(g)}$, the newly formed silica rapidly dissolves in the aqueous environment as silicic acid [250,251]. Since there exists abundant solubility of silica in high temperature water [252], and the rate of silica dissolution is much faster than the rate of its formation, the latter determines the rate-limiting step for hydrothermal corrosion of SiC. Hence the strong effect of oxygen activity in the water on the corrosion rate of SiC that was determined in recent years is not surprising [249,253]. Essentially, dissolved hydrogen reduces the oxygen activity and redox potential in the system, slowing the rate of silica formation. Hydrogen water chemistry (HWC) was always a characteristic of PWR primary loop coolant (owing to the H_2 gas in the pressurizer) [254] and is now routinely used in BWRs to reduce the oxygen activity in these systems and alleviate a host of aqueous corrosion concerns [255], primarily SCC of core internals.

A number of out-of-pile and in-pile corrosion tests to quantify the recession rate of CVD-SiC and NITE-SiC have been conducted to date and are summarized in Table 4. Similar tests on joined SiC specimens indicate that the SiC-based joining technologies that exhibit superior radiation stability are also the ones that are most corrosion resistant [256]. For a SiC-based fuel cladding, a recession rate of 0.1 mg/cm^2-month results in thickness loss of ~3.7 µm/year (assuming fully dense cladding) while ~55 kg of silica is expected to be dissolved and deposited in the primary coolant annually. During out-of-pile tests, depending on the composition of the sintering additives, the corrosion rate of NITE-SiC is ~1–2 orders of magnitude faster than CVD-SiC [257]. Radiation is known to exacerbate the hydrothermal corrosion rate of SiC due to increasing oxygen activity in the system as a result of radiolysis [258] and introduction of radiation damage in SiC that renders it a more corrosion-susceptible material [259]. Substantial dissolution during in-pile corrosion tests with NWC coolant conditions has been reported for CVI SiC/SiC specimens [260]. Utilization of HWC is known to greatly reduce the oxygen activity as radiolysis takes place [261] while also, as shown recently [262], largely mitigating the susceptibility of irradiated CVD-SiC to faster corrosion. It should be noted that even when HWC is utilized, selective migration of H to the gas phase as boiling takes place in BWR fuel bundles results in an increase in oxygen activity in the liquid phase [261].

Table 4. SiC recession rate in mg/cm^2-month from various out-of-pile and in-pile tests.

	Out-of-pile		In-pile	
	HWC	NWC	HWC	NWC
CVD-SiC	0.01–0.03 [249,253]	0.1–1.5 [246,249,253,263]	0.15–0.3 [260,264]	1–5 [260]
NITE-SiC	0.5–5 [257]	10 [257]	2–30 [265]	≥30

Only recently has systematic in-pile corrosion testing of SiC in a well-characterized water chemistry environment been initiated to produce high quality data [266]. However, to mitigate SiC dissolution entirely, the use of aqueous corrosion-resistant coatings has been proposed [267–269]. As noted earlier in Table 2, Cr, CrN, and TiN are such coatings. Preliminary results from coated SiC, neutron irradiated to 0.5 dpa, are promising, indicating that these coatings are able to accommodate swelling strain (saturating at ~0.1 dpa [203]) in the SiC substrate after 0.5

dpa and remain adherent and crack-free [270]. This is likely due to abundant irradiation creep in the coating layer.

5.2.2. Potential for radionuclide release due to microcracking

The potential for SiC/SiC fuel cladding failure due to microcracking is another area that merits deeper understanding. Note that failure in this instance is not defined by a catastrophic break in the material; rather it is characterized by formation of a pathway, likely a network of microcracks, that allows for release of radionuclides from the cladding internals to the coolant. SiC/SiC composites have been brilliantly engineered to exhibit macroscopic ductility and therefore are robust structural materials. However, the mechanism that facilitates this macroscopic ductility consists of microcracking of the SiC matrix (since matrix modulus is higher than the fiber [195], it cracks first) followed by fiber bridging and pullout. It is therefore postulated that matrix microcracking may provide a path for release of radionuclides when the fuel cladding application is considered.

While catastrophic failure in SiC/SiC composites occurs at an ultimate tensile stress (UTS) of ~200 to ~400 MPa and a total strain of ~0.5%, the microcracking in the matrix develops when proportional limit strength (PLS), manifesting at ~90 MPa (CVI) to ~200 MPa (NITE) stress or ~0.05% strain, is reached [195,231,271], Figure 9. The UTS and PLS of SiC/SiC may be described by certain statistical distribution functions such as a two-parameter Weibull distribution [195], with lower scale and shape parameters for the latter, facilitating the possibility of microcracking at stresses well below 90 MPa. In fact, the onset of microcracking has been attributed to stress levels at ~80% of PLS [272].

This modest level of stress, as shown by numerous thermomechanical analyses in recent years by various research groups [273–277], is expected to be present in SiC/SiC cladding during normal LWR fuel operation. Although the pioneering analyses [278,279] failed to capture this effect, the large swelling gradient [280] across the cladding thickness, itself a result of a large temperature gradient due to the low thermal conductivity of SiC/SiC cladding [281], presents a stress profile conspicuously distinct from that of the metal cladding. The result is that prior to pellet–cladding contact, unlike metallic cladding, the maximum tensile stress is on the inner surface of the SiC-based cladding [273]. This stress is increased further upon core shutdown [275] and PCMI [282]. Furthermore, the strong sensitivity of SiC swelling to temperature for the regime relevant to LWRs may result in large deflections when nonuniform pellet power profiles are considered [282].

The ability of SiC/SiC cladding to remain hermetic under irradiation and high heat flux is the subject of ongoing research [283]. Early experimental results appear to confirm the model predictions of high tensile stress on the inner surface of the cladding [284]. It is conceivable that microcracking initiated in the inner surface may come to rest as it propagates across the cladding thickness towards the region experiencing strong compressive stress; this hypothesis will be experimentally verified with a sensitive leak detection apparatus such as the one described in [285]. At the same time, the potential for microcracking in SiC/SiC further underlines the usefulness of an outer coating layer that may serve the dual purpose of protecting SiC from hydrothermal corrosion while serving as an additional barrier to radionuclide release.

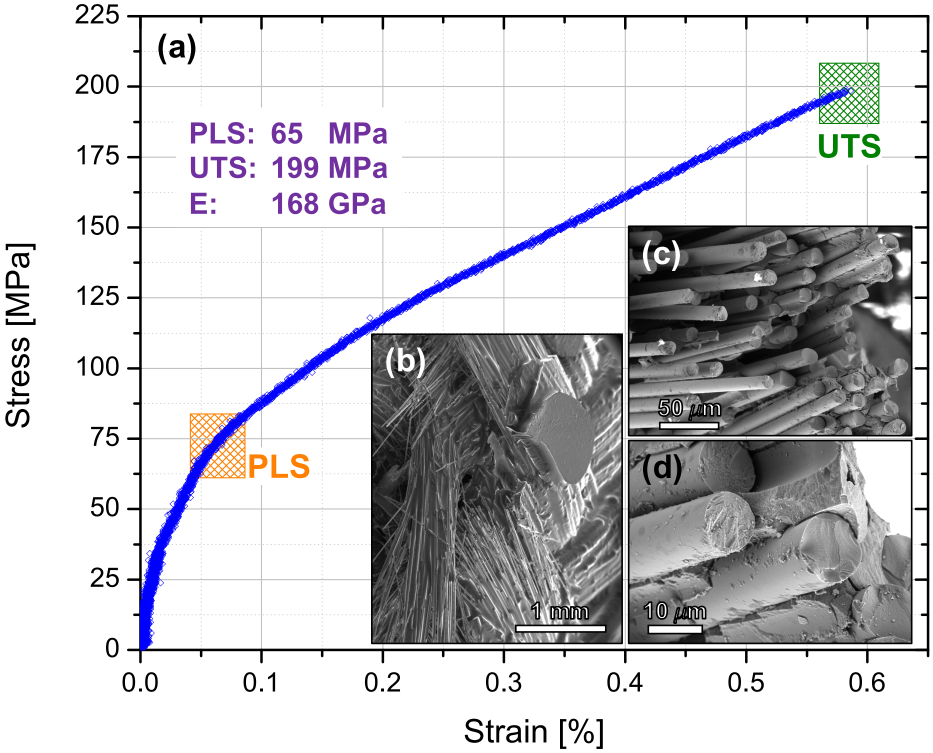

Figure 9. a) Typical stress–strain curve during axial tensile testing of a CVI SiC/SiC tube [286]. *b,c,d) Fracture surface of composite at different magnifications.*

5.2.3. Need for material standard and design architecture definition

SiC/SiC is a fundamentally different material than the various metal fuel claddings used since the inception of nuclear energy: its constituents are brittle ceramics and it is heterogeneous. Although, and as noted earlier, SiC/SiC has been engineered to exhibit pseudo-ductility, its treatment as a qualified material requires well-defined design rules. Although such rules and standards readily exist for metallic materials, they are currently being defined and developed for ceramic matrix composites [287]. This activity is of paramount importance and needs to be supported fully and broadly by and within the nuclear SiC/SiC community.

The heterogeneous nature of SiC/SiC (i.e., fiber inside the matrix) may manifest in a myriad of architectures, facilitating the opportunity to produce a composite tuned for a specific application. For instance, fully composite (woven or braided) [225], woven composite with an inner monolith [214,217,288], braided composite with an outer monolith [214], and metal–composite hybrid [289] concepts have all been produced and examined for the nuclear fuel cladding application. This implies that a unanimous understanding on an optimal architecture for SiC/SiC cladding is lacking. Defining a standard and optimal SiC/SiC fuel cladding architecture for adoption across the nuclear community should be deemed a high priority.

5.2.4. Need for elucidation of beyond DBA behavior

SiC undergoes oxidation–volatilization in flowing steam, best described by paralinear kinetics [290,291]. Furthermore, high steam pressures, which may be present during severe accidents, may increase the oxidation rate of the cladding by causing pore formation in the protective silica [39,292,293]. Recent studies indicate that pore formation can manifest even at atmospheric pressures when a high partial pressure of oxidation product gases (i.e., H_2, CO, $Si(OH)_4$) are present in the system [294]. Except for Avincola et al. [239], the temperature range of all other studies has been limited to ≤ 1700°C. Additional separate effects and integral tests are necessary to examine the behavior of SiC/SiC cladding technologies under beyond DBA scenarios.

6. Discussion

This section presents arguments in support of full development and deployment of ATF cladding concepts. The technical challenges discussed in the previous sections are not deemed insurmountable with focused R&D. Also, a brief attempt at presenting the fuel cycle cost is made here with the intention of showing that the overall benefits of ATF cladding concepts outweigh their higher cost. Finally, the need for additional R&D to generate data in support of informed deployment of these technologies is highlighted.

6.1. Zr is approaching its performance limit

It is clear to the readers of this journal that a broad range of advanced reactor concepts [295] are under active development across the globe. These advanced concepts aim to offer higher performance (efficiency and resource utilization) and enhanced safety, and thereby better economics than the current fleet of LWRs. Almost none of these advanced reactor concepts intend to utilize Zr-based alloys as fuel cladding or as any structural component. Aside from its low strength at elevated temperatures [296], as an HCP (hexagonal close-packed) metal, application of Zr to high dose, >30 dpa, and high temperature, >450°C, is not viable given its susceptibility to runaway irradiation growth [44].

Regarding the LWR application of Zr, any substantial further gains in fuel burnup are limited by zirconium's inherent affinity for hydrogen [8]. Material-specific challenges with Zr-based alloys under beyond DBA scenarios were discussed in Section 1.3, and real limitations under DBAs have been previously elaborated upon in [5]. The susceptibility of Zr-based alloys to severe degradation during post-CHF events is well-documented [297,298], imposing strict limits to core loading possibilities as well as the reactor's operational flexibility.

6.2. Partial gains provide additional coping time

In Section 1.3 the stated motivation for transitioning away from Zr-based alloys to ATF cladding materials is to reduce the rate of heat generation, Q in Equation (1), in the core. Note that this transition does not resolve the ultimate susceptibility of LWR cores with active safety systems to degradation under beyond DBAs. If the first term on the right-hand side of Equation (1), which represents heat removal from the core, is smaller than Q, the temperature in the core will rise. When loss of cooling capability is severe, adiabatic conditions best describe core temperature evolution. Under such a scenario, a smaller Q only delays the onset of core degradation. However, this smaller Q will positively impact accident progression in the core in two important ways.

First, additional coping time will be available to interfere with the core and to provide cooling, Figure 1. Depending on the accident scenario and the type of ATF cladding, this additional coping time will be on the order of minutes to tens of hours [189]. Although not substantial, the additional time may be exploited to facilitate injecting coolant to the core to mitigate the accident's progression. Given the physics of a LWR core, none of the ATF cladding concepts allow for a "walkaway safe" core. However, their utilization will delay the core's degradation processes, by prolonging the period during which active safety systems can be brought on line to cool the core.

Second, the rate and total magnitude of heat and hydrogen generated in the core are smaller [28] for ATF cladding concepts because their oxidation rate is lower. This implies that the threshold cooling capability necessary to stabilize the temperatures or cool the core is lower. This highlights the importance of partial cooling capability and the lower burden on ECCS capacity.

6.3. ATF cladding should not be deemed cost prohibitive

LWR fuel constitutes ~20% of nuclear electricity generation cost today, Figure 10. The fuel cost itself is dominated (>75%) by the cost of uranium, its conversion, and enrichment (note that SWU [separative work unit] prices have dropped in recent years to their lowest levels in decades [299], lowering the fuel price). At roughly \$30/m Zr-based cladding itself constitutes <3% of the fuel assembly cost, Table 5. In the case of coated Zr-based and SiC/SiC cladding materials, where thermal neutron absorption in the cladding is negligible [79], only the remaining fuel fabrication cost should be considered. Coating deposition on the surface of the cladding is not expected to increase the fuel cost notably. In the case of SiC-based cladding, the cladding cost today is high (>\$1000 per tube), dominated by the nuclear-grade (Gen III) SiC fibers at ~\$15,000/kg. These fibers are ~50× more expensive than the early generation ceramic-grade fibers. The current price of certain Gen III fibers does not necessarily reflect their actual production cost and is a result of low demand due to lack of large industrial application (much like the Zr price in its early days was too high). Once industrial demand for Gen III SiC fibers takes off, the fiber price is expected to drop greatly. Although the cladding cost itself is expected to be less compared to Zr-based cladding, the higher thermal neutron absorption in FeCrAl will require additional fissile loading (more uranium and enrichment) that will result in an additional cost of the fuel assembly on the order of 15–25% [5,131].

Note that in all cases in Table 5, the fuel pellet is considered to consist of urania that is stable in the highly oxidizing LWR coolant environments, whether high pressure water during normal operation [300,301] or steam to >2400°C under accidents [76,302]. Urania remains stable since H_2 is always present at very small quantities, retarding its further oxidation to U_3O_8. High density monolithic non-oxide fuels (>9.5 gU/cm^3), as they manifest today, are not considered safe for LWR application since they are highly reactive in O_2 and H_2O environments of >400°C [303–305], defeating the motivation discussed in Section 1.3.

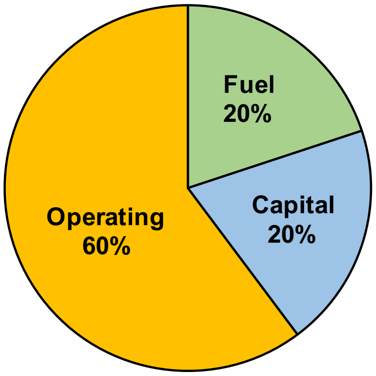

Figure 10. Fuel, operating, and capital costs as a fraction of the average total generating cost in 2016 for US-generated nuclear electricity, $34 per MWh [306,307].

Table 5. Price estimate in k$ for a PWR fuel bundle with various cladding materials (assuming the same fabrication cost).

	Cladding cost per bundle	Fuel pellet cost per bundle	Fabrication cost	Total cost	Difference from base case (%)
Zr-based (base case)	30	950	140	1120	-
Coated Zr-based	<40	950	140	<1130	<1
FeCrAl	<30	1140	140	≤1310	≤17
SiC/SiC	>300*	950	140	≥1390	≥24

*The cost is dominated by fiber and may be greatly reduced once fibers are produced in large volumes.

In today’s electricity market, at least in the deregulated regions of the United States, price competition is fierce. Most prominently, owing to hydraulic fracturing, natural gas is available in abundance; priced at <$3/MMBtu in the United States, 2–3 times cheaper than in the European Union and Asia, it is a cheap source of electricity. Given this pressure, there exists a strong sensitivity to any increase in nuclear fuel price. However, there are a number of opportunities to reduce the overall fuel cycle cost [308]. These opportunities are real and include the following:

- Expanded fuel operational window (higher CHF limits and resistance to degradation during post-CHF events as well as the absence of a cladding ductility loss mechanism due to hydride precipitates)

- Increased fuel burnup, no longer limited by cladding performance (although >5% ^{235}U enrichment will likely be necessary)
- Increased limits on peak cladding temperature and exposure duration under DBA scenarios
- Application of risk-informed categorization and treatment of structures, systems, and components for nuclear power reactors (i.e., 10 CFR 50.69) and regulatory guidelines, to change operational models and reduce maintenance costs [309]
- Enhanced back-end performance of used fuel and less susceptibility to degradation during storage and transport

Most important, the nuclear industry, unlike other heavily regulated industries (e.g., aerospace), has failed to adopt advanced materials over the past decades [310]. This failure is a recipe for obsolescence, and it is imperative that the community moves forward with adoption of ATF cladding materials to break this trend. Also, these materials are often the same ones being considered for advanced (non-LWR) systems, and their successful deployment in LWRs will greatly facilitate their adoption in future nuclear energy systems.

6.4. Experimental data remains a critical need

Large datasets of high quality are necessary to support predictive understanding of fuel behavior under all the scenarios listed in Table 1. This body of data, as it pertains to Zr-based alloys, was collected over the decades and needs to be replicated for the ATF cladding technologies. Note that over two-thirds of the references cited in this article were published since the Fukushima Daiichi accidents in 2011. This highlights the level of effort directed at developing these technologies internationally and the resulting technical output from those studies. However, the need for complete datasets remains critical, as they are key facilitators of furthering the technology readiness level for these advanced fuel systems [311]. Particular focus should be placed on in-pile testing of these fuel concepts in prototypical environments [312] as well as experimental data to inform fuel degradation behavior during severe accidents [313].

7. Summary

The motivation for developing alternative fuel cladding materials with significantly enhanced oxidation resistance to replace Zr-based alloys in LWRs is to delay and reduce the heat and hydrogen generated during severe accidents as a result of cladding oxidation. Three leading concepts under active development are Cr-coated Zr-based cladding, FeCrAl cladding, and SiC/SiC cladding, with improved severe accident performance in that order. A review of development status, challenges, and data gaps for these concepts shows promise without identifying any insurmountable technical challenges. Additional R&D in key areas and generation of high quality experimental data to explain their properties and integral performance behavior are necessary to further the technology readiness level of these cladding concepts toward commercial-scale deployment and realization of their safety benefits. Full development and adoption of ATF cladding technologies will likely improve the economics of the overall fuel cycle and will showcase the nuclear industry as adopting new materials and technologies, paving the way for other advanced nuclear systems.

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