Technology readiness level (TRL) assessment of cladding alloys for advanced nuclear fuels

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Abstract

Reliable fuel claddings are essential for the safe, sustainable and economic operation of nuclear stations. This paper presents a worldwide TRL assessment of advanced claddings for Gen III and IV reactors following an extensive literature review. Claddings include austenitic, ferritic/martensitic (F/M), reduced activation (RA) and oxide dispersion strengthened (ODS) steels as well as advanced iron-based alloys (Kanthal alloys). Also assessed are alloys of zirconium, nickel (including Hastelloy®), titanium, chromium, vanadium and refractory metals (Nb, Mo, Ta and W). Comparison is made with Cf/C and SiCf/SiC composites, MAX phase ceramics, cermets and TRISO fuel particle coatings. The results show in general that the higher the maximum operating temperature of the cladding, the lower the TRL. Advanced claddings were found to have lower TRLs than the corresponding fuel materials, and therefore may be the limiting factor in the deployment of advanced fuels and even possibly the entire reactor in the case of Gen IV.

Introduction

Reliable fuel claddings are essential for achieving the safe, sustainable and economic operation of nuclear stations. Therefore, this paper presents a worldwide assessment of the technology readiness levels (TRLs) of cladding alloys for advanced nuclear fuels. The paper is aimed at highlighting the potential benefits of the various advanced fuel cladding alloys and their current status with regards to commercialisation. This assessment can be used to compare the different alloys and their ceramic competitors.

The assessment has considered both "evolutionary" claddings (i.e. improvements to existing commercial claddings) as well as radically different "revolutionary" claddings. The nuclear reactor systems that could deploy these advanced claddings include current Generation III light and heavy water reactors (LWRs and HWRs) in addition to the potentially revolutionary Gen IV systems which are aimed at generating nuclear energy in a significantly more sustainable and secure manner [1].

The goals of these systems can only be fully realised with the deployment of advanced fuel and cladding technology. Indeed for the Gen IV systems, the deployment of the entire reactor design is dependent on the development and qualification of advanced fuels and cladding technology in order to underpin its safe and efficient operation [1].

Method

Down-selection of advanced cladding materials

In order to conduct TRL assessments, it was first necessary to down-select candidate advanced fuel cladding alloys and ceramics for deployment in Gen III and IV systems. In addition to those claddings most commonly suggested for deployment in these reactors, some materials currently of interest as fusion structural materials have also been down-selected, which have yet to be widely considered as cladding. This is because there has been a trend in recent years for advanced structural materials developed for fusion research programmes to be subsequently considered as cladding materials for fission systems, especially for Gen IV, but also for Gen III systems. In a similar vein, there has been a trend for some Gen IV cladding technologies to be postulated for application in Gen III reactors.

These down-selections were made by applying knowledge regarding the relevant systems. Fuel particle coatings and potential pipe-work/vessel materials for a Molten Salt Reactor (MSR) are considered alongside the more traditional cladding types, as they also function as a primary barrier to release of fuel material and fission products.

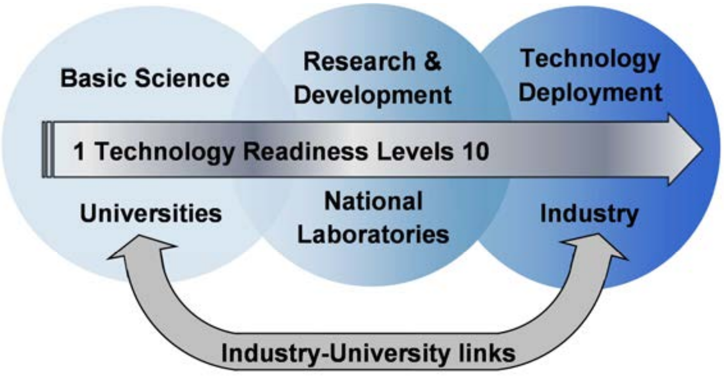
The down-selections are summarised below in order of approximate maximum operating temperature from low to high. This a useful measure of benefit as higher reactor temperatures allow for more thermodynamically efficient electricity generation. Higher temperatures also give the potential for direct heat use in a larger variety of chemical process applications (extraction of hydrogen gas from water is a particular goal of proposed Gen IV reactors with the highest operating temperatures [1]).

- **zirconium alloys**
  - standard
  - surface treated
  - advanced
- **steels**
  - ferritic/martensitic (F/M)
  - reduced activation (RA) F/M
  - surface treated F/M
  - standard austenitic
  - advanced austenitic
  - oxide dispersion strengthened (ODS) F/M
- **semi-refractory alloys**
  - advanced iron (Fe)
  - titanium (Ti) and titanium aluminide (Ti/Al) intermetallics
  - nickel (Ni)
  - hastelloys® (for MSR pipework)
  - vanadium (V)
  - chromium (Cr)
- **refractory alloy liners**
  - niobium (Nb), tantalum (Ta), molybdenum (Mo), tungsten (W)
- **ceramic-based**
  - MAX phase
  - cermets
  - SiCf/SiC
  - Cf/C
  - SiC TRISO fuel particle coatings
  - ZrC TRISO fuel particle coatings

Technology readiness levels (TRLs)

The TRL system is a means of measuring technology maturity, with a degree of standardisation, which allows for comparison between different technologies. Originally defined by Mankins (1995) of NASA [2], TRLs have become adopted by many industries around the world. As the technology matures from the lower TRLs to the higher TRLs, it moves from a scientific idea through to a fully developed application that has demonstrated its usefulness by being deployed in the real world in an operational situation. Figure 1 illustrates where the development of technologies at different TRLs may be conducted to advance their TRL in a typical national technology supply chain.

Figure 1: TRLs and the national technology supply chain



It should be emphasised that the NASA TRLs were defined for systems for individual space missions (often electronic) and the terminology is not always suitable for nuclear industry applications. Hayes and Porter (2007) of INL (Idaho National Laboratory) [3] reported an adaptation of the TRL system for application to fast reactor fuel.

There is some apparent inconsistency between the NASA and the INL definitions (which both have TRLs 1 to 9). For example, INL TRL 1 seems to correspond more closely with NASA TRL 2, due to INL not including basic principles research at the bottom of the scale. Furthermore at the top of the scale, INL TRL 9 could feasibly correspond to an assumed NASA TRL 10, if their definitions were extrapolated to operation of many actual systems as opposed to single missions. Therefore overall, the INL scale appears to be shifted down by one TRL with respect to NASA's.

For the cladding TRL assessments in this paper, a combined approach has been used that includes elements of both the NASA and the INL definitions. Some additional simplification and generalisation was also employed to allow a flexible approach to what is in reality a highly complex situation. Consistency has been maintained with the NASA scale, with TRL 1 still corresponding to basic principles research. In addition, aspects of the more precise INL approach have been incorporated including their guidelines regarding out-of-reactor/in-reactor testing, lead assemblies and core reloads. A TRL 10 has also been defined based on the INL TRL 9 to consider long-term operation of many actual systems. The TRL definitions used for this paper are presented in Table 1 using a "traffic light" colour coding that is employed throughout.

Assessing the TRL of the cladding material down-selections

For each down-selection, a literature search was performed seeking papers in peer-reviewed journals and reports produced by international nuclear organisations (IAEA, OECD/NEA, WNA) as well as the work of national nuclear institutions. Further information was sought where appropriate through attending various conferences and by contact with partners in the international nuclear community. Using this information, it was then possible to use the TRL definitions in Table 1 to assess the TRL of the down-selections.

The ascribed TRLs represent an international best case scenario for each technology with the TRL being representative of its most developed form in the country or countries that have progressed it to the greatest extent. These international TRLs do not necessarily equate to the TRL applicable to individual countries. The assessment also considered TRLs with respect to the reactor type/s for which the cladding is most developed. A lower TRL would apply to the use of the same cladding in a different reactor type.

Table 1: TRL definitions for nuclear fuels and claddings

TRL	Definition and description
10	Widespread, reliable and long-term operation of many actual systems e.g. long-term use of a fuel within a commercial reactor fleet/fleets with many thousands of hours of operating experience and data
9	Successful operation of actual system e.g. assemblies have performed successfully under irradiation in reload quantities (demonstrated by surveillance programme)
8	Actual system constructed and commissioned e.g. assemblies fabricated in reload quantities, may include irradiation with only limited success
7	Prototype successfully demonstrated e.g. lead use assemblies have performed successfully in a prototype or commercial reactor (demonstrated by PIE and/or in-core monitoring)
6	Prototype construction (much more representative than the basic system) e.g. lead use assemblies have been fabricated, and potentially irradiated in a prototype or commercial reactor but with only limited success
5	Basic system successfully demonstrated e.g. test rods have been irradiated and performed successfully in a test reactor (demonstrated by in-reactor instrumentation and/or post irradiation examination (PIE) and/or post irradiation mechanical testing)
4	Integration of components into a basic system e.g. representative assembly sections have been manufactured and subjected to out-of-reactor tests and/or test reactor irradiation trials of individual rods have been conducted with only limited success
3	Basic components fabricated and successfully demonstrated e.g. fuel and/or cladding components have been manufactured and tested out-of-reactor and/or irradiated as a component only
2	Practical applications suggested and concepts formulated e.g. fuel, cladding and/or fuel assembly designs have been established
1	Research identifies the basic principles that underlie the technology e.g. promising materials and/or geometry have been identified

Table source: This table represents an amalgamation of those presented in [2] and [3]

Limitations of TRL assessments

It should be emphasised that a TRL assessment is at best a crude measure of a complex and ever changing international technological situation. Interpretation and use of the definitions in a TRL assessment is inevitably somewhat subjective and challenging to apply consistently.

TRL assessment also gives no indication of the amount of time/effort/cost required to increase a technology's TRL. For example, if two technologies are currently at the same TRL, then there is no guarantee that these will continue to be developed successfully at the same rate. Indeed a technology currently with a lower TRL may reach deployment sooner than another technology which currently has a higher TRL due to increased R&D effort, fewer feasibility issues etc. Importantly, there is no guarantee that any technology will ever reach the highest TRL as it may ultimately be found to be unfeasible during further development.

In spite of these limitations, a TRL assessment may still prove useful as a guide for further study. It should be noted that TRL values are potentially more useful for comparisons between technologies than they are when considered individually as absolute values. TRLs themselves give no indication of the relative benefits of the different technologies if they were fully deployed, though this weakness can be overcome by plotting TRLs against appropriate measures of benefit.

Results

The full details of the TRL assessment of each cladding material down-selection are given in an NNL report produced for the UK Department of Energy and Climate (DECC) [4]. Details include a full description of each material and its benefits, a full written justification for each ascribed TRL, and the most important literature references (typically

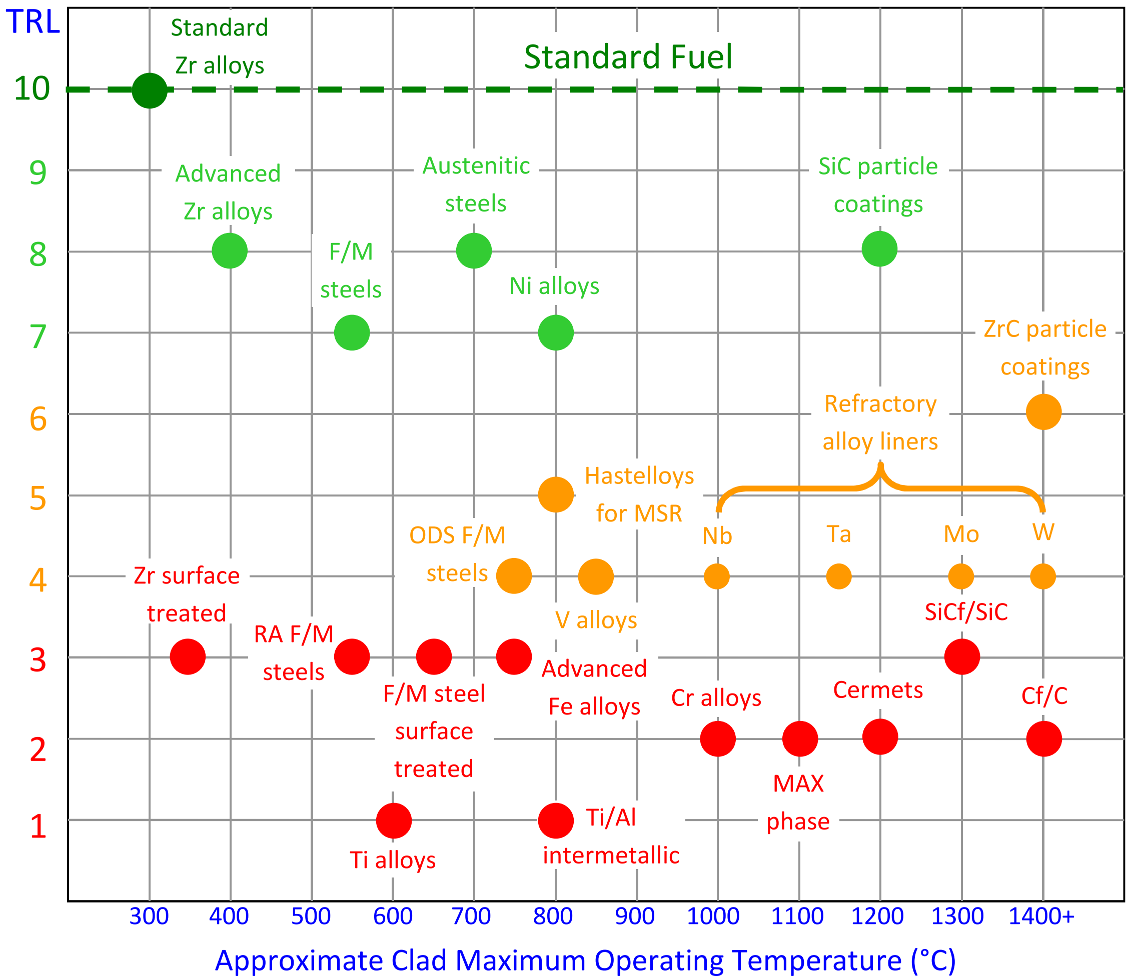
4-8 per technology). A summary of the assessments from the full report is given in this paper using the same “traffic light” colour coding as Table 1.

Table 2 gives the ascribed international best case TRLs for the down-selected cladding materials alongside a brief justification and a key reference. Figure 2 then plots these international best case TRLs against their approximate maximum operating temperature from low to high.

Table 2: International best case TRL assessments for advanced claddings

Advanced cladding categories *In order of approximate maximum operating temperature (low to high)*		**Best case TRL**	**TRL Justification and Key Reference**
Zirconium	Standard alloys	10	Used in PWR, BWR and HWR stations worldwide for many years [5].
	Surface treated	3	Some development in Republic of Korea, United States and Russia [6].
	Advanced alloys	8	New products from United States, France, Russia and Japan nearing market [5].
Steels	F/M	7	Successful cladding in a number of prototype SFRs [7].
	RA F/M	3	Extensive development by fusion programme. Qualification in its infancy for fission reactors [8].
	F/M surface treated	3	Development and testing being driven by LFR programmes [9].
	Standard austenitic	8	Past standard LWR clad material that would require updating to modern standards to use once more [10].
	Avanced austenitic	8	Moderate success in large SFRs (BN600 in Russia, Superphenix in France) [7].
	ODS F/M	4	Moderately successful tests of Russian and Japanese rodlet claddings in a basic SFR [7].
Semi-refractory alloys	Advanced Fe	3	Fe-Cr-Al Kanthal alloys being tested for LWR cladding application [10].
	Ti and Ti/Al intermetallic	1	Some development of Ti [11] and Ti/Al [12] for fusion programmes. Yet to be widely considered for fission.
	Ni	7	Tested successfully in UK and US fast reactors in 1970s-1990s [13].
	Hastelloys	5	Pipework in US Molten Salt Reactor Experiment (MSRE) in 1960s with reasonable success [1].
	V	4	Tested as liner in UK fast reactor in 1950s-1960s with limited success [14]. Extensive development of new alloys by fusion programme [15].
	Cr	2	Currently only a few fuel concepts. Very limited irradiation data [16].
Refractory alloy liners	Nb, Ta, Mo & W	4	Tested in fast reactors in 1950s-1980s with limited success (US space reactor programme) [17].
Ceramic-based	MAX phase	2	Capability first discovered in the 1990s. Now a few concepts and first irradiation trials are underway [18].
	Cermets	2	Currently only a few fuel concepts for various reactors [19].
	SiCf/SiC	3	Leading candidate for GFR application and now considered for LWR, SFR and LFR. Extensive development by fusion programme [20].
	Cf/C	2	Some consideration for fission applications. Extensive development by fusion programme [21].
	SiC particle coatings	8	SiC coatings performed successfully in attempted commercial HTRs in Germany and United States. Current programmes in China and Japan [22].
	ZrC particle coatings	6	Needed for VHTR. Irradiation programmes underway in HTR prototypes with some success [22].

Figure 2: Advanced clad TRLs vs. approximate maximum operating temperature



Finally, Table 3 shows which reactor types the cladding materials are relevant to and gives TRLs with respect to each. The reactor types considered are Gen III reactors (LWRs and HWRs) and the six Gen IV systems as listed below [1]:

- **SFR**, **LFR** & **GFR** - sodium, lead & gas-cooled fast reactor respectively.
- **HTR/VHTR** – high- and very-high-temperature reactor.
- **SCWR** - supercritical water reactor.
- **MSR** - molten salt reactor.

Discussion

It can be seen from Table 2 and Figure 2 that in general as the maximum operating temperature of the potential cladding materials increases, the TRL decreases. Therefore Zr alloys have the highest level of technology readiness followed by steels. Refractory and semi-refractory alloys with the exception of nickel alloys have low technology readiness as do ceramic cladding tubes. This was in line with pre-assessment expectations given the revolutionary nature of the materials with the highest potential operating temperatures.

Table 3: TRLs of advanced clads vs. reactor systems

In order of approximate maximum operating temperature (low to high)	**Generation**	**III**	**IV**					
	Reactor	**LWR/ HWR**	**SFR**	**SCWR**	**LFR**	**MSR**	**GFR**	**HTR/ VHTR**
Advanced clad categories	**Outlet temperature**	~325°C (PWR)	550°C	510 – 625°C	480 – 800°C	700 – 800°C	850°C	650 – 1 000°C
Zirconium	Standard alloys	10				**Grey indicates that the material is not relevant to the reactor type**		
	Surface treated	3		2				
	Advanced alloys	8		3				
Steels	F/M		7		4			
	RA F/M		3	3	3			
	F/M surface treated			2	3			
	Standard austenitic	8 (former standard)	6	3	4		3	
	Advanced austenitic	6	8	3	4		3	
	ODS F/M		4	2	3		3	
Semi-refractory alloys	Advanced Fe	3	2	2	2	2		
	Ti and Ti/Al intermetallic	1	1	1	1			
	Ni		7	3			3	
	Hastelloys					5		
	V		4		2		3	
	Cr	2	2	2	2		2	
Refractory alloy liners	Nb, Ta, Mo & W		4		3		3	
Ceramic-based	MAX phase	2	2	2	2		2	
	Cermets		2		2		2	
	SiCf/SiC	3	3	2	3		3	
	Cf/C	1	1	1	1		2	
	SiC particle coatings						3	8
	ZrC particle coatings						2	6

Notable exceptions to this trend are the SiC and ZrC TRISO fuel particle coatings which have attained reasonably high TRLs in HTR/VHTR programmes. It might be anticipated therefore that these non-traditional cladding forms will be deployed significantly in advance of traditional tube-shaped claddings for such high temperatures. Table 3 shows the diverse and complex relationship between cladding TRLs and reactor types. However, the same general inverse relationship between TRL and operating temperature applies with the same notable exception of HTR/VHTR. It is also evident that the TRLs for individual cladding technologies are higher for just one or two reactor types and much lower for the others. Indeed, for four out of the six proposed Gen IV reactor types the cladding TRL is no higher than 5 at best.

Conclusions

A significant level of further development of advanced claddings will be required before commercial use is possible. Many have TRLs within the usual development range of national laboratories and universities (TRL 7 and below), which suggests that these organisations will need to play a crucial role in bringing them to industry readiness. When compared with a similar TRL assessment of advanced fuels [4], it was found that the corresponding claddings often had a lower TRL. Therefore cladding development may be the limiting factor in the development of new fuel designs, and crucially this may in turn limit the deployment of advanced reactor systems. Therefore, further international development of cladding technology needs to be pursued urgently as an integral part of future advanced reactor and nuclear fuel cycle development programmes.

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References

[1] US DOE and GIF (2002), "A Technology Roadmap for Generation IV Nuclear Energy Systems", GIF-002-00.

[2] Mankins, J.C., NASA (1995), "Technology Readiness Levels", NASA White Paper, 6 April.

[3] Hayes, S.L. and D.L. Porter, INL (2007) "SFR Fuel Performance and Approach to Qualification", Presentation to DOE/NRC Seminar Series on Sodium Fast Reactors, 27-28 November.

[4] Shepherd, D. et al., NNL (2013) "Technology Readiness Level assessment of advanced fuels, cladding and associated manufacturing technology", NNL (13) 12502.

[5] Lemaignan, C., CEA (2012), "Zirconium Alloys: Properties and Characteristics", Comprehensive Nuclear Materials, Elsevier, ISBN 978-0-08-056033-5, R.J.M. Konings (Editor), Volume 2.07, pp. 217-232.

[6] Ivanova, S.V. et al., MEPhI and partners (2010), "Methods to Increase Operation Properties of Zirconium Components for New Generation LWR Active Cores", LWR Fuel Performance Conference, 26-29th September 2010, Orlando Florida, United States, Proceeedings, Paper 120, pp. 587-593.

[7] IAEA (2012), "Structural Materials for Liquid Metal Cooled Fast Reactor Fuel Assemblies – Operational Behaviour", IAEA Nuclear Energy Series, No. NF-T-4.3, ISBN 978-92-0-131610-3.

[8] Jones, R.H. et al., PNNL and INL (1999), "Low activation materials", Journal of Nuclear Materials, 271 and 272, pp. 518-525.

[9] Takaya, S. et al., JAEA and partners (2012), "Al-containing ODS steels with improved corrosion resistance to liquid lead-bismuth", Journal of Nuclear Materials, 428, pp. 125-130.

[10] Terrani, K.A. et al., ORNL (2013), "Advanced Oxidation-Resistant Iron Alloys as LWR Fuel Cladding", Enlarged Halden Programme Group Meeting, 10-15 March 2013, Gol, Norway, Proceedings, HPR-378-1, pp. 215-221.

[11] Bilobrov, I. and V. Trachevsky, NASU (2011), "Approach to modify the properties of titanium alloys for use in the nuclear industry", Journal of Nuclear Materials, 415, pp. 222-225.

[12] Hishinuma, A., JAERI (1996), "Radiation damage of TiAl intermetallic alloys", Journal of Nuclear Materials, 239, pp. 267-272.

[13] Rowcliffe, A.F. et al., ORNL (2009), "Perspectives on radiation effects in nickel-base alloys for applications in advanced reactors", Journal of Nuclear Materials, 392, pp. 341-352.

[14] Sinclair, V.M., UKAEA (1961), "Canning materials for D.F.R. fuel elements", UKAEA TRG Memorandum 178.

[15] Chen, J.M. et al. (2011), Southwestern Institute of Physics Chengdu China and partners, "Overview of the vanadium alloy researches for fusion reactors", Journal of Nuclear Materials, 417, pp. 289-294.

[16] Le Flem, M. et al., CEA (2011), "Advanced Materials for Fuel Cladding in Sodium Fast Reactors: From Metals to Ceramics", Presentation to 3rd MATGEN Summer School, 19-23rd September 2011, Lerici Italy.

[17] Cox, C.M. et al., Hanford Engineering Development Laboratory (1984), "Fuel Systems for Compact Space Reactors", HEDL-SA-3065-FP.

[18] Hoffman, E.N. et al., SRNL and partners (2012), "MAX phase carbides and nitrides: Properties for future nuclear power plant in-core applications and neutron transmutation analysis", Nuclear Engineering and Design, 244, pp. 17-24.

[19] Le Flem, M. et al., CEA (2008), "Microstructure and thermal conductivity of Mo-TiC cermets processed by hot isostatic pressing", Journal of Nuclear Materials, 380, pp. 85-92.

[20] Snead, L.L. et al., ORNL (2013), "An Overview of SiC-Based Fuel and Cladding Technologies in Support of Accident Tolerant Fuel Development", Enlarged Halden Programme Group Meeting, 10-15 March 2013, Gol (Norway), Proceedings, HPR-378-1, pp. 203-213.

[21] Venugopalan, R. et al., BARC (2010), "Neutron irradiation studies on low density pan fibre based carbon/carbon composites", Journal of Nuclear Materials, 404, pp. 19-24.

[22] IAEA (2010), "High Temperature Gas Cooled Reactor Fuels and Materials", IAEA-TECDOC-1645.