36th Annual Conference of the Canadian Nuclear Society and 40th Annual CNS/CNA Student Conference
Toronto Marriott Downtown Eaton Centre Hotel, Toronto, ON, Canada, June 19-22, 2016

Advances in Materials Evaluation for the Canadian SCWR Core Concept

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Abstract

Canada has, since 2007, undertaken research to develop a conceptual design of a pressure-tube based supercritical water-cooled reactor (SCWR). With an outlet temperature of 625 degree C and a coolant pressure of 25 MPa, this concept requires fuel-cladding materials that can sustain high-temperature and high-pressure in-core conditions. An overview of the key aspects of the design related to in-core materials and the recent development of materials assessment of this program are presented in this paper. Much progress has been made in selection and testing of candidate alloys, and the more rewarding scientific outcome of the program is the fact that significant amount of knowledge has been gained of the mechanisms and factors of key microscopic processes taking place under SCWR in-core conditions. Further materials R&D work should focus on improving the resistance to stress corrosion cracking (SCC) and irradiation-assisted SCC, high-temperature strength and ductility as well as microstructural stability.

1. The Canadian SCWR

The supercritical water-cooled reactor (SCWR) is one of the six advanced reactor systems developed in the collaborative effort of the Generation IV Forum. While Japan, the European Union and China are working on pressure-vessel SCWR core concepts, Canada is focusing on a pressure-tube SCWR concept [1, 2]. Rather than using a large pressure vessel as the pressure boundary, individual pressure tubes (336 in the latest design [1]), held in a heavy-water filled calandria vessel, serve as the pressure boundaries and the main conduits of coolant (Figure 1). In this design the heavy water moderator in the calandria is maintained at a low pressure and temperature (~0.3 MPa, ~100 °C) and such conditions allow zirconium-alloy pressure tubes to be used; by using improved Zr alloys for the pressure tubes, the strength needed to contain the supercritical water (SCW) coolant at 25 MPa is provided without sacrificing neutron economy. In order to limit the temperature of the pressure tube, an encapsulated zirconia insulator is installed in the fuel assembly to insulate the pressure tube from the SCW coolant.

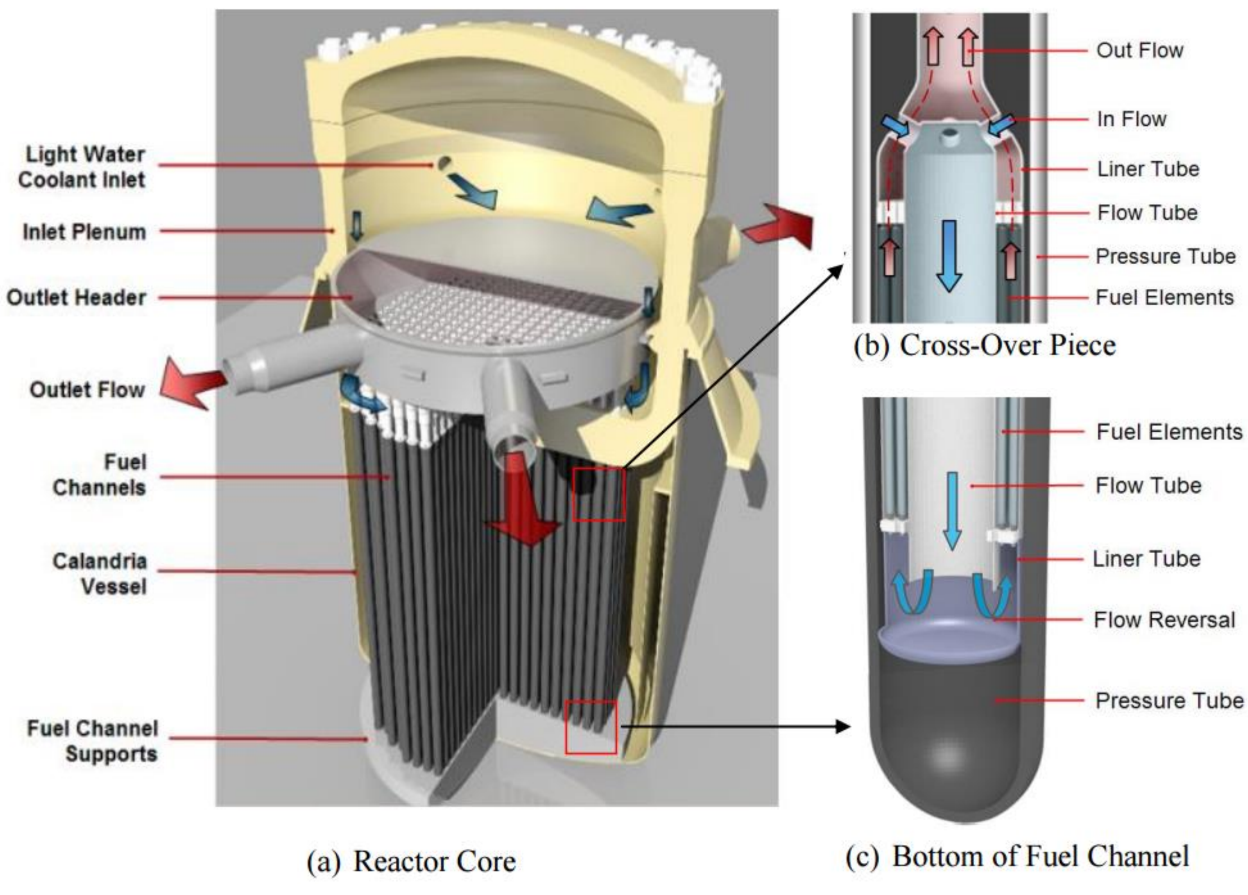

Figure 1: Schematic of the Canadian SCWR core concept and fuel channel [1].

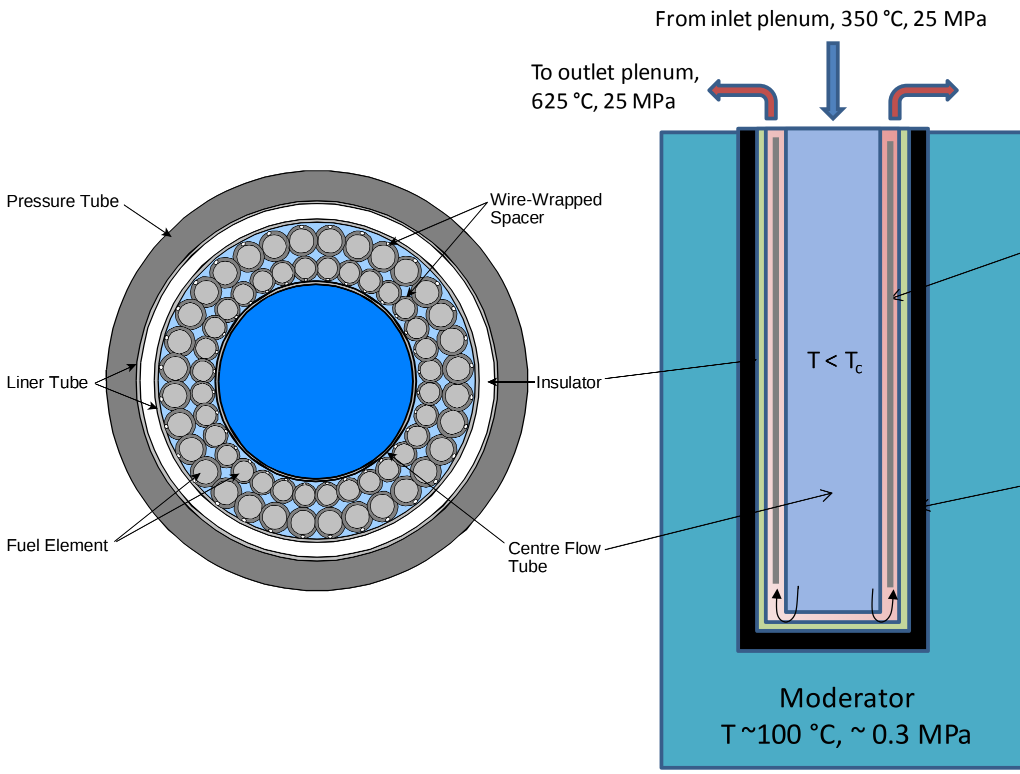

Figure 2. Canadian SCWR fuel channel/fuel assembly concept cross-section (left) and vertical flow path (right). The vertical flow path has been simplified to highlight specific aspects of the concept that relate to materials performance [1].

Figure 2 illustrates the details of the fuel channel and fuel assembly concepts, in supplement of the details shown in Figures 1a and 1c. In Figure 2, the full-length of the channel is not shown, as the intention is to show the gap and the relative layout between the pressure tube and the fuel bundle assembly [1, 2]. The pressure tubes are connected to the tubesheet in a leak-tight fashion; the fuel channel also has a guide tube that extends the fuel channel into the inlet plenum and an expansion bellows connecting the guide tube to the outlet header space.

As shown in Figure 2, the fuel assembly consists of a central flow tube, a ceramic insulator on the outside, an inner liner tube separating the fuel and the insulator, an outer liner encasing the insulator, and a two-ring fuel-element configuration [1, 2].

Above the fuel assembly, each channel has a cross-over mechanism (Figure 1b) that allows the inlet coolant to enter the central flow tube. The coolant first travels downwards through the central flow tube and after reaching the bottom, going upwards through the fuel rods, picking up the fission heat along the way, as shown in Figures 1b, 1c and 2.

The entire fuel assembly is replaced during refueling. As a result, all the components in the fuel assembly are subject to high temperature, high pressure and irradiation for only about 3 years, which significantly reduces the materials requirements; this is a major advantage of the Canadian SCWR concept. For other components that will be exposed to a temperature ranging from 350 to 625 °C and low neutron flux, materials developed for use in fossil-fired SCW power plants can be used as good references. One difference, though, will be the effects of the hydrogen, oxygen and hydrogen peroxide generated in the coolant by water radiolysis as it passes through the core [3].

As discussed in a recent summary paper [4], the fuel cladding will experience the most demanding conditions of all the in-core components of the main reactor assembly, in terms of temperature, stress load, irradiation and corrosion attack by general corrosion and stress-corrosion. As the peak surface temperature of the cladding can reach as high as 800 °C at certain locations along the fuel assembly, high-temperature mechanical strength is another concern; this is the reason why nickel alloys and austenitic stainless steels are preferred.

The use of stainless steels as a high-temperature fuel-cladding is not new. For example, 20Cr25Ni type stainless steel fuel cladding was used in the Commercial Advanced Gas-Cooled Reactors that were operational in the UK in the late 1970s and 1980s. With an outlet coolant (CO_2) gas temperature as high as 640 °C, the cladding temperature in the CAGRs was well over 700 °C in the hot sections of the fuel assembly. The use of SCW as a coolant leads to concerns related to corrosion, environmentally-assisted cracking, and water chemistry control [5, 6], similar to issues encountered in the current generation of water-cooled (WCRs). Because of the higher temperature and higher coolant pressure in the SCWR, some in-core material degradation mechanisms found in WCRs will be accelerated, including corrosion, stress-corrosion cracking, irradiation-induced microstructural degradation, and creep. In selection and evaluation of the cladding alloys for the Canadian SCWR, these aspects have been the focus of materials research effort. Recent advances from SCWR fuel cladding research are presented in this paper.

2. Assessment of fuel-cladding alloys

Zr alloys, currently used in Gen III LWRs, are not suitable for use as an SCWR fuel cladding due to their high corrosion and creep rates. Canada selected five candidate alloys for a detailed evaluation as potential fuel cladding materials: 310 SS, 347 SS, Alloy 800H, Alloy 625 and Alloy 214. Oxide-dispersion-strengthening (ODS) of ferritic and austenitic alloys can lead to much improved strength at higher temperature, as well as corrosion and irradiation resistance, and therefore the development of this class of materials is still pursued, as discussed in the paper by Kuyucak et al [7] of this conference proceedings.

2.1 Corrosion

Presently data for the corrosion of alloys under SCWR conditions exist for more than 100 alloys [8, 9]. Figure 3 compares the corrosion (measured by weight gain) of Zr alloys, low-Cr ferritic steels, austenitic stainless steels and a Ni alloy [10].

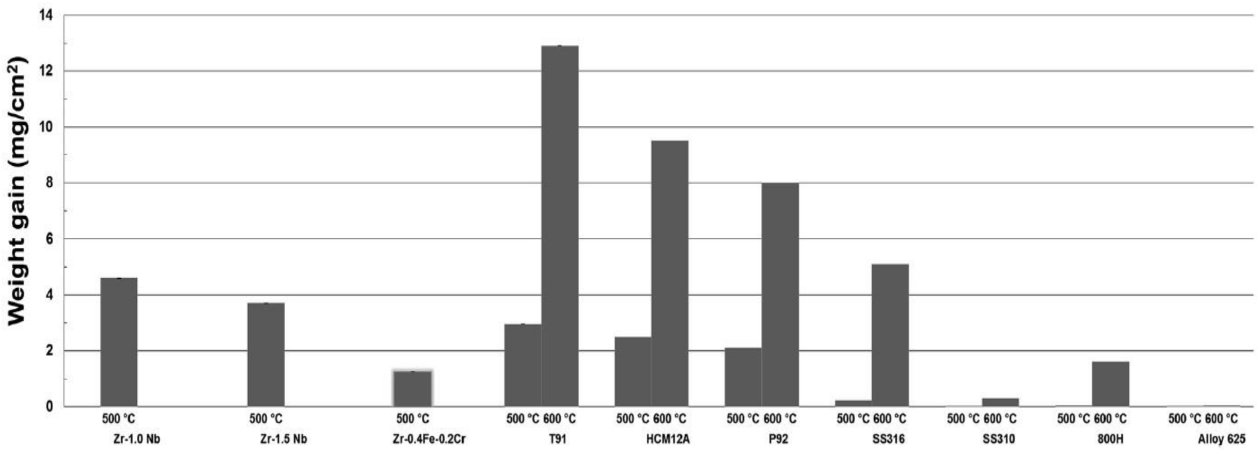

Figure 3 Example of recent studies of the corrosion of different alloys in SCW [10].

Considerable reactor operating experience exists on the use of stainless steel fuel cladding in BWRs and PWRs [11]. Russia operated two pressure-tube type reactors with separate boiling and superheated steam channels (up to 550 °C and 8 MPa) at Beloyarsk NPP between 1964 and 1989, using the stainless steels Kh18Ni10T and EI-847 in-core [12]. General corrosion was not reported to be a cause of failures.

It should be noted that significant scatter is sometimes observed when corrosion data for the same alloy under nominally identical test conditions are compared. To investigate the reproducibility of corrosion rates, an international round-robin test was carried out by seven organizations in GIF member countries [13]. Three alloys (800H, 310, 08H18Ni10T) were tested for 500 hours in SCW at 550 °C and 25 MPa with 8 mg/kg dissolved oxygen, and a surprisingly large degree of scatter, typically greater than 100% of the average weight gain, was seen for each of the three alloys when data from different labs were compared [13]. The variation was attributed to differences between the various test facilities, such as flow rate. It was concluded that while the absolute value of weight gain data should be treated with some caution, the relative ranking of the weight gain measured by different groups is reliable.

Despite the observed variability, the general trend of the weight gain data for most alloys is found to follow a generic functional form [6, 14]:

$$WG(t,T) = a \cdot T + b \cdot exp\left(\frac{-E_a}{RT}\right) \cdot t^x \qquad \text{Equation (1)}$$

with a and b being empirically-determined constants, E_a the activation energy, T the temperature (in K), t the exposure time, R the gas constant, and x ranging from 0 to 1. Measurements of a, b, and E_a enable extrapolation of the weight gain data to obtain the corrosion penetration at the different cladding temperature at the cladding design lifetime.

The estimated amounts of corrosion of 310 SS, 347 SS, Alloy 800H, Alloy 625 using Equation (1) and x=1 are shown in Figure 4. In the conceptual design [1], the minimum fuel-cladding thickness is about 0.40 mm, a thickness enough to avoid ridging during service. Therefore a corrosion allowance of 200 micrometers would be available if the cladding was fabricated with an initial thickness of 0.60

mm. As shown in Figure 4, all of four alloys meet the specified performance criterion, and general corrosion resistance should not be an issue for the alloys considered.

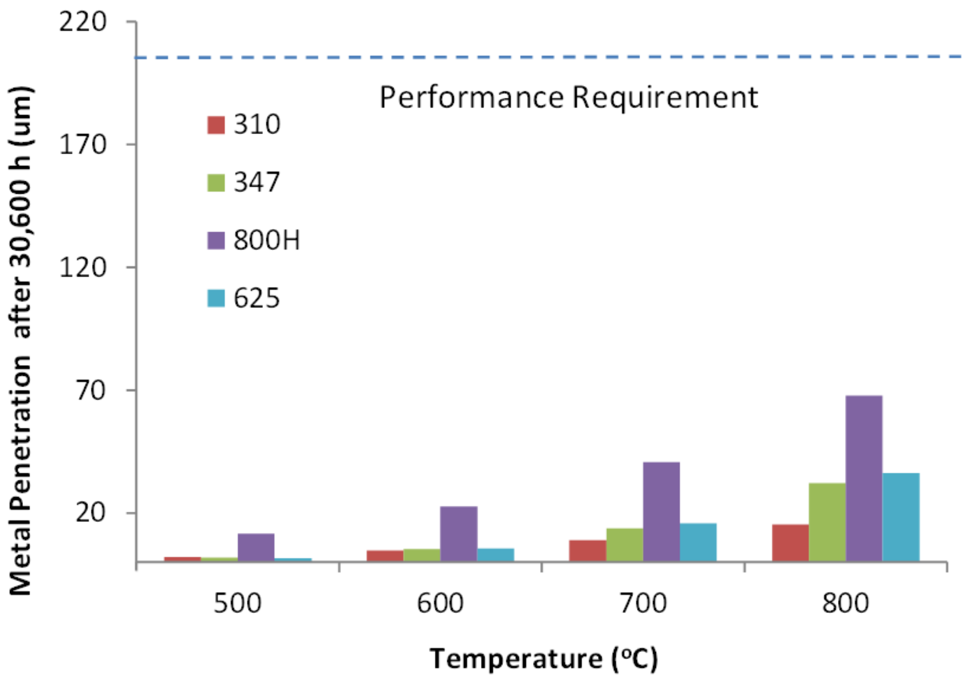

Figure 4. Estimated amount of corrosion for 310, 347, 800H and 625 at various temperatures relevant to the Canadian SCWR fuel cladding after three fuel cycles [6, 14].

2.2 SCC and IASCC:

Of the five alloys considered for the Canadian SCWR cladding, Alloy 347, 310 [16] and 800H all showed SCC in conventional strain-to-failure SSRT tests [4, 15]. SSRT testing of Alloy 625 in de-aerated SCW between 400-550 °C [16] also showed intergranular cracking at all temperatures. The average crack length increased with increasing temperature and the cracking was more extensive than that found in austenitic alloys. Was et al. [17] reported SCC growth rates between $3x10^{-9}$ and $3x10^{-8}$ mm/s for this temperature range based on measurements of the crack depth and total test time.

A key controlling parameter in the SCC susceptibility of austenitic stainless steels in SCW is the level of strain applied on a macroscopic level. Mechanistically the cracking is of intergranular mode and the data from the SCC tests using SSRT methods suggested that there is a threshold level of strain at the grain boundary below which SCC initiation would be difficult [4].

As only 1.5% total deformation is expected over the lifetime of the Canadian SCWR fuel cladding, in recent SSRT tests for the Canadian SCWR program straining was stopped after the engineering strain reached 5%. All samples without prior tensile cold-working strained to 5% in SCW at 500 °C showed no cracking in these “5% strain” tests. This finding was confirmed for Alloys 310S and 800H in tests at 625 °C; in these latter tests the samples were strained to 5% in SCW and then held at that strain level for at least 300 h [4, 15]. In SSRT tests in SCW at 500 °C, cold-worked 310S and 800H that were seen to have developed deep cracks that propagated across the deformation bands in the microstructure, a characteristic similar to that observed in SCC in cold-worked austenitic steels under light water reactor (LWR) conditions [18-20].

As irradiation of sufficient dose can induce the formation of deformation channels [21], there is a similitude of impact from cold-working and irradiation in terms of this 'localization' phenomenon. Work is needed to determine if after ~10 dpa of irradiation damage dose the cladding alloy can develop cracks of critical size within the strain range expected in the conceptual design.

Radiation induced segregation (RIS) in stainless steels can lead to depletion of Cr in grain boundaries, a known cause of intergranular cracking in stainless steels [22]. This chemical effect of the irradiation on the candidate alloys, and the implications for corrosion and SCC, is being studied in the Canadian SCWR program [23].

There are so far few studies of SCC tests of irradiated materials under SCW conditions. Zhou et al., [24] reported that SSRT results for Alloy 800H, including test samples irradiated to 7 dpa. It showed that, at a strain rate of $3x10^{-7}s^{-1}$ and with the test alloys pulled to failure, the depth of cracks was three times greater in the irradiated samples. Although 7 dpa of damage is close to the expected end-of-service dose for the fuel cladding in the Canadian SCWR concept, the strain levels applied in these tests far exceeded the expected range of interest. Further work in this area is needed.

2.3 Creep

A large mechanical property database [25] has been developed in an effort to select and assess promising commercial alloys and new alloys under development. It contains data on ferritic-martensitic (F/M) steel, austenitic stainless steels, Ni-Fe based alloys, Ni-based alloys, and oxide-dispersion-strengthened (ODS) alloys, the latter including both commercial and new experimental alloys. Comparisons were made for given alloy types of their tensile strengths and creep rupture life (i.e., the Wilshire–Scharning model for F/M steels, and Larson–Miller Parameter for other types). The irradiation effects on mechanical properties of the candidate alloys were also surveyed [26]. Figure 5 shows predicted creep rupture stress for the five alloys after 30,000 hours in the high temperature regime relevant to SCWR. Some data were collected from published literature while the results of commercial SS 347H and SS 310S were confirmed by in-house experiments. The creep rupture strengths at 1073 K (800 °C) and 30,000 h of 347H, 800H and 310S were too low for a free-standing fuel cladding concept (as used by the EU and Japanese SCWR concepts), and thus a collapsible cladding was selected; the Zr-alloy fuel cladding used in CANDU reactors is also collapsible.

In addition to thermal creep (time-dependent deformation under temperature and stress only), alloys under irradiation can experience irradiation creep as well as volumetric expansion-induced deformation. Irradiation creep can increase the creep rate over thermal creep or induce creep in temperature regimes where thermal creep is negligible [27]. It is therefore important to find ways to improve the alloys that have otherwise sufficient corrosion and SCC resistance. During initial screening of materials, ferritic steels were eliminated from further consideration for two reasons: (1) the corrosion rate in SCW for ferritic alloys with Cr content less than 19% is too high in the conventional wrought alloys and (2) when the Cr content is above this value the formation of a close-packed tetragonal sigma phase creates brittleness and loss of fracture toughness. Mechanical alloying is therefore receiving much attention as a way of improving resistance to irradiation and thermal degradation. Some promising creep test data have been obtained for the Fe-Cr ODS alloys prepared in-house at CanmetMaterials. The best batch has been subject to 80 MPa load at 750 °C for six months, no sign of failing can be seen at the present time [7].

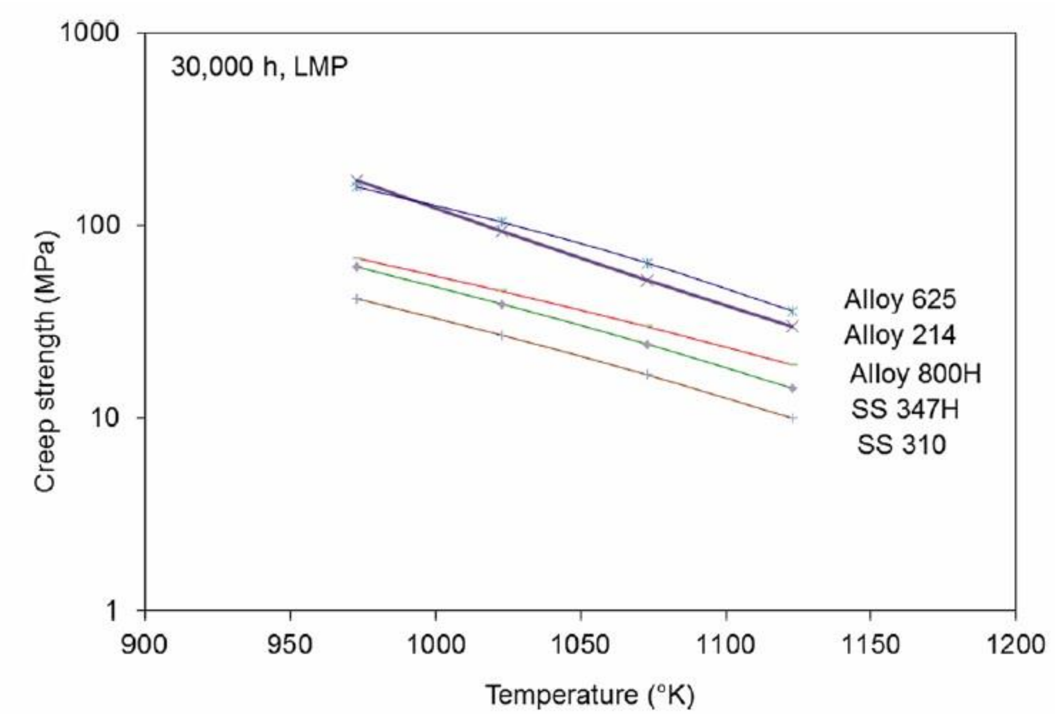

Figure 5. Creep rupture stresses for 30,000 h of the five candidate alloys [25]

2.4 Irradiation damage

Formation of helium bubbles along grain boundaries, a well-known problem in Ni-containing alloys, is a process that can unfavorably affect the creep performance of alloys, ductility and fracture toughness [28]. Ductility, as shown in elongation, may significantly decrease as a result of irradiation at temperatures above ~600 °C. Cold-worked stainless steels show some softening effects due to thermally-activated recovery. The best example in this regard is perhaps the paper by Thiele et al. [29], who reported the ductility of Alloy 800 (which has smaller grains than the high-temperature version, 800H) and other materials at various temperatures after irradiation at elevated temperatures to a thermal fluence of 1.1×10^{21} $n \cdot cm^{-2}$, a dose of about 1.4 dpa. At 700 °C most of the alloys studied showed elongation in the high single digits or to just above 10%. At 850 °C most materials showed elongation below 5% [29]. In terms of the Canadian SCWR concept, it remains to be determined as to how much post-collapse ductility is required for the fuel-cladding to survive 30,000 h. The expectation is that in the case of excessive hydrostatic loading the fuel cladding would collapse relatively early during operation [1,2]; at that stage there would have been little accumulation of irradiation dose and the material should essentially behave as the un-irradiated alloys during collapse.

Void swelling is another mode of irradiation damage. Austenitic stainless steels and nickel-based alloys are known to be susceptible to void swelling [30, 31]. Alloys 800H and 625 are estimated to be the most resistant to void swelling, followed by Alloy 214. Type 310 stainless steel is considered at risk for void swelling. Type 347 stainless steel is predicted to swell over the life of the fuel cladding [14].

3 Future Research

The overall outcome of evaluation of five alloys, as presented to the Check and Review panel of SCWR experts in October 2015, has been summarized using a color-coded score card, recaptured here as Table I. While none of the candidates can meet all the expected performance requirements for the Canadian SCWR fuel cladding, there are sufficient data available to suggest that Alloy 800H, 310s and Alloy 625 are promising for further, more detailed, material testing including in-pile testing.

While much progress has been made in selection and testing of candidate alloys, an equally rewarding technical outcome of the program is the fact that significant amount of knowledge has been gained regarding the mechanisms and factors of key microscopic processes taking place under conditions relevant to the SCWR claddings, i.e., under high-temperature and pressure and with intense irradiation. There remain many gaps that need to be addressed in a future validation phase of the program, for example, the effect of flow rate and water radiolysis on general corrosion, the effects of surface finish, and the effects of irradiation. In terms of SCC, there are indications that a threshold strain level exist for a given microstructure below which crack initiation would be difficult [4]. As irradiation can cause accumulation of localized deformation, especially at grain boundaries, the effects of irradiation on this SCC 'strain-threshold' should be investigated for the final cladding material. Understanding the microstructural evolution of irradiated alloys, using micro-beam analyses or computational tools [32], is essential for not only developing improved cladding materials but also for future service- integrity assessment. A major gap is the lack of suitable irradiation facilities.

Recent creep test data of a new ODS alloy developed in-house show a real feasibility to improve mechanical performance through mechanical alloying [7]. Optimization of the alloy composition for corrosion and irradiation resistance is the next step in further experimental development.

Table I. Results of overall assessment of candidate alloys [14]

Alloy	Corrosion	Oxide thickness	SCC	IASCC	Void Swelling	Creep rupture	Ductility	Strength
347								
310S								
800H								
625								
214								

GREEN : Available data suggest that alloy **meets** performance criteria under all conditions expected. (Alloys are assumed to be in fully annealed condition)
YELLOW : Some (or all) available data suggest that alloy **may not meet** performance criteria under some conditions expected

RED : Some (or all) available data suggest that alloy **will not meet** the performance criteria under some conditions expected
GREY : **Insufficient data available** to make an informed guess as to the behavior

Acknowledgement

The authors wish to acknowledge the multi-year funding support by the National Gen IV program managed by Office of Energy Research and Development (OERD), under the leadership of Shannon Quinn, Ramesh Sadhanker (on assignment from the former AECL) and Daniel Brady. Significant contributions and collaborations from CNL colleagues, particularly Laurence Leung, Metin Yetisir, Jeremy Pencer and Matt Edwards, are acknowledged with appreciation. Input from many CMAT staff, particularly in the area of mechanical and corrosion testing, materials processing and characterization, computation, over the past 8 years has been valuable in achieving the planned deliverables for both the national program and for the Generation IV International Forum (GIF). Many of the corrosion and materials results are achieved through collaboration with University of Alberta, Carleton University, Queens, McMaster, UNB, and University of Toronto. Among the GIF members, VTT (Finland), EU Joint Research Centre (JRC-IET), Hitachi Research Lab (Japan), also contributed to corrosion and SCC work.

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