PECULIARITIES OF STAINLESS STEELS APPLICATION AS ATF IN VVER'S

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ABSTRACT

Analysis of the peculiarities of steels application as ATF in VVERs was made. Three classes of steels are under consideration now. Ferritic-martensitic steels (FeCrAl alloy included), austenitic steels as well as Ni-Cr steels. Each type of steel has its advantages and drawbacks and was analyzed using the following ATF criteria: melting temperature, neutron penalty, change in shape, radiation induced hardening, stress corrosion cracking and high temperature hardening. For each class of steel some modifications were suggested to improve their properties.

1. Introduction

The majority of nuclear reactors currently accommodate traditional Zr clad/UO_2 fuel design, which has an excellent performance record for normal operation. However, a significant hydrogen production, resulting from high temperature Zr/steam interaction requires the design improvement developing more accident tolerant fuels (ATFs)-clad systems. One of them is an application of stainless steel claddings of various types, particularly FeCrAl cladding. Russia accepted a concept to use at the first stage as ATF already developed steels, particularly 42HNM alloy, which revealed an excellent behavior in water-cooled and fast reactors.

Steels advantages in comparison with Zr clad alloys are well-known [1-5]. First of all, it is high corrosion resistance in accidence hence, low hydrogen producing. There are a lot of experiences of its application in various types of reactors, water-cooled included and existent industrial scale technology. Moreover, steel claddings allow fabricating any complicated shape of cladding (Fig. 1) [6, 7]. At the same time some disadvantages exist that can create difficulties in its implementation in LWRs. Mainly, they are noticeable neutron capture, stress corrosion cracking (for austenitic steels) and radiation induced hardening (for ferritic steels) as well as tritium release to the coolant [3-5].

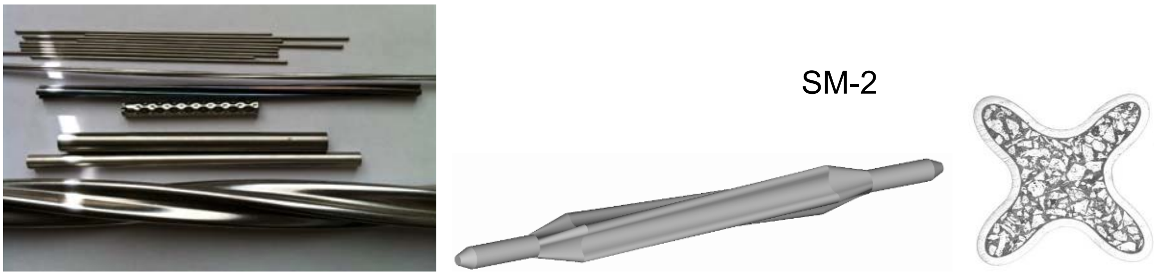


Fig. 1. Complicated shape of steel cladding to increase the reactor power [6, 7]

In this paper consideration is given for peculiarity of stainless steels application as ATF in LWRs as well as proposes ways of their modifications to improve properties.

2. Consideration of various types of steel concepts

Briefly various types of steels as candidates for ATF can be conditionally divided into 3 groups:

1. Ferritic-martensitic steels
2. Austenitic steels
3. Ni-Cr steels

It should be noted that the majority of steels have qualitatively low oxidation rates at accidence in comparison with Zr claddings. Of course, corrosion resistance differs for various steel types that require composition modification to improve steels properties. At the same time steels corrosion resistance in high temperature steam is an order of magnitude better than that of zirconium and satisfies ATF requirements. Moreover the beginning of leakage and fuel element damage occurs by inner gas pressure already at 800^0C. Hence further modification of the steels corrosion resistance should not be at the expanse of the other steels properties.

First group is mostly represented by FeCrAl alloys. Its composition varies by Cr and Al content, usually Cr 12-20% and Al 3-8%. Small amount of Mo and some other elements can be also added. It is a well-known fact that the more alloying elements (mostly Cr and Al) added to steels, the more corrosion resistance and tensile strength is resulted and respectively less ductility. But ductility is quite enough for tube fabrication and satisfies BOL conditions. However, the situation changes during irradiation. The experience of using this class of steels with a high chromium and aluminium content in Russian design fast reactors shows a drastically loss of ductility upon irradiation.

Conditionally austenitic steels can be divided into 3 subgroups – steels having relatively low Ni content (15-19%) average content (24-26%) and high content (39-46%). Nickel improves corrosion resistance and in case of complex doping high temperature strength. At the same time it has large neutron capture and can produce a small amount of He at high burn-ups and neutron fluence. Its application experience at normal conditions in Russian water-cooled reactors has shown that all austenitic steels, regardless of the nickel content, are all sensitive to stress corrosion cracking [7-14].

The third group refers to so called Ni-Cr steels. Correctly they should not be named as steels since they have minimum or nothing Fe content in their composition. First of all, it is 42HNM steel developed in Russia as well as foreign 625 and 690 (Tab. 1). Their high-temperature strength as well as corrosion resistance are of maximum value among the other steel types. In comparison with Fe-Cr-Al they retain most of their high properties after irradiation. But large neutron capture and some other disadvantages (see chapter 4) also create some restrictions to their usage. Nevertheless 42HNM alloy is the main candidate for implementation as ATF as we have a great successful experience of its usage in water-cooled small reactors [10-12].

Steel	**Element content % wt.**				
	Ni	**Cr**	Fe	**Mo**	**Co+Ta**
42HNM	base	42	-	1	-
625	base	21.5	4	9	3.5
690	base	29	9	-	-

Tab 1: Chemical composition of Ni-Cr alloys

As can be seen from table 1 the principal difference between the Russian alloy 42HNM (bochvaloy) from its foreign analogues is the lack of iron in its composition. Perhaps it is for this reason, formally referring to austenitic steels with a FCC lattice this alloy is not subjected to stress corrosion cracking, which makes it possible to widely use in VVER's [10-12].

3. Steel cladding application in Russia

A.A. Bochvar Institute has great experience in fabrication and use of steel cladding either for sodium-cooled fast reactors (SFR), or for small water cooled reactors, particularly FNPP and icebreakers, as well as high-flux SM-2 research reactor (Fig. 2) [7-12]. Steel developments included three group of materials - ferritic-martensitic steels, austenitic steels with different nickel content as well as Ni-Cr steels. If we compare their compositions of Russian and foreign steels we can clear see that ferritic steels, developed in Russia have ~ 12% chromium content (not more) as chromium provokes radiation induced hardening, which leads to embrittlement and fuel element destroying, particularly at refuelling operation. This fact was experimentally confirmed by irradiation tests in thermal reactors.

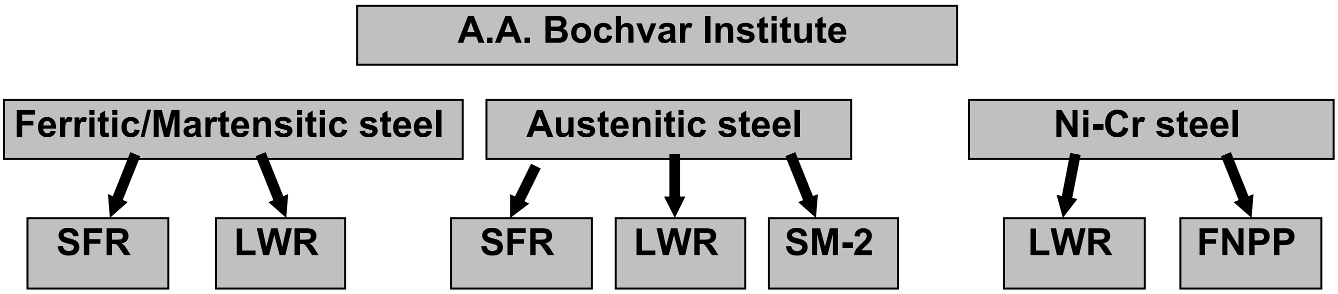


Fig. 2. Main directions on ATF steel cladding developments in A.A. Bochvar Institute.

Steel claddings with outer diameter from 2 to 30 mm and thickness from 0.1 to 0.7 mm can be manufactured on the industrial scale, which allows the expanding the range of fuel elements designs and to use them, for example in IMF design (Fig. 3).

Fig. 3. Steel tube appearance produced in Russia at the industrial scale

Besides steel claddings for fast reactors, steel claddings in Russia implemented in water-cooled reactors, operating in severe transients: Bilibinskaya Power Plant, icebreakers, Floating Nuclear Power Plants (FNPP), high flux SM-2 research reactor [7, 8, 13, 14].

A database has been collected on fuel and claddings properties and in-pile tests of fuel clad with steel and calculation programs on its application in icebreakers, FNPP, SM-2 and partly VVER type have been developed [15].
For steel cladding developed at A.A. Bochvar Institute with dispersion type fuel the following characteristics were achieved in water cooled reactors [6, 7, 12-14]:
Fuel for small reactors:
Max burn-up: 1,0 g/cm^3 under the cladding or 120 MW*d/kgU as recalculated for the standard VVER-1000 fuel rod).
Clad thickness: 0.20 - 0.30 mm
Fuel for SM research high flux reactor (100 MW power)
Steel Cladding – EI-847.
Clad thickness – 0. 15 mm.
Outer diameter – 5,15 mm.
Max burn-up – 80 at.% or 1,6 g/cm^3 under the cladding or 200 MW*d/kgU as recalculated for the standard VVER-1000 fuel rod.

4. Problems of steel application as ATF and possible ways of its improvements

ATF steels as any proposed novel system except existent advantages can have some drawbacks on the opposite site. Their analysis can help to modify and improve steel properties for application as ATF in LWRs.

Steels were considered using the following ATF criteria: melting temperature, neutron penalty, change in shape, stress corrosion cracking, radiation induced hardening, high temperature hardening,

4.1. Melting temperature

All of the above mentioned classes of steels have lower melting temperature in the range of 1350-1550 ^{0}C in comparison with Zr (1860 ^{0}C). It is natural as iron melting temperature is only 1540 ^{0}C and the main alloying elements – Ni, Cr, Al do not increase it. The lowest melting temperature of 1350 ^{0}C has Ni-Cr based 42HNM alloy, which is the most radiation resistant and perspective for ATF implementation. Moreover, the oxide films formation at accidence does not significantly increase its melting temperature - 1565 ^{0}C for Fe_2O_3 in comparison with 2700 ^{0}C for ZrO_2. But at the same time alloying elements oxides in steels have higher melting temperatures (NiO - 1955 ^{0}C, Cr_2O_2 - 2335 ^{0}C, Al_2O_3 - 2050 ^{0}C) and at their primary formation on the steel clad surface can promote the cladding shape retaining at high temperatures.

Taking into account that most of ATF steels contain sufficient quantities of these alloyed elements in steel compositions, with the appreciated oxides forming, the real melting temperature of such composites (clad with oxide film) will not be less than 2000 ^{0}C especially for Ni-Cr alloys.

Besides, when analyzing fuel behavior at accidents we should take into account not only cladding properties but fuel element as a whole. Therefore, before steel fuel element clad reaches melting temperature it will be destroyed by inner factors (inner gas pressure, interaction, etc) and failure scenario mostly will be determined by vapor – dioxide uranium pellets interaction, that leads to combustible hydrogen generation with thermal effects.

Therefore, to our mind steels melting temperature is not noticeable limitation factor at accidents. Nevertheless some of modifications can be made on this point.

For example, to use having higher melting point Cr coatings on steel claddings, especially produced by cheaper cold spray technology. It is likely that the use of ODS steels with oxide hardening, due to the higher high-temperature strength, will also contribute to conservation of the cladding shape stability [16].

4.2. Neutron penalty

Any proposed fuel system should coincide economically with the current UO_2-zirconium alloy fuel system. The thermal neutron absorption cross section of steels is about ten times that of Zircaloy [1, 17-20]. Fig 4 illustrates this fact [17]. At the same time in spite of different steel types and compositions (ferritic austenitic and Ni-Cr steels) their neutronic properties are not significantly different from each other.

The neutronic penalty necessitates thinner cladding [17, 20]. This allows to slightly larger pellets to give the same cold gap width in the rod. However, the slight increase in pellet diameter is not sufficient to compensate for the neutronic penalty and enriching the fuel beyond the current 5% limit appears to be necessary [1-3, 17-20]. Fuel claddings in the range of 0.3 to 0.5 mm width are under consideration now. For cladding thickness of 0.4 mm 6% enrichments is necessary for ferritic steels, 6.5% for austenitic steels and 7% for Ni-Cr steels [20]. Current estimates indicate that this neutronic penalty will impose an increase in fuel cost of 15-35% [3-5, 20]. It is included not only the cost of higher enrichment uranium using, but also the increase of fuel mass due to increasing of pellets diameter and hence, fuel volume.

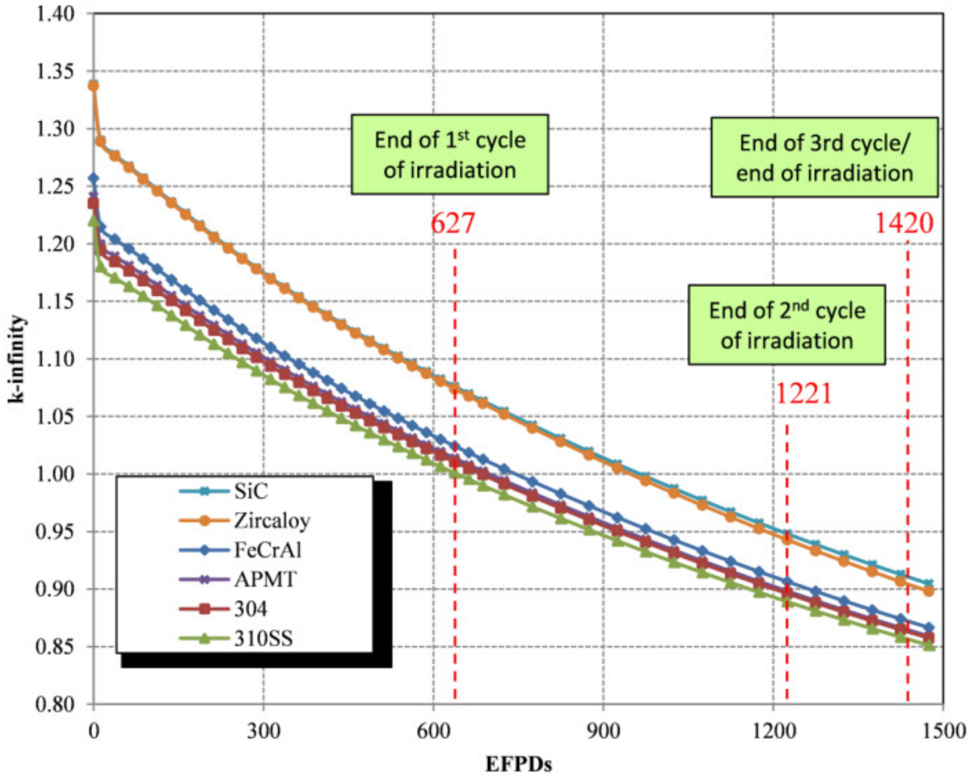
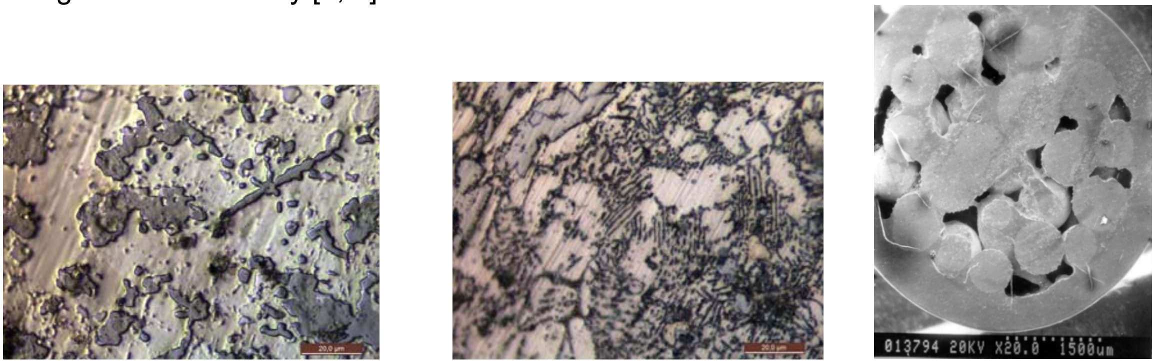


Fig. 4. Calculations of various types steel claddings with pelletized oxide fuel [17]

The other way to compensate neutron penalty is implementation of high density fuel instead of uranium dioxide, in particular, uranium disilicide (U_3Si_2), (benefit of 17% heavy metal density) [2, 3, 17, 20]. However its noticeable swelling as well as relatively poor corrosion resistance in water minimizes its advantages. To increase uranium capacity it is promising to use heavily doped uranium-molybdenum fuel as well as modified uranium-silicide fuel (Fig. 5) [6, 7, 21-25]. It is also possible to consider dispersion type fuel, in particular, composite fuel with high uranium density [6, 7].



a b c

Fig. 5. Microstructure of high density uranium alloys (fuel): U-Mo-C (a), U-Mo-(C-O-Si) (b) and macrostructure of composite fuel (c) [6, 7, 21-25]

4.3. Change in shape

Clad thickness reducing to compensate steels clad neutron penalty can lead to a loss of cladding shape stability from cylindrical to ellipse type due to radial gap width existing by coolant pressure at BOL and operating conditions. This effect can lead to fuel element failure under irradiation in pressurized water. To eliminate this effect the investigation was carried out to find the range of steel cladding stability versus steel composition, clad thickness, mechanical properties (out-of-pile and in-pile), temperature, gas and coolant pressure etc. Preliminary calculations showed that clad thickness of more than 0.4 mm for all types of steels will be stable under outer pressure and 0.35 mm for most of ferritic-martensitic steels in normal operation conditions. If we intended to use steel claddings with smaller wall tube thickness we should verify this experimentally. Implementation of more mechanical strength steels using in fast reactors as well as ODS steels will be also increase shape stability.

It should be mentioned that experimentally proved radiation and corrosion resistance of some kinds of steels in LWRs (42HNM) permits to implement 0.15-0.20 mm cladding wall thickness [11-12].

4.4 Radiation induced hardening

One of the problems of steels implementation in LWRs is radiation induced hardening at normal operation conditions, particularly for ferritic-martensitic steels (FeCrAl included) having BCC lattice. This process leads to fully loss of ductility, which finally results in cracking of fuel elements. Figure 6 illustrates the loss of short-term plasticity of one of the austenitic steels (EI844) in comparison with Ni-Cr steel (42HNM) after irradiation [12, 14, 15].

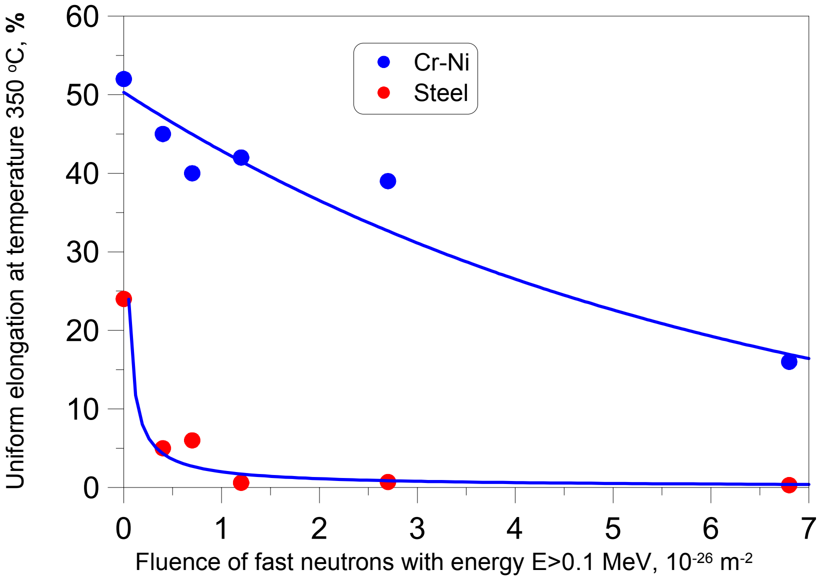


Fig. 6. Depending on the uniform elongation of the alloy Cr-Ni steel and EI844 on the fluence [12, 14, 15]

In spite of low residual ductility austenitic steels, particularly EI847, showed good-quality behavior in LWR – Bilibinskaya PP and small water-cooled reactors.

Another situation arises for ferritic-martensitic steels with BCC lattice. Formation of the Cr-rich α' phase under irradiation causes significant radiation induced hardening and embrittlement for traditional high-Cr alloys [13-15]. Chromium concentration more than 12% would limit the irradiation performance of the alloys that is especially actual for FeCrAl alloys having more than 20% (Cr + Al). Therefore, steels for Sodium Fast Reactors (SFR) contain no more than 12% Cr [8]. The influence of Al is the same. Hence, if we intend to retain minimal ductility at the end of campaign, necessary to removing fuel assembly from the core, the total Cr+Al content in steel composition should be less than 12%, that is twice less than for existing FeCrAl steels. It was also confirmed by irradiation tests of EI852 steel having 13.5% Cr. Moreover, the failure scenario was more dangerous than for Zr claddings. Cracking was occurred along the whole fuel element – from plug to plug. At the same time at accidence the Cr-rich precipitates dissolve and embrittlement disappears. It also accompanies by radiation induced defects annealing that leads to the increase of ductility.

The work towards diminishing the Cr+Al content below 12% is executing now. Naturally this can lead to a partial loss of corrosion resistance and high-temperature strength. At the same time additional alloying with elements that increase the corrosion resistance of steels, in particular, silicon, etc., may solve this problem.

Some loss of strength properties can be compensated by other small additions of alloying elements as well as thermal treatment as it is established for SFR claddings developments. It is also possible to use ODS steels with reduced Cr content, but having high-temperature strength as well as high corrosion resistance.
Bimetal claddings (FeCrAl alloy outside and austenitic steel inside) can also be one of the ways resolving the problem of radiation embrittlement.

4.5 Stress corrosion cracking

To our mind austenitic steels are the most promising variant for application in ATF as they not so sensible to irradiation induced hardening as ferritic-martensitic steels and the great experimental experience of its application in LWRs of various types is existed. But at the same time they are sensitive to stress corrosion cracking (SCC). Stress corrosion cracking is an important consideration in materials selection for accident tolerant fuel because the first instances of SCC in LWRs occurred in stainless steel fuel cladding in high temperature water (Fig 7) [4, 7, 15]. The ferritic alloys as well as Zr and Ni-Cr based alloys exhibit excellent resistance to SCC, even under quite aggressive conditions of elevated oxidants.

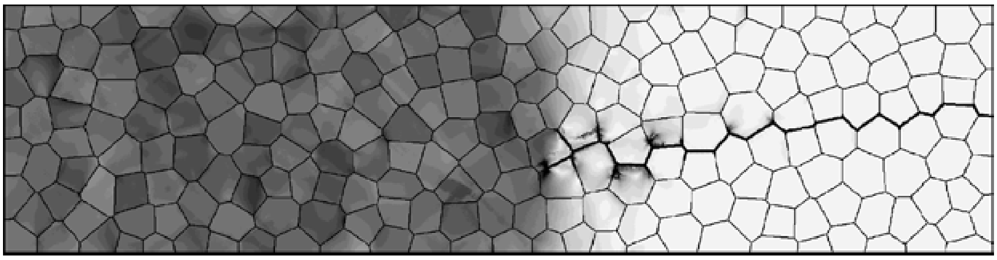


Fig. 7. Mechanism of stress corrosion cracking in austenitic steels (FCC lattice)

At A.A. Bochvar Institute a unique methodic of out-pile ampoule test of susceptibility to SCC of various types of materials has been developed. It was confirmed by numerous irradiation tests in **a** special loop of MIR reactor (NIIAR, Dimitrovgrad). The irradiation results fully coincide with methodic and showed that Zr clad alloys, Ni-Cr steels as well as all types of ferritic steels show excellent resistance to SCC in comparison with austenitic steels of various nickel content (Fig. 8) [12, 14, 15].

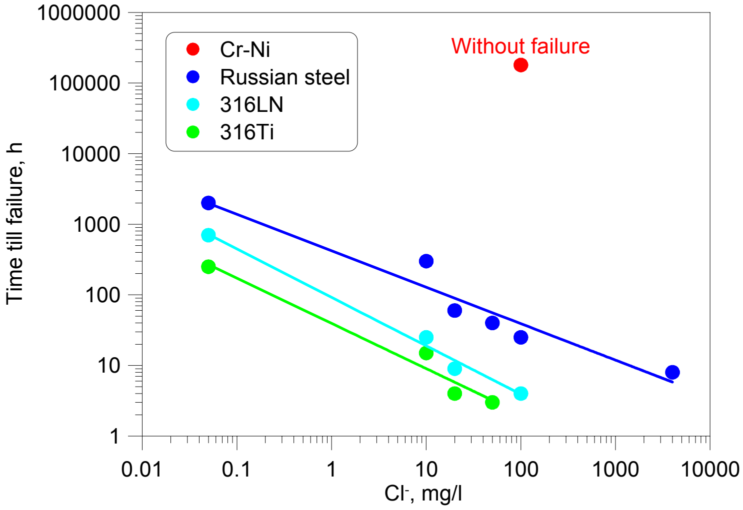


Fig. 8. Stress corrosion cracking in austenitic steels [12, 14, 15]

The mechanism of this process is not clear yet. It depends on not only of austenitic steels with FCC lattice, but also on the temperature. The attempts to understand this mechanism clearly and on this base to solve this problem did not lead to a success. Nevertheless, this process needs some time and is induced only by stresses. As the fuel-clad gap at steel application not diminishes by outer water pressure it can compensate fuel swelling up to average LWRs burn-up. Besides, even when SCC mechanism starts to operate the austenitic claddings can reach nearly 1.0 % of elongation by fuel swelling without leakage (0.3 mm initial cladding thickness). Hence, with some probability the more resistant to SCC austenitic steels in first approximation can be implement as ATF.

No doubt, efforts to find optimal composition to avoid SCC should be continued. As one of the variants to avoid SCC is to use Cr, FeCrAl or other types of coatings as well as implement bimetallic claddings.

4.6 High temperature hardening

Some types of Ni-Cr alloys are sensible to high temperature hardening at 700-800 ^{0}C temperature range. But this process requires some time that most likely would not be realized in accidents. For Ni-Cr alloys even short-term exceeding of this temperature will lead to structure changes as well as to dissolving brittle Cr precipitates along grain boundaries and neglecting this effect. At the same time Ni-Cr alloys are one of the most promising candidates for ATF thanks to its high corrosion and irradiation resistance proved by numerous claddings and fuel elements reactor tests.

5. Discussion

Analysis of the peculiarities of steels application as ATF in VVERs has shown that some ATF criteria can be modified for steels. For example, the melting temperature and corrosion resistance, since the beginning of leakage and fuel element damage occurs by inner gas pressure already at 800^{0}C. Therefore a lower (1400-1500^{9}C) than zirconium melting point of steels is not a critical limiting factor. The same applies to the steels corrosion resistance in high temperature steam, which is an order of magnitude better than that of zirconium and satisfies ATF requirements. Hence further modification of the steels corrosion resistance should not be at the expanse of the other steels properties.

The use of FeCrAl alloy as one of the main variants of ATF claddings may prove to be unsuitable since the experience of using this class of steels with a high chromium and aluminium content in Russian design fast reactors shows a drastically loss of ductility upon irradiation. Therefore, in order to preserve the minimum residual ductility at the end of the campaign, which is necessary to extract the fuel assembly from the core, the total Cr+Al content in ferritic-martensitic steels should be less than 12%, which is half the current FeCrAl steels for ATF

To compensate for the partial loss of corrosion resistance and high-temperature strength of steels at low total Cr+Al content additional alloying is suggested with elements that increase the corrosion resistance of steels, in particular, silicon, etc. The increase in high-temperature strength can be compensated by other small additions of alloying elements as well as thermal treatment as it is established for SFR claddings developments. It is also possible to use ODS steels with different Cr content. Bimetal claddings (FeCrAl alloy outside and austenitic steel inside) can also be one of the ways resolving the problem of radiation embrittlement.

It is of interest also to use austenitic claddings, which are more resistant to radiation embrittlement in comparison with ferritic steels in spite of their inclination to stress corrosion cracking (SCC). The availability of fuel-clad gap can compensate fuel swelling up to average LWRs burn-up without transferring tensile stresses to the clad. Also one of the options for preventing SCC is the use of Cr, FeCrAl or other type of coatings as well as bimetal claddings.

The use of steel claddings requires an increase in uranium enrichment above a limited 5% for uranium-235 even in spite of implementation of thinner wall claddings. It is known that the implementation of more dense fuel instead of uranium dioxide is one of the ways to solve this problem.

We propose to use heavily doped uranium-molybdenum fuel as well as modified uranium-silicide fuel that are developing in A.A. Bochvar Institute. It is also possible to consider dispersion type fuel, in particular, composite fuel with high uranium density.

Pros and cons of each steel variant are summarize in Tab. 2.

Properties, pros and cons	**Steels variants**			
	Traditional ferritic-martensitic (≤12%Cr)	**FeCrAl alloy**	**Austenitic**	**Cr-Ni (42HNM)**
Melting temperature	+	+	+	+/–
Resistance to neutron capture	–/+	–/+	–/+	–
Corrosion resistance in water	+/–	+	+	+
Corrosion resistance at LOCA	–/+	+	+/–	+
Resistance to radiation induced hardening	–/+	–	+/–	+
Resistance to SCC	+	+	–	+
Needs improvements by alloying	+	+	+	No need
Needs improvements by duplex clad implementation	+	+	+	No need
Current implementation in water-cooled reactors	–/+	–	+/–	+
Current implementation in SFR	–/+	–	+	–
Manufacturing on an industrial scale	+	–/+	+	+
Ready for use in VVERs in normal operation conditions	–	–	–	+
Need LOCA experiments	+	+	+	+

Tab 2: Pros and cons of each steel variant

6. Conclusion

Analysis of the peculiarities of steels application as ATF in VVERs was made. Three classes of steels are under consideration now. Ferritic-martensitic steels (FeCrAl alloy included), austenitic steels as well as Ni-Cr steels. Each type of steel has its advantages and drawbacks and was analyzed using the following ATF criteria: melting temperature, neutron penalty, change in shape, radiation induced hardening, stress corrosion cracking and high temperature hardening. For each class of steel some modifications were suggested to improve their properties.

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