

# **TECHNICAL ANNEX**

TO THE DOCUMENT

## **MATERIALS FOR SUSTAINABLE NUCLEAR ENERGY**

*The Strategic Research Agenda (SRA) of the Joint Programme on Nuclear Materials (JPNM)  
of the European Energy Research Alliance (EERA)*





## TA.1 Detailed nuclear materials research agenda

### TA.1.1 Structural materials

#### TA.1.1.1 Pre-normative research: qualification, test procedures, design rules

This section concerns the development of the assessment tools and the collection of the materials data in support of the design, licensing, and construction of the ESNII demonstrators, with a view also towards the FOAK prototypes and commercial reactors to be deployed later.<sup>1</sup>

An especially delicate issue in this context is the **extension of the operational life of non-replaceable components from 40 to at least 60 years as a general Gen IV requirement.**<sup>2</sup> All relevant slow long-term degradation processes need to be accounted for, especially high-temperature processes (creep, fatigue, thermal ageing), but also corrosion and low-dose-rate long-term irradiation effects. A fundamental issue is **how to measure material properties representative for long-term operation.** This is a tremendous challenge shared by all Gen IV concepts, for which design and assessment methodologies need to be developed and translated into codes and standards. For instance, for creep extrapolation by a factor 3 is considered feasible, but this would require tests of 20 years duration for reliable 60 years design data. In a future low-carbon energy mix, the nuclear contribution will also need to operate in a load-following mode, whereby the reactor components will accumulate more load cycles, which should also be taken into account in a long-term operation perspective. Although the 60-years operational life is a requirement for the commercial deployment, thus a long-term need, it is a short-term R&D need to start in due time, i.e. now, in view of the long-term test programmes<sup>3</sup>.

Specifically for the HLM-cooled systems, the most important issue is to demonstrate the structural integrity of components in contact with the corrosive coolant. There are no dedicated design rules, assessment procedures or material data in the codes to address HLM environmental effects. Thus there is an **urgent need to perform pre-normative research on the compatibility between components and HLM coolants** and develop procedures to be included in codes and standards.<sup>4</sup>

Importantly, most structural integrity issues in metallic components occur at the welds, the assessment of which is especially complex. This means that **much emphasis in the pre-normative research should be dedicated to welded components.**

Finally, the high investment cost is perhaps the largest obstacle for the development and deployment of innovative nuclear reactors. Thus, **in addition to demonstration of safety, the codes and standards should also ensure cost efficiency.** Pre-normative research related to reducing cost without reducing safety margins include: reduction of undue conservatism in design rules by applying more advanced assessment procedures; assessment of alternative materials and designs; more accurate and less conservative descriptions of loads; updated materials curves based on additional tests methods as well as extended test conditions. This implies that conceptually the development of codes and standards should be based on a physical understanding of materials degradation processes.

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<sup>1</sup> Pre-normative research has been a key objective in the FP7 projects MATTER and MATISSE (<http://www.fp7-matisse.eu/>), and is the key focus in the H2020 project GEMMA (<http://www.eera-jpnm.eu/gemma/>) and in ongoing JPNM Pilot Projects. See in particular the summary report from the MATTER project for an overview of work done and envisaged: "Deliverable 9.1: The relevance of MATTER results for the design of ESNII reactors", <http://www.eera-jpnm.eu/gemma/>.

<sup>2</sup> Technology Roadmap Update for Generation IV Nuclear Energy Systems, January 2014, <https://www.gen-4.org/gif/upload/docs/application/pdf/2014-03/gif-tru2014.pdf>

<sup>3</sup> See Final Report EERA JPNM Task Force 60 years Operational Life *Future Reactors Review and Roadmap for Future Activities*, available from <http://www.eera-jpnm.eu/filesharer/documents/> and Technical Area 1a of the MoU between EERA and SNETP: *Development of a methodology for design and plant life management for 60-years operational life for non-replaceable components of Generation IV reactors*

<sup>4</sup> See Technical Area 1b of the MoU between EERA and SNETP: *Development of Design Rules, qualification and mitigation strategies for reactor components in heavy liquid metal coolants*

The **pre-normative research includes closely integrated experiments and modelling work at the meso- and macroscale**. The engineering scale is the basis for reactor design, but the understanding of the degradation mechanisms and of how to include them in the design rules and assessment procedures requires the proper length and time scale to be addressed for the particular degradation mechanism. One example is the extrapolation of accelerated data to operational conditions where the proper relevant degradation mechanism must be explicitly taken into account. The end product would then be engineering tools based on a multi-scale *informed* approach.

In what follows the main pre-normative research issues are described for the key materials of the ESNII demonstrators. The 60-years design lifetime is not mentioned anywhere as a specific issue as it corresponds to a combination of several of the issues in what follows and in the table. The **reference structural materials are austenitic steels, on which most research effort is focused**, in particular 316L and 316L(N), that are planned to be used **for the core of all prototypes. In the case of the GFR, Ni-base alloys are also reference materials**, in particular the high-Ni alloy 800H for out-of-core components. Alloy 800H is qualified up to 750°C for the steam generator; but for the other high temperature components – turbine blades, hot gas header, intermediate heat exchangers – Ni-base alloys with higher creep rupture strength are required, e.g. Inconel 617, Haynes 230, or Hastalloy-XR.<sup>5</sup> **Some emphasis remains on 9Cr F/M steels** which, although not used for MYRRHA and ALFRED, and only to a limited extent in ASTRID, remain **candidate materials for the phase II of some demonstrators**. For **cladding** the reference material is the **15-15Ti austenitic steel**.

### High temperature behaviour and degradation of metals

Creep effects need to be taken into account above the negligible creep temperature, which according to RCC-MRx is 470, 425 and 375°C, for Alloy 800, 316SS and F/M steels, respectively. Many fast reactor components operate in the creep regime, where **creep, creep-fatigue and thermal ageing** limit the operational life of components.<sup>6</sup> In addition to the 60-years design life, high-temperatures must also be considered for accident conditions, where temperature excursion could be well above the normal operational temperature.

#### *Creep and creep-fatigue deformation*

Design rules and assessment procedures need to describe creep and relaxation curves for the reference materials under the operational conditions. Most models and material test data in design codes are restricted to relatively high temperatures and stresses where creep is governed by dislocation climb. At the operational conditions the stresses and temperatures are generally lower and creep deformation is mainly by diffusion and creep rates are very small.<sup>7</sup> Extrapolation of dislocation-climb creep data from accelerated tests to operational conditions where creep is by diffusion will give non-conservative predictions.<sup>8</sup> **For the long-term operation it is necessary to have models and data that incorporate the degradation mechanisms or microstructural evolution itself by addressing the proper length scale.** Moreover it may not be feasible to base design rules for very small creep rates on creep rupture. Instead other approaches, for instance to base design on small creep deformation (e.g. 1%), should be explored.

Fast reactors operate at low pressure, but cyclic thermal loads with a typical temperature variation of 100°C are significant. Thus creep-fatigue, or more specifically **non-isothermal creep-fatigue, will be a dominant mechanism**. Design rules, assessment procedures and material curves are much less developed for creep-fatigue than for creep or fatigue, so more accurate models are needed. **This problem is especially serious for F/M steels that undergo significant softening under cyclic loading.** Understanding the underlying processes and developing suitable models are necessary to establish design rules without undue conservatism. Creep and relaxation may occur simultaneously in

<sup>5</sup> There has also been effort to develop special ODS alloys with high temperature strength and stability (MA6000).

<sup>6</sup> For a general overview of high-temperature assessment approaches see *Comprehensive Structural Integrity, Fracture of Materials from Nano to Macro Vol. 4 Cyclic Loading and Fatigue and Vol. 5 Creep and High-Temperature Failure*, 2007, Elsevier-Pergamon. For nuclear applications see *Structural Materials for Generation IV Nuclear Reactors*, Edited by Pascal Yvon, Woodhead Publishing Series in Energy, Elsevier, 2017

<sup>7</sup> Frost and Ashby, *Deformation Mechanism Maps: the plasticity and creep of metals and ceramics*. Oxford: Pergamon, 1982.

<sup>8</sup> Cui et al. Modelling and experimental study of long term creep damage in austenitic stainless steels, *Engineering Failure Analysis*, 58 (2015) 452.

components under cyclic loading. The goal should therefore be to have unified constitutive models that include forward creep and stress relaxation, plastic deformation, and include key features such as cyclic hardening (austenitic steel and Ni-based alloys) and softening (F/M steels), progressive accumulated deformation (ratchetting) and creep or relaxation during hold-times. The obvious way forward is to use visco-plastic constitutive models that include kinematic hardening and isotropic hardening/softening, and possibly also other effects such as recovery.<sup>9</sup> Since loads are predominantly thermo-mechanical with a very large variation in the creep rate in the temperature range 450-550°C, the temperature dependence needs to be taken into account for the constitutive models.

### ***Creep and fatigue damage and crack propagation***

To predict the life-time of components it is also necessary to **incorporate the initiation of damage and crack propagation until final failure**. The current approach for creep-fatigue is based on creep-fatigue interaction diagram and is not very accurate, which in the design must be compensated by large safety margins. Models that actually **integrate creep and fatigue with proper damage criteria** are needed for more reliable predictions. Methods are required to predict whether a given defect in a component may become critical. This demands the application of fracture mechanics models and in particular for crack growth in the creep-fatigue regime more reliable models need to be developed. An issue for both damage initiation and crack propagation is the very long test durations needed to get test data representative for temperature, load amplitude and hold-times.

### ***Thermal ageing***

Thermal ageing is caused by microstructural evolutions and affects the long-term material properties and, most importantly, reduces the ductility of the material.<sup>10</sup> The microstructural evolution depends both on stresses and temperature and is difficult to predict. In the ASME and RCC-MRx codes, thermal ageing is only included in reduction factors for some selected properties such as yield and ultimate strength and fracture toughness, and only for a few materials.

To include thermal ageing effects in design codes and assessment procedures requires first that the **microstructural evolution is predicted by thermodynamic codes** such as CALPhad or METCALC and secondly knowledge of how it affects the mechanical properties. Models of this type are the subject of section 3.1.2.

### ***Interaction creep, creep-fatigue and thermal ageing and upscaling***

Models that combine the effects of creep, fatigue and ageing are needed for total life assessments of components.<sup>11</sup> To upscale from mechanistic models to polycrystal continuum models requires homogenization schemes, for instance through mean-field theories.<sup>12</sup>

### ***Data collection and production for 60 years design life***

The **key to the development of a methodology for a 60 years reactor operation life is access to representative long-term material data**. There will never be "enough" data so three different, but complementary, approaches should be pursued.

- Large test programmes, including long-term tests, were performed in the previous decades, in particular for 316L stainless steel grades and welds. **Compilation and assessment of such "historical" data** should be the starting point. Unfortunately, data management plans were not developed at that time and the data available are rather limited and not so easy to use.<sup>13</sup>
- A second extremely useful approach would be to perform **mechanical tests and microstructural analyses of materials from reactor components operated in the past**, such as the French sodium fast reactors and the UK AGR reactors - the reactor type which has the largest accumulated operational life in the relevant temperature range (500 – 600°C).

<sup>9</sup> Chaboche, Constitutive equations for cyclic plasticity and cyclic viscoplasticity. International Journal of Plasticity 5(3) (1989) 247.

<sup>10</sup> Pickering, Physical Metallurgy of Stainless Steel Developments, International Metals Reviews, 12 (1976) 227.

<sup>11</sup> There are very few examples in the literature where the combined effect of creep and thermal ageing are addressed. One example is B. Dyson, Use of CDM in Materials Modelling and Component Creep Life Prediction, Journal of Pressure Vessel Technology, 122 (2000) 281.

<sup>12</sup> Mercier et al., Validation of an interaction law for the Eshelby inclusion problem in elasto-viscoplasticity, International Journal of Solids and Structures 42 (2005) 1923.

<sup>13</sup> One example is the collection of rupture data for 316 welded joints Escaravage et al. CEC Study Contract RA1 - 0179 F, Evaluation of Mo containing austenitic weld metal stress rupture strength, Part I : Collection of Data, 1990.

- **New test programmes need in any case to be defined and performed.** This should include materials and components manufactured in accordance with the most recent specifications in RCC-MRx. The bulk of the new testing should be different accelerated tests, complemented with long term validation tests to include ageing. The key to the relevance of accelerated tests is that the same degradation mechanism should be activated as for the long-term tests. Mechanistic models are needed to define an optimized test plan, as well as to evaluate the post-test results. Acceleration is not simply a question of increasing the loads and the temperatures. New test procedures need to be developed. For instance, creep damage can be accelerated by increasing the stress-triaxiality.<sup>14</sup> In stress relaxation tests, the creep rates decrease to in-service relevant levels much faster than in classical uniaxial (forward) creep tests. Stress relaxation could potentially be used as accelerated creep tests provided a visco-plastic model exists that can describe both phenomena.<sup>15</sup>

## Environmental compatibility between coolant and structural materials

### *HLM-cooled systems*

While liquid sodium is relatively benign to steels, it is well known that material in contact with **heavy liquid metals (HLM)**, i.e. liquid lead and lead alloys such as lead-bismuth eutectic (LBE), may degrade severely by **corrosion, dissolution and erosion**. In addition, **liquid metal embrittlement (LME)** is a phenomenon specific to some materials that may affect severely the mechanical performance and determine the failure of components. So, in **HLM-cooled reactors the main issue is to qualify materials** in order to be sure that they will retain their integrity **in contact with the coolant for the lifetime of the component**.

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Concerning resistance to corrosion, it is essential to **map, for the selected materials and coolant, the corrosion rate as a function of all the variables involved**, namely temperature, impurity content (especially oxygen), and fluid velocity. It must be kept in mind that in the reactor it will be impossible to guarantee the same level of variable control as in a laboratory test, thus the range of testing should exceed the range of nominal service conditions and emphasise the most penalizing conditions. Corrosion tests need to be conducted in flowing conditions whereas mechanical tests (slow strain rate, fracture, creep, fatigue and fracture toughness tests) can partially be done under stagnant conditions. Importantly, **test procedures and standards are not fully developed for HLM environments so such activities need to be considered**. The resulting mapping should be eventually the basis for the elaboration of design rules.

Three **steps towards design rules and licensing**, valid also for LME, can be identified:<sup>16</sup>

- Demonstrate "immunity", i.e. that testing in environment is equivalent to testing in air;
- If immunity cannot be demonstrated but environmental effects are relatively slow, acceptable and predictable, establish a mitigation programme to ensure long-term integrity based on adaptation of the systems parameters, acceptance criteria and inspection plans to accommodate affordable corrosion allowances.;
- If no solid mitigation programme can be identified, then the mapping of the behaviour in environment of non-conventional materials needs to be considered: these may be surface engineered steels (coatings) or alumina forming steels.

Based on an approach of this type it has been seen that F/M steels suffer from severe LME at the operational conditions for both MYRRHA and ALFRED and have therefore been ruled out as structural materials for demonstrators that use HLM as coolant. In contrast, **austenitic 316 steels seem to be**

<sup>14</sup> Hosseini et al., The LICON methodology for predicting long-time uniaxial creep rupture strength of materials, International Journal of Pressure Vessels and Piping 111-112 (2013) 27.

<sup>15</sup> Woodford, Test Methods for accelerated development design and life assessment of high-temperature materials, Materials & Design, 14 (1993) 231.

<sup>16</sup> This follows the Code evolution proposal PG2/CE-01: "Use of the code in innovative coolant environment" and associated R&D proposal PG2/RD-01: "Development of Design Rules and Characterisation of Material Behaviour in Heavy Liquid Metal (HLM) Environments" of the CEN WS064 Design and Construction Code for mechanical equipments of innovative nuclear installations as well as the Technical Area 1b of the MoU between EERA and SNETP

**quite resistant to HLM degradation, but this needs to be further demonstrated** by an experimental programme that should **include welded components** and define clear **boundaries of operation**.

The environmental degradation under operational conditions may be quite slow and **long-term tests may be needed before the effects can be observed**. Complementary **accelerated tests by a more aggressive environment** are a possible shortcut. More aggressive environments are also needed to establish boundary conditions. In addition to varying temperature and loads (for mechanical tests), the liquid can also be made more aggressive by changing the chemical composition. The interpretation and use of accelerated test data will be much more complicated than for high temperature degradation, though.

As for the high temperature degradation, accelerated tests need to be **complemented with appropriate models and microstructural characterization** to ensure consistency of degradation mechanisms. Importantly, for completeness, a qualification test programme should also include the simultaneous effect of irradiation and environment (see also next section). The modelling of HLM degradation mechanisms corrosion and liquid metal embrittlement is much less advanced than for creep and irradiation. Engineering models need to be developed to interpret test data and support design rules. Such models will, to a large extent, be semi-empirical and based on semi-analytical mathematical models. Corrosion models that can calculate the corrosion rates, corrosion and oxidation layer thickness and transport of corrosion products have been developed.<sup>17,18</sup>

**Liquid metal embrittlement** may manifest in different ways: increased crack growth rate, reduced fracture toughness, reduced tensile elongation. Some basic models such as have been developed and applied but mainly applied to single solid/liquid couples.<sup>19</sup> These models differ with respect to assumptions and the specific mechanisms, e.g. adsorption versus dissolution, transgranular versus intergranular fracture. The models are complementary and there is no consensus on the best approach. In general a mechanistic understanding is still lacking and models that account for the microstructure, in particular grain boundaries, including the transport of embrittling atoms, are needed.

### **Gas-cooled systems**

For applications in GFR, the candidate materials (austenitic and F/M steels as well as Ni-based alloys) need qualification at high temperature (see previous section) in contact with flowing He.

The alloy **corrosion behaviour depends to a large extent on the relative impurities level** in the He coolant and whether **oxidation, carburization or de-carburization** occurs.<sup>20,21</sup> Carburization is linked to loss of ductility whereas decarburization reduces the creep rupture strength. The effect is however much smaller than for LME. Ideally a continuous passivating oxide layer develops which provides corrosion resistance. Due to high dilution of gaseous species, the He-coolant is not in thermodynamic equilibrium. This and the fact that the alloy composition changes makes the prediction of the conditions for stable oxide formation very difficult. Furthermore, the **possibility of environment and high stress synergism on corrosion and crack formation** should also be investigated. To understand the corrosion mechanisms and their impact on materials properties and how they are affected by alloy and gas composition requires **test programme complemented by physical and empirical models**. The approach to demonstrate the suitability for candidate materials in high-temperature He is similar to the HLM outlined above. However, due to the higher temperatures of GFR, **coupling with creep and creep-fatigue** becomes more important. Moreover, the **number of candidate materials** with different features is higher than for LFR, which means that **screening is needed**. For example, the alloy 800H is a qualified material, but it is relatively weak in strength at high temperature (above 750°C), exhibiting also a significantly higher coefficient of

<sup>17</sup> Zhang et al., Review - Models of liquid metal corrosion, J. Nucl. Mater. 404 (2010) 82.

<sup>18</sup> Zhang, A review of steel corrosion by liquid lead and lead-bismuth, Corrosion Science 51 (2009) 1207.

<sup>19</sup> See overview articles; M.H. Kamdar, Liquid Metal Embrittlement, Treatise on Materials Science and Technology, edited by Academic Press 25, 1983, 361-459; P.J.L Fernandes and D.R.H Jones, Mechanisms of Liquid metal induced embrittlement, International Materials Reviews, 42, 1997, 251-261; B. Joseph, M. Picat, and F. Barbiera, Liquid metal embrittlement: A state-of-the-art appraisal, Eur. Phys. J. AP, 5, 1999, pp 19-31

<sup>20</sup> Cabet et al., High temperature corrosion of structural materials under gas-cooled reactor helium, Materials and Corrosion 57 (2006) 147.

<sup>21</sup> Christ et al., High temperature corrosion of the nickel-based alloy Inconel 617 in helium containing small amounts of impurities, Mater. Sci. Eng. 87 (1987) 161.



thermal expansion compared to Inconel 617 and Haynes 230. This makes this alloy potentially susceptible to thermal fatigue. Therefore, a significant effort must be made to select other corresponding materials, i.e. Inconel 617,<sup>21,22</sup> and to develop data to support ASME code cases in order to extend its operating range.

## Irradiation effects

For irradiation effects it is necessary to distinguish between fuel cladding, necessarily exposed to high irradiation dose rates, and structural components that are exposed to very low irradiation fluxes over long periods of time.

In the RCC-MRx code, irradiation of **structural materials** is taken into account by very basic design rules for tensile strength, uniform elongation, fracture toughness and the elastic allowable stresses. There are two essential border lines: the **negligible irradiation curve** that defines conditions for absence of significant irradiation effects, and the **maximum allowable irradiation**. If negligible irradiation conditions are fulfilled, then radiation effects can be ignored. For values in-between, the irradiation effects need to be taken into account.

Nuclear reactors are designed so that all structural components should remain in the negligible irradiation regime. The irradiation data, however, are only provided for a small number of materials, and even when known the negligible irradiation curve and maximum allowable irradiation are based on a rather limited number of tests of irradiated materials. In RCC-MRx, the negligible and maximum allowable irradiation damages in the temperature range 425-550°C for 316L(N) are 2 and 24 dpa, respectively. These values are based on **irradiation hardening and embrittlement**. There is some concern that **with 60-years operational life low dose may have significant effects due to prolonged exposure and thus helium production induced by transmutation**. As a matter of fact, He is essentially insoluble in metals and the entrapped He from transmutation tends to precipitate as nano-scale bubbles at grain boundaries. At **elevated temperatures**, helium bubbles grow rapidly under the influence of both temperature and stress. The **growth of grain boundary helium bubbles may result in the weakening of the grain boundaries and intergranular fracture**, leading to severe embrittlement in the materials.

For **fuel claddings** the main concerns are **irradiation creep and swelling**, the latter also related to He production. These two processes are less problematic in F/M than in austenitic steels. However, in the case of **F/M steels**, not yet fully codified in RCC-MRx, **low temperature (<350°C) radiation embrittlement** may lead to a significant shift of the ductile-brittle transition temperature (DBTT) already after fractions of one dpa, and potentially to severe **loss of elongation due to plastic flow localisation** may ensue.<sup>23</sup> The latter requires intense modelling effort to be fully understood, so as to justify a revision and extension of design rules for F/M steels.

Another important aspect to be quantified under irradiation is the possible influence on the response of materials to coolant exposure. **Synergistic effects may exist that exacerbate corrosion, dissolution, or LME.**<sup>24,25</sup> These effects need to be quantified and taken into account, possibly at the level of design codes and certainly at the level of component lifetime management.

In summary, for structural materials pre-normative research may be needed to:

- Expand the class of materials for which irradiation data are provided;
- Refine the irradiation rules by design by analysis, for instance by support of mechanistic models for plastic flow localization and irradiation embrittlement;
- Quantify the effects of helium production long-term low dose exposure;
- Quantify the potentially synergistic effects between irradiation and corrosion.

<sup>22</sup> Christ et al., Oxidation of Metals, 30 (1988) 27.

<sup>23</sup> See e.g. Klueh and Nelson, Ferritic/martensitic steels for next-generation reactors, J. Nucl. Mater. 371 (2007) 37.

<sup>24</sup> Stergar et al., LEXUR-II-LBE an irradiation program in lead-bismuth to high dose, J. Nucl. Mater. 450 (2014) 262.

<sup>25</sup> Magielsen et al., Irradiation of structural materials in contact with lead bismuth eutectic in the high flux reactor, J. Nucl. Mater. 415 (2011) 311.



How to translate test data and mechanism-based irradiation models to engineering tools remains largely an open question.

## Assessment of complex loadings

All structural components in a nuclear reactor are exposed to complex loadings. A good **description of the loads and the computation of the stress and strain distributions**, which are directly related to the constitutive models, are **necessary to predict safety margins, evolution of damage and remaining lifetime of components**. Typical complex loadings include:

- Multiaxial stress distributions are the norm whereas mechanical properties are generally based on uniaxial tests. In particular very complex stress fields are found in welded joints.
- Non-isothermal thermo-mechanical loads. These are caused by temperature variations on the surface and the constraint imposed by the component. Temperature gradients give large variations stresses and strains through the component wall thickness that depend on frequency and amplitude of the thermal loads.<sup>26</sup>
- Load transients and beyond design conditions. These are characterized by load transients and temperature excursions.

To address complex loadings correctly the following **pre-normative activities** are required.

- Development of constitutive models and governing equations that include temperature variations, strain rate and other dynamic effect dependence and constraint effects. Their calibration requires specific material characterization test programmes.
- Transferability between specimen and component tests. All standard tests, which are the basis for all constitutive models, are designed to give simple and well-defined loadings. Tests that include complex loading effects in components, i.e. **component tests**, are needed to demonstrate transferability between standard tests data and component assessment. Such tests include for instance bi-axial tests and thermal shock tests.

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## Integrity and qualification of weldments and welded components

The operating experience of all nuclear reactor types clearly demonstrates that welds are the weak spots in metallic components. Testing and assessment of welds and welded components must thus be an integrating part of pre-normative research programmes for GenIV conditions.

Welds are very complex as they include at least two materials and the welded microstructure will depend on the energy supplied during welding, and is thus linked with process. Moreover the microstructure and the local material properties vary across the weld.<sup>27</sup>

**Welds will always contain defects**, the propagation of which will determine the lifetime of the welded component: **defect assessment through inspection and non-destructive examination can reduce costs significantly and increase safety** (see page 9, component and material health monitoring).<sup>28</sup>

**Welding will also introduce internal residual stresses**. These have been the cause of many in-service failures and thus cannot be ignored by designers. Informed design of **post-weld heat treatment to minimize residual stress and optimize properties and microstructure is especially important**. Another problem is that it is notoriously difficult to quantify residual stresses in components by experiments or modelling. **Residual stress measurements in welded components** by neutron diffraction as well as other simpler methods **need to be pursued**. In parallel **models** that predict

<sup>26</sup> Nilsson et al., Assessment of thermal fatigue life for 316L and P91 pipe components at elevated temperatures, Engineering Fracture Mechanics 168 (2016) 73.

<sup>27</sup> For an overview of defect assessment methods for welded components see Comprehensive Structural Integrity, Fracture of Materials from Nano to Macro, Vol. 7, Practical Failure Assessment Methods, 2007, Elsevier-Pergamon. Defect assessment is included in Appendix 16 of RCC-MRx and at greater depth in the R6 and R6 defect assessment codes.

<sup>28</sup> Defect assessment is included in Appendix 16 of RCC-MRx and at greater depth in the R6 and R6 defect assessment codes.

residual stresses with higher predictability by simulating the physical processes also **need to be further developed**.

Since the properties and the behaviour of welded components depend strongly on the welding process it is essential that **welded components in test programmes are manufactured in accordance with code requirements**, which for the GenIV reactors correspond to RCC-MRx.

Other points:

- **Novel welding technologies** such as electron beam, friction stir or laser have considerable promise, but must be thoroughly understood and characterized before use by pre-normative research.
- **Design rules** are based on **weld factors for creep and fatigue**. It should be verified if the same weld factors can be used for creep-fatigue.
- Weldment design codes must consider through-life degradation in an informed manner, based upon service experience and advanced assessment procedures. It is not possible to test for 60 years prior to entering service, long-term exposure to aggressive environments or neutron irradiation, so physically based **mechanistic understanding of degradation and failure mechanisms** for all regions of the weld is vital, to allow prediction of service exposed behaviour.

### Sub-size and miniature specimen test standardization

Tests methods and associated procedures and standards for very small volumes of material using small and miniature specimens<sup>29</sup> become essential for:

- Neutron irradiated materials available in limited quantity, or in order to minimize the radioactivity;
- Very thin material layers that are affected by charged particle irradiation (typically a few  $\mu\text{m}$ );
- Service-exposed materials, i.e. tested in service during inspections;
- Thin-walled cladding tubes;
- Evaluation of local properties, for instance heat affected zone of a weld (mm scale), coatings, single crystals ( $\mu\text{m}$  scale), or graded functional materials.

Specimens involved range from those for sub-sized standard and less conventional miniature tests, complementary to the standard specimen and component tests, to truly miniaturized samples:

- **Miniaturised specimens for standard mechanical property assessment.** This type of testing involves two types of issues. Firstly, testing setup and specimen preparation need to be modified to adapt them for small specimens. Secondly, modified evaluation procedures have to be assessed and validated by comparison with larger specimen data, for full transferability of test results. In particular, the issue as to whether these specimens can be used to predict real component behaviour is addressed. Validation of specimen geometry and testing procedure should be undertaken by inter-laboratory exercises as a first step to achieving the standardization of the procedures. One final goal is the determination of the fracture toughness and crack growth rates.
- **Thin-walled cladding tube tests.**<sup>30</sup> This can include tests of different sizes such as small-punch, ring-compression, pressure and cone-mandrel. Since fuel-claddings become highly irradiated, focus should be on testing irradiated material in hot cell conditions. The different tests are complementary, so there is a need to identify the strengths and limitations of each method, and to provide recommendations on procedures and then develop standards on how to perform the tests and evaluate the data. In particular, the **small punch** test is now becoming widespread, also for other applications, due to its relative simplicity in preparing specimen and performing tests, but the evaluation of the test result is complex and how to extract material properties is not obvious.

<sup>29</sup> Hyde et al., Requirement for and use of miniature test specimens to provide mechanical and creep properties of materials: a review, *Int. Materials Reviews* 52 (2007) 213.

<sup>30</sup> Holmström et al., Test methodologies for determining high temperature material properties of thin walled tubes, JRC Technical Reports: EUR 28642 EN 2017, Publications Office of the European Union, Luxembourg,

- **Micro- and nano-mechanical testing methods.**<sup>31</sup> These include nano-indentation, as well as compression, tension and bending tests on FIB-produced micropillars, where in-situ approaches combine mechanical testing with advanced imaging characterization. Such methods allow single grain properties needed for crystal plasticity models to be measured. Testing of bi-crystal specimens can be used to study grain boundary strengths. Nano-indentation allows local hardness variation to be mapped, as tool to understand dislocation dynamics. They all can be applied to both neutron- and ion-irradiated materials and may foster the exploitation of ion irradiation as a surrogate of neutron irradiation. Research is needed to i) further develop high-temperature procedures for nano-hardness measurements, and ii) elaborate test and evaluation procedures for micro-pillar compression tests, including identification of material properties that can be extracted.

There are some restrictions to the applicability of miniature test methods. Creep properties have been studied by small punch test, but the tests are restricted to high creep rates. The possibility to derive fracture and in particular fatigue properties is quite limited. The methods are presently mainly aimed for screening and ranking of materials. To what extent design specific material properties (e.g. tensile curve, creep curve, fracture toughness) used in design rules or design by analysis can be extracted is an open issue.

An exception are sub-sized tensile specimens, from which tensile properties, except total elongation, can be accurately obtained: except for total elongation, sub-size samples are already included in ASTM and ISO standards. The use of sub-size specimen for Charpy, fatigue, creep and fracture mechanics tests (fracture toughness and crack growth), in contrast, still needs to be included in the standards.

In summary **for miniature tests there is a need to review in depth the test methods in terms of strength, weaknesses and restrictions on providing material data.** This should be followed by the development of common procedures and standardization of these tests. For the sub-sized tests, the emphasis should be on fracture and fatigue properties. The standardization work must go through the involvement of national and international standard committees with participation of the main testing laboratories and designers. In the case of small punch tests, some activities have already started in Europe (ECISS<sup>32</sup>) and USA (ASTM), mainly with international inter-laboratory studies.

### Component and material health monitoring

Non-destructive examination (NDE) of components is crucial to verify that components are performing according to expectations and to detect well ahead of time cracks that potentially could lead to failure. However, it is not obvious how to deduce precise information on microstructural evolution and associated changes in mechanical properties from NDE. The approach today is thus to monitor the material degradation by analysing surveillance specimen exposed to in-service conditions by mechanical tests and, if required, microstructural analysis.

Typical methods used for NDE are ultrasound, X-rays, Foucault or eddy currents and thermoelectric power. Ideally it would be desirable to be able to deduce from these techniques not only information about cracks, but also about how the material has deformed and its microstructure has evolved, possibly by identifying signatures that correspond to situations that deserve attention. The current industrial tendency is towards online materials monitoring and lifetime estimation based on NDE, possibly applying artificial intelligence-based big data analysis to derive patterns that are capable of giving warning sufficiently in advance of the possibility that the component may fail.<sup>33</sup> Clearly the GenIV nuclear reactor environment, with components immersed in e.g. liquid metals and exposed to irradiation at high temperature, is not ideal for online component monitoring. However, it is

<sup>31</sup> Hosemann et al. An exploratory study to determine applicability of nano-hardness and micro-compression measurements for yield stress estimation, *J. Nucl. Mater.* 375 (2008) 135; Uchic, Application of micro-sample testing to study fundamental aspects of plastic flow, *Scripta Materialia* 54 (2006) 759; Heintze et al., Ion irradiation combined with nanoindentation as a screening test procedure for irradiation hardening, *J. Nucl. Mater.* 472 (2016) 196.

<sup>32</sup> ECISS TC 101 WG 1 on Small Punch standard is now under review.

<sup>33</sup> Chang et al., *Inventions* 3 (2018) 41; Liang et al. *Digital Comm. & Networks*, 2 (2016) 97.

considered that exploring this possibility, starting from non-destructive tests for the controllability of materials, would provide a clear added value in terms of reduction of conservativeness in design coupled to higher safety standards.

### TA.1.1.2 Advanced structural materials modelling and characterization

The issues to be addressed with an ICME approach are necessarily strongly linked to those addressed in pre-normative research (previous section), as they correspond to the main degradation processes that affect structural materials for GenIV systems. Since the early 2000's, several European projects have addressed from a modelling point of view material degradation processes connected to irradiation, such as radiation hardening and embrittlement (in bainitic, austenitic and ferritic/martensitic steels), as well as, to a certain extent, plastic flow localisation and irradiation assisted stress corrosion cracking in water, namely FP6/PERFECT,<sup>34</sup> FP7/GETMAT,<sup>35</sup> FP7/PERFORM60,<sup>36</sup> FP7/MatISSE.<sup>37</sup> Some of the relevant activities currently continue in the H2020/SOTERIA<sup>38</sup> and the H2020/M4F<sup>39</sup> projects. Issues connected with the modelling of the high temperature behaviour of F/M steels have also been partially addressed in FP7/MATTER<sup>40</sup> and FP7/MatISSE, while the modelling of oxidation and dissolution, as well as prolonged irradiation, in austenitic steels is being partly addressed in H2020/GEMMA<sup>41</sup>. In general, several modelling activities are included in EERA JPNM pilot projects. All these projects provide solid bases on which to progress further, both in terms of modelling techniques that have been developed and results obtained, as well as in terms of experimental work, performed with a view to understanding mechanisms and for model validation. However, the development of models necessarily proceeds from simpler to more complex materials and issues, therefore not only is there still a lot of room for the improvement of existing models and several open issues to be addressed, but there are in fact cases where no defined computer-simulation based physical modelling approach has been identified yet, beyond classical continuum approaches.

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The most important issues to be addressed are described in what follows.

#### Microstructural/microchemical evolution

Irradiation produces damage in materials through processes of interaction between energetic subatomic or atomic particles with the atoms and molecules that form the target. Therefore, in the case of nuclear materials, which have irradiation as their most specific origin of degradation, **large effort needs to be devoted to the development of models that describe the material at the microscopic submicroscopic and atomic scale**, accompanied by relevant modelling-oriented experiments.

Modelling the microstructural and microchemical evolution under irradiation and/or while exposed to high temperature is essential in order to understand, among others, the following key degradation processes:<sup>42</sup>

- The kinetics of formation of **microstructural features that are responsible for radiation-induced hardening and embrittlement**<sup>43</sup> of steels, and metallic materials in general, including loss of

<sup>34</sup> FP6 IP PERFECT Project: Prediction of Irradiation Damage Effects in Reactor Components, Journal of Nuclear Materials – Special Issue, vol. 406, issue 1 (2010).

<sup>35</sup> [http://www.iaea.org/inis/collection/NCLCollectionStore/\\_Public/46/040/46040904.pdf?r=1](http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/46/040/46040904.pdf?r=1)

<sup>36</sup> <http://www.sciencedirect.com/science/article/pii/S0029549311001804?via%3Dihub>;  
<http://proceedings.asmedigitalcollection.asme.org/proceeding.aspx?articleid=1628620>;  
<https://www.openaire.eu/search/project?projectId=corda::190daa7e4bf2ff682991a1c9e23ccfd2>.

<sup>37</sup> [www.fp7-matisse.eu](http://www.fp7-matisse.eu); <http://www.fp7-matisse.eu/wp-content/uploads/2015/12/MatISSE-2015-GEN-IV-materials-Holmstrom.pdf>

<sup>38</sup> [www.soteria-project.eu](http://www.soteria-project.eu)

<sup>39</sup> <http://www.h2020-m4f.eu/>

<sup>40</sup> <http://www.eera-jpnm.eu/?q=jpnm&sq=nboard>;  
<http://proceedings.asmedigitalcollection.asme.org/proceeding.aspx?articleid=1627574>

<sup>41</sup> <http://www.eera-jpnm.eu/gemma/>

<sup>42</sup> A summary of key irradiation effects in structural materials can be found in, e.g.: Zinkle, Phys. Plasma. 12 (2005) 058101; Zinkle and Was, Acta Mater. 61 (2013) 735.

<sup>43</sup> Monnet, J. Nucl. Mater. 508 (2018) 609.

**elongation due to plastic flow localisation**,<sup>44</sup> at temperatures  $<0.3-0.4T_m$ , (where  $T_m$  is the melting point in K). These phenomena are often the main lifetime limiting factors for nuclear components and affect materials already after irradiation to very low dpa.

- The kinetics of **formation of voids and the onset of radiation-induced swelling** of steels,<sup>45</sup> and metallic materials in general, at intermediate temperature ( $>0.3-0.4T_m$ ), above a threshold dpa dose, and the steady-state swelling rate above such dose. In particular, predicting the **threshold dose for steady-state swelling onset is crucial**, because this dose limits the lifetime of cladding materials, thereby affecting the maximum burnup that can be reached. This dose is very challenging to predict: it may vary between a few to hundreds of dpa, depending on several variables, not only material type and composition, but also temperature, dose-rate, and material history.
- **Segregation and/or precipitation of chemical species, including transmutants like He, at extended defects** (dislocations, grain boundaries) induced by irradiation or thermal ageing in steels, and metallic alloys in general.<sup>46</sup> These processes may significantly change the property of the materials, influencing their response in terms of mechanical properties (hardening, loss of grain boundary strength and promotion of intergranular fracture, ...), but also of reduced resistance to corrosion and oxidation in contact with coolants. Irradiation environments may trigger segregation and precipitation also against thermodynamic equilibrium, making them difficult to predict.
- The **stability and size distribution evolution of carbides and oxides**<sup>47</sup> in stabilized or strengthened steels, and metallic alloys in general, during material fabrication, thermal treatment, thermal ageing, and/or irradiation. Obtaining and maintaining under operation a microstructure characterized by fine and uniform dispersions of particles is indeed key to guarantee the integrity of components at high temperature for sufficiently long time.

Although significant advances have been made in the development of microstructural and microchemical evolution models in the last couple of decades, multiple challenges remain to be addressed in order to develop reliable ICME approaches that provide truly predictive capability. These essentially aim at being able to **describe, with a single tool, the evolution of both the microstructure (redistribution of lattice defects) and the microchemistry (redistribution of chemical species)**, be this based on kinetic Monte Carlo,<sup>48</sup> cluster dynamics<sup>49</sup> or phase field<sup>50</sup> approaches. Among others, the following modelling developments need to be pursued:

- Develop methods to simulate **radiation damage production in collision cascades** using atomistic simulation methods such as molecular dynamics<sup>51</sup> not only in pure elements or simple compounds,<sup>52</sup> but also for complex alloys, **including all involved interactions**, e.g. phonon-electron.<sup>53</sup>
- Develop **atomistic models allowing multi-million atom simulations for chemically complex systems** such as for example steels. While ideally interatomic potentials fitted to electronic structure calculations with density functional theory (DFT)<sup>54</sup> would be desirable,<sup>55</sup> for many applications rigid lattice models based on cluster expansion concepts may be comparatively simpler to fit to DFT.<sup>56</sup>

<sup>44</sup> Farrell, Byun and Hashimoto, J. Nucl. Mater. 335 (2004) 471.

<sup>45</sup> Garner, "Radiation Damage in Austenitic Steels", Comprehensive Nuclear Materials, vol. 4 (2012) 33.

<sup>46</sup> Thuin et al. Comp. Mat. Sci. 149 (2018) 324.

<sup>47</sup> Wharry et al. J. Nucl. Mater. 486 (2017) 11-20

<sup>48</sup> Becquart et al. J. Nucl. Mater. 406 (2010) 39; Soisson et al. ibid. p. 55.

<sup>49</sup> Reinhard and Suraud, Introduction to Cluster Dynamics, Wiley (2008).

<sup>50</sup> Tonk et al. Comp. Mater. Sci. 147 (2018) 353.

<sup>51</sup> [http://cms.sjtu.edu.cn/doc/reading/md/A\\_Molecular\\_Dynamics\\_Primer\\_\(Ercolessi\).pdf](http://cms.sjtu.edu.cn/doc/reading/md/A_Molecular_Dynamics_Primer_(Ercolessi).pdf).

<sup>52</sup> K. Nordlund et al. "Primary Radiation Damage in Materials", NEA/NSC/DOC(2015)9, [www.oecd-nea.org/science/docs/2015/nsc-doc2015-9.pdf](http://www.oecd-nea.org/science/docs/2015/nsc-doc2015-9.pdf).

<sup>53</sup> Rizzi, Real-Time Quantum Dynamics of Electron-Phonon Systems, Springer (2018).

<sup>54</sup> Di Zhou, An Introduction of Density Functional Theory and its Application, <http://citeseerx.ist.psu.edu/viewdoc/summary?doi=10.1.1.569.2831>.

<sup>55</sup> Bonny et al., Modell. Simul. Mater. Sci. Eng. 26 (2018) 065014.

<sup>56</sup> Qu Wu et al., Comp. Mater. Sci. 125 (2016) 243.

- **Include the effect of applied loads and stress-strain states** as variables in microstructural and microchemical evolution models.<sup>57</sup> This has a two-fold application: on the one hand it is needed to describe the influence of stresses on the mobility and stability of radiation defects; on the other it is necessary in order to describe correctly the removal of radiation defects at sinks such as dislocations and grain boundaries, including preferential segregation of chemical species, as well as to describe processes such as heterogeneous nucleation of precipitates.
- Use DFT and other DFT-based atomistic models, e.g. based on cluster expansion approaches, to **go beyond and improve models based on the Calphad approach**<sup>58</sup>, for a full physically-based prediction of the phase diagrams for complex alloys and compounds, including kinetics of phase separation.
- Find suitable **compromises between atomistic details and larger scale descriptions**, in microstructural and microchemical evolution models, **to enable high irradiation doses and temperatures in sufficiently large volumes to be modelled**, given that current models have strong limitations in this sense.

**Machine learning schemes based on artificial intelligence**, e.g. artificial neural networks, could be of valuable help to address the modelling problems related with high chemical complexity, as well as to bridge between scales, by providing in a simplified form the input coming from a lower scale to a larger scale model.<sup>59</sup>

### Mechanical behaviour after and under irradiation

**Microstructural and microchemical evolution models** and the corresponding understanding should be used as **inputs to move to larger scales** and address, for example, the **prediction of the subsequent plastic behaviour**, particularly in tensile tests, as a function of materials type and composition, dose received, irradiation and test temperatures.

The issues to be addressed are, in particular:

- Prediction of **radiation hardening** (increase of yield strength and flow behaviour) and **embrittlement** (reduction of elongation) after irradiation below  $0.3-0.4T_m$ , due to the impediment to dislocation motion when load is applied, caused by radiation-produced defects.<sup>43</sup>
- Onset of **plastic flow localization**, also after irradiation below  $0.3-0.4T_m$ , with subsequent **loss of engineering work-hardening and uniform elongation**, and its impact on the overall mechanical behaviour:<sup>60</sup> this phenomenon is intimately connected with radiation hardening, but it is strongly influenced by the nature of the defects that irradiation produces and how they interact with dislocations (absorbable versus shearable). This radiation-specific effect needs to be correctly accounted for if appropriate design rules need to be developed, especially for F/M steels.
- Deformation at constant load due to **irradiation creep**, which occurs in a wide range of temperatures, including low temperatures, and is controlled by point-defect diffusion and unbalanced elimination of vacancies and self-interstitials at the dislocations, therefore differing substantially from thermal creep.<sup>61</sup>

Model developments are necessary to address these issues, essentially aiming at **describing in a single framework the kinetic evolution of the irradiation-induced and the dislocation microstructures, to deduce constitutive laws for crystal plasticity**<sup>62</sup> and **devise homogenization schemes**<sup>63</sup> for continuum mechanics approaches applied to components.

- As for microstructure/microchemical evolution models, **reliable atomistic descriptions of chemically complex systems applicable to multi-million system atoms are necessary to describe**

<sup>57</sup> Subramanian et al. Phys. Rev. B 87 (2013) 144107.

<sup>58</sup> [http://www.calphad.com/calphad\\_method.html](http://www.calphad.com/calphad_method.html); Schick et al. Calphad 37 (2012) 77.

<sup>59</sup> Castin et al. Comp. Mater. Sci. 148 (2018) 116.

<sup>60</sup> Doyle et al. J. Nucl. Mater. 499 (2018) 47.

<sup>61</sup> Matthews and Finnis, J. Nucl. Mater. 159 (1988) 257.

<sup>62</sup> Volegov et al., Physical Mesomechanics 20 (2017) 174 <https://link.springer.com/article/10.1134/S1029959917020072>.

<sup>63</sup> Allen, Comp. Sci. & Technol. 61 (2001) 2223.



**dislocations and grain boundaries, their mutual interaction and their interaction with radiation defects**,<sup>64</sup> if this interaction has to be representative of real situations. In this case, rigid lattice atomistic models are of no use and interatomic potentials remain to date the only tool.

- **A methodology to transfer systematically the above understanding to dislocation dynamics models applicable at single-crystal level including irradiation effects**, in the form of **local rules** needs to be established: tools exist,<sup>43,65</sup> but their application to a specific material requires extensive *ad hoc* calculations, and takes for granted a knowledge of the microstructural and microchemical changes induced by irradiation and their effects on dislocation motion, grain boundary properties, etc. It remains challenging in particular to introduce **chemical effects** (e.g. decorated dislocation loops or heterogeneously nucleated precipitates). In the case of (irradiation) creep, the coupling between microstructural and dislocation evolution needs to be especially effective because *both microstructures evolve simultaneously over time*.

Concerning **continuum mechanics**, in order to introduce the effect of the presence of boundaries between single crystals in the transfer of stress and strain benefit could be taken from a most widespread use of **strain gradient plasticity**.<sup>66</sup> Moreover, it is not yet completely clear up to what extent current **homogenization schemes** are able to fully transfer to component level the effects of irradiation or thermal ageing, including **local or localized effects**, such as for example plastic flow localization.

### Mechanical behaviour at high temperature

The model developments sketched above are potentially applicable also in the case of high temperature mechanical behaviour, mainly describable as **thermal creep**, associated or not with **cyclic loading**, i.e. **fatigue**, via **creep-fatigue interaction**. However in these cases **visco-plasticity comes into play** to describe the fact that materials at high temperature deform and lose strength also under constant load, through several mechanisms. These models are currently not developed to the same level of advancement as microstructure evolution or plasticity models.

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**Points of attention** for models of high temperature mechanical behaviour are the following:

- At high stresses and temperatures thermal creep is controlled by dislocation climb and glide; at lower temperatures and stresses vacancy diffusion (Nabarro-Herring creep)<sup>67</sup> or grain boundary sliding (Coble creep)<sup>67</sup>. Since **creep tests** are usually at **higher temperature or load** than in the operational conditions, **in the laboratory** creep is controlled by **dislocation climb** whereas **under operation conditions** the creep is governed by **diffusion**. So **extrapolation of creep rates from laboratory conditions to operational conditions is generally non-conservative**. The creep failure mechanisms are also different. To correctly extrapolate, all mechanisms need to be understood and modelled correctly.
- Crystal plasticity models so far are primarily limited to static tensile loads. In order to address cyclic loading and thus fatigue, however, **time-dependent crystal plasticity**<sup>68</sup> **models** need to be **developed and calibrated**.
- Separate engineering models are often developed for creep and plasticity but to understand for instance the **coupling between creep and fatigue**, **unified visco-plastic models** are needed. Examples of these models that include kinematic and isotropic hardening, memory and recovery effects exist<sup>69</sup> and allow features such as cyclic softening and hardening, strain rate effects, etc.,

<sup>64</sup> Liang Zhang et al. Comp. Mater. Sci. 118 (2016) 180.

<sup>65</sup> Vattré et al. J. Mech. & Phys. Solids 63 (2014) 491.

<sup>66</sup> Guha et al. Sadhana 40 (2015) 1205 <https://www.ias.ac.in/article/fulltext/sadh/040/04/1205-1240>.

<sup>67</sup> Kassner, Fundamentals of Creep in Metals and Alloys, Elsevier, Second Edition (2009); Riedel, Fracture at High Temperatures, Springer Verlag, Berlin (1987).

<sup>68</sup> Crystal plasticity is formulated as rate equations, i.e. time effects are included in the formulation. The problem of the time-dependent application is much related to the calibration and the fact that computational times get excessively large. Full-field modelling (individual grains modelled in a polycrystal) is generally not possible. Most work is for single crystals. Homogenization is often used. See e.g. Dunne and Petrinic, Introduction to Computational Plasticity, Oxford University Press (2005); Roters et al., Overview of constitutive laws, kinematics, homogenization and multiscale methods in crystal plasticity finite-element modeling: Theory, experiments, applications, Acta Materialia 58 (2010) 1152.

<sup>69</sup> Chaboche, A review of some plasticity and viscoplasticity constitutive theories, International Journal of Plasticity 24 (2008) 1642.



to be accommodated. However, the application is limited due to **difficulties in the calibration of model parameters** with experiments and in the **implementation of models into finite element codes**, requiring large computational resources.

It is important to emphasise that visco-plasticity approaches such as those sketched above assume that continuum mechanics equations, solvable with finite element methods, can be actually written. However, **for several engineering applications connected with, in particular, high temperature operation, and therefore affecting crucially GenIV systems, finite element models may not be usable**. This generally happens because there is no sufficient understanding of the underlying processes to be able to elaborate a conceptual model, which is an essential guide to develop a consistent integrated modelling approach. In these cases, where only or mostly empirical correlations for quantities of engineering interest exist, appropriate microstructural examination together with the (generally semi-empirical) identification of the important variables are of essential use to cast some light onto the fundamental mechanisms and eventually guide an integrated modelling approach. Specifically **in the case of total creep-fatigue life assessment, only semi-analytical engineering models currently exist**, because the underlying processes are not yet clearly identified or are so complex that it is difficult to separate variables and effects. It would be desirable to have more robust, ICME-type models also for these applications, but to date building reliable models of this type for these processes remains an open challenge that needs to be addressed.

### Fracture mechanics

All materials contain defects, i.e. microcracks from where cracks may initiate and then propagate to failure. In design codes the assumption is normally that the components are defect free, but for the **structural integrity assessment** of components the existence of **microcracks** needs to be accounted for, especially for **welded components**. Existing models need to be improved and further validated, but, importantly, **non-destructive examination techniques are key** in order to get a picture of the initial situation of a material in terms of pre-existing (or developed) microcracks.

**Neither plastic nor visco-plastic models explicitly account for** the degradation that leads to final failure by **fracture**.<sup>69,70</sup> Degradation can be incorporated through damage models by introducing additional state variables into the thermodynamic framework. In this way, by integrating the effect of pre-existing damage, e.g. initiation, growth and coalescence of voids and micro-cracks, into the constitutive continuum mechanics models, the evolution from the virgin state to macroscopic crack initiation should become possible. This implies including criteria for damage on ideally observable quantities (e.g. crack length, crack density).

**Crack initiation and propagation** are cases of processes for which conceptual models to guide an ICME approach do exist, but the variability of possible situations is so large, the scales simultaneously involved so different, and the phenomena so stochastic in nature, that integrated modelling approaches are inherently difficult to develop and apply. Here too, appropriate **microstructural examination** for the identification of the important variables, although challenging, because they involve the direct observation of loaded materials, are of essential use to cast some light onto the fundamental mechanisms and eventually guide an integrated modelling approach.

The **propagation** of a given **crack** can be relatively well described for simple loading conditions. However, in the case of more complex loadings, for instance combination of creep and fatigue, non-proportional loading, thermo-mechanical loads and mixed-mode (combination of normal and shear stresses) the predictability is much more limited.

Fracture mechanics models can be generally separated into two classes: (i) **classical fracture mechanics**<sup>71</sup> where crack propagation is governed by a crack tip parameter such as stress intensity

<sup>70</sup> McDowell, Viscoplasticity of heterogeneous metallic materials, Materials Science and Engineering R 62 (2008) 67.

<sup>71</sup> Anderson, Fracture Mechanics Fundamentals and Applications, Second Edition, CRC Press LLC, 1995; Milne, Karihaloo and Ritchie, Comprehensive Structural Integrity – Fracture of Materials from Nano to Macro, Elsevier, 2003 (updated 2007).

factor, J-integral or C-integral, and (ii) **local approaches**<sup>72</sup> where the fracture processes are specifically modelled, e.g. void growth and coalescence for ductile tearing. While models of the first type of models are the most used and widespread, especially at industrial level, it is desirable, especially to address the problem of irradiated materials, that **local approach models should be more developed**.

### Coolant compatibility models

The processes that govern the **interaction between solids and coolants** are crucial in the context of GenIV reactors and need to be appropriately modelled beyond current capabilities. Basic equations for mass transport in the liquid and the solid, as well at the liquid/solid interface can be written and solved under appropriate assumptions with simple boundary conditions.<sup>17,18</sup> However, the understanding and detailed description of the origin of the processes observed through microstructural examination calls for more detailed approaches, involving the treatment of atomic-level interactions as driving force for complex interplays between liquid and solid. A full description of these processes requires the **identification of new appropriate modelling approaches**, because the complexity of the relevant physical and above all chemical processes challenges the possibilities offered by existing simulation tools.

The model developments sketched above for microstructural/microchemical evolution under irradiation largely apply here as well and the tools currently available are essentially the same. However **the level of chemical and structural complexity is, in this case, higher, even for the simplest model systems that can be devised** (e.g. Fe or Fe-Cr solid substrate and  $Pb_{liq}$ -O as liquid). Models should strike a balance between complexity and level of detail that is not easily identified. Atomistic models can provide clues about mechanisms, but the scales involved and the simultaneous presence of several different phases make the application of purely atomistic approaches very challenging and ultimately probably impossible. Thus atomistic models should inform and match suitable thermodynamic models, for example based on the phase field formalism, describing the **stability of oxide scales**, together with kinetic models describing the processes of **oxidation and dissolution** in, especially, **heavy liquid metals** ( $Pb_{liq}$  and its alloys). A proper and fully defined integrated approach is still missing.

Especially challenging is the problem of **liquid metal embrittlement** (LME) which requires models for the solid-liquid interaction and for the embrittlement and their coupling. The degree of the embrittlement and the fracture mechanism depend on the specific liquid/solid couple. Although LME reduces the ductility at the macro-scale, the actual embrittlement mechanisms operate at the atomistic scale. Different models, all based on fracture analysis, have been proposed in the last decades: Adsorption Induced Reduction in Cohesion Model; Adsorption Enhanced Dislocation Emission Model, Dissolution-Condensation Mechanism and Grain Boundary Embrittlement and Penetration models. These models assume different mechanism but no model can reproduce all experimental observations, which indicates that LME is a multi-physical phenomenon, and the models are complementary. One approach could be to combine models describing competing mechanisms. Recent development for atomic-scale mechanism, grain boundary penetration, and crystal plasticity should be considered to further develop LME models. Any model development for specific liquid/solid combination needs to be closely integrated with microstructural analysis to establish the specific degradation mechanism, The development of mesoscopic models for these phenomena, rooted in a solid description of atomic-level interactions and mechanisms and linkable to the macroscopic mechanical response, remains an open challenge that needs to be addressed.

### Models in support of the use of charged particle irradiation

Neutron irradiations are long and expensive experiments and access to the relevant facilities is generally restricted. In a context of **modelling-orientated experiments** on radiation effects, but also

<sup>72</sup> A. Pineau, Development of the local approach to fracture over the past 25 years: theory and applications, International Journal of Fracture 138 (2006) 139–166.

of **screening of different routes to more radiation-resistant materials**, charged particle irradiation (ions, protons, electrons, ...) is a very valuable and affordable tool. However, transferability issues exist between charged particle and neutron irradiation environment (see also section TA.6.1.3).<sup>73</sup> To design and interpret correctly the results of these experiments, it is **essential that suitable models address the specificities of these modes of irradiation, which differ in many respect from neutrons**. Developing models of this type beyond the use of standard binary collision approximation models coupled to ion stopping power calculations to draw the profile of ion penetration in terms of displaced and injected atoms,<sup>74</sup> remains an open task.

While the scheme sketched above for microstructural/microchemical evolution under irradiation remains broadly valid, **ad hoc solutions need to be devised to take into account the specificity of charged particle irradiation**, namely: limited damage penetration and presence of close-by surface, damage profile and gradient, injected interstitials, effectively pulsed irradiation, possible self-annealing effects, injection of unwanted species ...

Moreover, a strong limitation of charged particle irradiation experiments is that **the quantity of material affected is very small, and yet not uniformly irradiated**. This introduces specific difficulties in the PIE. Advanced micro-specimen fabrication techniques for microstructural characterization, based on the use of focused ion beams (FIB) to select specific regions, are needed. More importantly, it is impossible to perform a standard evaluation of mechanical properties. It becomes therefore **essential to develop the capability of obtaining meaningful assessments of mechanical properties using alternative, microprobing techniques**, such as nanoindentation, small punch, micropillars, ... . This capability should also go hand-in-hand with suitable mechanical behaviour models, to be able to interpret correctly the results of the experiments.

Overall, the **goal** is to develop an established and possibly **standardized methodology**, based on the guidance of **models**, to perform **charged particle irradiation and relevant PIE, including mechanical characterization**. This is not expected to be suitable for the qualification of materials for pre-normative purposes, but should at least provide robust support for the development of models, ranging from microstructural evolution to mechanical behaviour. It should also allow different possible materials to be screened in terms of response to irradiation, limiting neutron irradiation to the most promising ones.

### Development of standard methodology for model validation

The validation of models is a complex and costly task that requires specific attention also from a methodological point of view. In fact, validating a model means mainly reducing the uncertainties on all fronts, from model parameters to experimental error bar so that only well-identified sources of uncertainty remain and an error can be associated with the model outcome. For this purpose, a number of actions are needed:

- Experiments aimed at **validating models** need to expose and then characterise materials with a view to **identifying mechanisms**, thus in them the **variables** need to be **separated**. **Model** materials are as important as **technological materials** and the characterization needs to combine several techniques, as only their combination can provide a sufficiently global and complete picture to allow comparison with all the model results.<sup>75</sup>
- The way of applying microstructural examination techniques (TEM, APT, SANS, PAS, ...), as well as micromechanical characterization methods (nanoindentation, micropillars, ...), and the way of analysing the outcome of the examination, is currently not standardized. This creates difficulties when collecting and putting together results from different laboratories. Thus **protocols** for the execution of **microstructural examination and the analysis of the data** need to be established, to enable inter-laboratory comparison.

<sup>73</sup> Was, J. Mater. Res. 30 (2015) 1158.

<sup>74</sup> SRIM code: <http://www.srim.org/>; MARLOWE code: Robinson, Nucl. Instr. & Meth. B. 67 (1992) 396 and Robinson, Phys. Rev. B 9 (1974) 5008.

<sup>75</sup> Hernandez-Mayoral et al. GETMAT (FP7-212175) D4.9 deliverable (2013).

- To facilitate experiment/simulation comparison, **methods that enable the simulation of the signal that the experimental techniques provide**, for instance how a given microstructural feature as simulated is going to appear to APT or TEM or PAS, need to be developed. They exist only in a few cases and their use is not sufficiently widespread.<sup>76,77</sup>
- **Protocols according to which the comparison between simulation and experiment should be performed**, in terms of which quantities can be safely compared and using which criteria, need to be identified.
- A methodology to identify the main **sources of uncertainty** and the error bar associated with the application of a model should also be established.

## Other issues

Quite specific issues for which not much has been done in terms of modelling, but that deserve mention, concern the problem of assessing the residual stresses in welded zones and the problem of the deformation and fracture modes of steels in general and specifically ODS steels.

## Note on materials of interest and different levels of model development

It is important to emphasise that **the level of development of advanced models is not equal for all materials of interest for GenIV reactors**. For example, even if austenitic steels are currently the candidate materials for GenIV prototypes and demonstrators, therefore corresponding to the shortest-term needs, **the development of microstructure evolution physical models for austenitic steels is much less mature than for F/M steels**. The reason is a combination of historical factors and of ease of application of the available modelling techniques. The crucial importance of radiation-induced embrittlement of the steels used to fabricate the pressure vessel of current GenII/III nuclear reactors focused a lot of effort on the development of models dedicated to these materials and that problem, which have very much in common with ferritic-martensitic steels. Importantly, both reactor pressure vessel steels and F/M steels can be modelled as body-centred-cubic iron alloys, starting from Fe or Fe-C as model material, and adding later the effect of other alloying elements, which is an especially robust way of progressing in model development. The other reason for austenitic steels lagging behind is that they are concentrated alloys that are not stable at 0 K and are characterised, moreover, by a complex magnetic state. They thus prove to be quite challenging materials to be modelled using atomic-level calculation and simulation tools. Equally challenging is the connected development of physics-based models to deal with microstructural and, above all, microchemical evolution. In turn, however, plasticity models suitable to address the mechanical behaviour from the mesoscopic to the macroscopic scale are somewhat more evolved in face-centred-cubic crystallographic structures (such as austenitic steels).

This situation has two consequences:

- **The modelling of F/M steels is certainly worth being pursued** not only because these materials remain important in the long-term for GenIV applications, but also because they represent a sort of reference case to be used as example and starting point for the development of models for other, atomistically more complex, materials, including austenitic steels. In addition, problems such as cyclic softening or liquid metal embrittlement seem to be specific of these steels and require to be addressed, in order to guide the qualification of these materials for future reactors.
- **The modelling of austenitic steels needs now to be pushed forward**, in such a way that, by “catching up” with the level of maturity that characterises models for F/M steels on the low-scale side, it can then benefit from the more advanced level that characterises the modelling of face-centred-cubic alloys, when it comes to the scale of dislocations and continuum mechanics, thereby being accelerated.

Since the development of models, in particular at the low-scale, necessarily proceeds from simpler to chemically more complex systems, F/M steels represent also the starting point also to develop

<sup>76</sup> Barthel, Ultramicroscopy 193 (2018) 1-11; Schäublin and Stadelmann, Mater. Sci. & Eng. A 164 (1993) 373.

<sup>77</sup> Hyde et al. Microscopy and Microanalysis 23 (2017) 366.

models of use to simulate **ODS and/or alumina-forming alloys** (FeCrAl), with a view to optimizing them. In the case of ODS, specific problems need to be addressed in terms of mechanical behaviour concerning their modes of deformation, especially at high temperature, and residual stresses after the fabrication process. On the other hand, high-entropy alloys, for which modelling support could be interesting in order to streamline among the possible combinations, as well as to understand the underlying reasons for their promising properties, are generally face-centred cubic systems.

Also in order to address the problem of **compatibility with liquid metals and other coolants** in connection with the prototypes, while models for austenitic steels are the main goal, including the problem of optimizing alumina-forming austenitic alloys, experience with “simpler” ferritic alloys is expected to help.

Concerning **ceramic materials**, these are often chemically and crystallographically complex systems (e.g. MAX phases), that require specific developments in terms of atomic-level modelling. Of all, SiC and Al<sub>2</sub>O<sub>3</sub>, are probably the only two systems on which some solid modelling background exists. However, the activities are globally very limited and scattered. More importantly, these materials are hardly ever used in their monolithic form and never as structural materials: in order to improve their mechanical properties, they are used in the form of composites. This opens completely different types of problems for modelling, because the macroscopic behaviour of these materials will be determined much more by the type of architecture of the composite materials and its degradation than by the degradation of the bulk material. Therefore, the monitoring of the behaviour of the architecture becomes of overarching importance, posing problems such as the need to obtain the correct 3D structure of the material and to model it correctly at the level of continuum mechanics, knowing that in fact the material is not a continuum. Clearly, the role of the interfaces between e.g. fibre and matrix becomes key to correctly simulate the macroscopic behaviour of the material.

### TA.1.1.3 Development of advanced structural materials

The development and codification of new materials solutions for nuclear application is a very long process, due to both the overall strict requirements to comply with the safety constraints of the nuclear industry and the requests of the regulators, and the unavoidable need to perform long and costly irradiation campaigns. This prevents, for example, the use of optimised materials for the short term ESNII prototypes. Yet, for further demonstrator phases, FOAK prototypes and longer term applications in commercial reactors it is important to pursue the development and codification of better performing materials and materials solutions.

The main targets are to **improve high temperature behaviour, minimise radiation effects and mitigate environmental degradation**. In this section, R&D needs for six material classes are discussed:

- Improved austenitic steels
- Ferritic/martensitic steels
- SiC<sub>f</sub>/SiC composites
- Refractory metallic alloys
- Modified surface layers for protection from corrosion
- Other perspective materials

For all these materials there are general issues of **fabrication processes to be developed/optimized and/or made less expensive**. For some of them **additive manufacturing** may be a way forward that is worth pursuing. There are also general issues of **protection against coolant attack** for those solutions that do not explicitly address this problem. Finally, for all of them different solutions need to be considered and **screening procedures** applied, which are especially costly when irradiation is involved: for some, but not for all, ion irradiation can be a way to go.



## Improved austenitic steels

Austenitic steels have been used as cladding and core component structural materials in past fast reactors. Their behaviour is therefore relatively well-known as a large database of irradiated materials is available. Overall, they offer excellent mechanical properties in a wide range of temperatures, including satisfactory creep resistance up to 700°C. The **main shortcoming for the use of austenitic steels as core components is their susceptibility to irradiation void swelling**<sup>78</sup> probably both an inherent property connected with the fcc crystal structure and also a consequence of the He production by transmutation through nuclide chains that start with Ni. Swelling leads to unacceptable embrittlement above 3% ( $\Delta V/V$ , where V is the volume).

Another issue affecting austenitic steels is that, despite their generally higher resistance to corrosion than e.g. F/M steels, they **may not offer sufficient guarantees of corrosion-resistance in HLM-cooled systems**<sup>79</sup>, especially with a view to pushing up the operating temperature for higher efficiency.

### *Swelling-resistant austenitic steels*

The 15-15 Ti-class of austenitic steels, which is presently the reference material class for the fuel cladding tubes and core internals for all the ESNII systems, at least in their first configuration, has been developed, starting from the AISI 316 formula, to improve the performance against swelling, through Ti addition with optimized C, the increase of Ni content and the fine tuning of minor alloy elements, above all P and Si. The beneficial effect of Ti, which largely remains in solid solution, has been associated primarily with the formation of complexes that trap vacancies, thereby enhancing their recombination with interstitials and decreasing point-defect supersaturation.<sup>80</sup> In addition the precipitation of a fine distribution of carbides may stabilize the dislocation microstructure, e.g. obtained by cold-working: a stable high dislocation density acts as efficient sink for point defects, thereby also delaying swelling. The 15-15 Ti microstructure thus allows 90 dpa to be reached. Further improvements are probably possible, especially with the help of suitably physical models that may help to identify and quantify in detail the mechanisms responsible for swelling.

A relatively recent specification of 15-15Ti with better swelling resistance is AIM1, which should push the maximum allowable dose to 115 dpa and is being qualified as cladding material for ASTRID (target burnup: 110 dpa). For the future, in addition to ODS cladding, or while this is being developed, the effect of swelling inhibitors in solid solution should be explored, towards a further optimized composition (AIM2)<sup>81</sup>. These are elements such as Ni, Si and P (partially optimised in AIM1), which are expected to decrease the kinetics of void formation, but also Nb and V that, by promoting carbide precipitation, should further stabilize the dislocation network. A downside is that an excess of carbides might lead to embrittlement, which needs to be avoided. The large database of properties after ion, electron and neutron irradiation makes it possible to establish correlations and ion irradiation is a very useful tool to be used for the screening between several nuances of composition.

### *Alumina forming austenitic steels*

Recent materials research has focused on the development of alumina forming austenitic (AFA) steels, with the aim of increasing the corrosion/oxidation resistance and the creep strength for high temperature applications.<sup>82</sup> AFA steels may contain Al in the range 4-6 %wt and a maximum of 25 wt% Ni, exhibiting superior oxidation resistance up to 900°C due to the formation of a protective Al<sub>2</sub>O<sub>3</sub> scale rather than the Cr<sub>2</sub>O<sub>3</sub>-rich scales that form on conventional stainless steels. They offer creep strength comparable with some superalloys that contain a much higher amount of nickel, thanks to strengthening via precipitation of NiAl particles. Some alumina forming alloys have been tested in molten lead alloys environment showing good corrosion resistance. For this reason, they

<sup>78</sup> C. Cawthorne, E.J. Fulton Nature, 216 (1967), pp. 575-576.

<sup>79</sup> K. Lambrinou, et al Journal of Nuclear Materials, Volume 490, 2017, Pages 9-27.

<sup>80</sup> Rouxel et al., EPJ Nuclear Sci. Technol. 2 (2016) 30; Terasawa et al., J. Nucl. Sci. & Technol. 19 (1982) 646.

<sup>81</sup> J.-L. Séran, M. Le Flem, Structural Materials for Generation IV Nuclear Reactors, Woodhead Publishing, 2017, Pages 285-328.

<sup>82</sup> Yamamoto et al., Intermetallics 16 (2008) 453.

are being considered as one of the most promising materials for future HLM-cooled plants, in particular for the steam generator.

AFA steels cladding tubes can be accordingly considered as a promising option for long term deployment in the LFR, provided that neutron-resistant materials are produced<sup>83</sup>. The materials under development are very similar to the  $\gamma'$  precipitate-hardened<sup>84</sup> Ni-base superalloys studied in the context of the US liquid metal fast breeder reactor programme, for which neutron irradiation data are available.<sup>85</sup> The lesson learned during the above research is that the use of thermodynamic simulations and ion irradiations could be used for the accelerated development of radiation resistant AFA. Nevertheless, their application as fuel cladding materials will require several compositional changes and qualification under neutron irradiation, before converging to a suitable composition. Moreover, the threat of LME remains open.

**LME** corresponds to a **loss of elongation that appears when a specific metal is tested in contact with a specific liquid metal**, with effects also on fracture toughness, fatigue behaviour, and possibly even creep-resistance, certainly in F/M steels. A large amount of experimental data from mechanical testing in LBE and, to a smaller extent, in pure lead environment, has been obtained during the last decades and, thanks to the extensive work done, the features of the phenomenon are at least qualitatively delineated<sup>86</sup>. This work has highlighted the need for direct and intimate contact between liquid and solid metal and the conditions under which the susceptibility to LME is especially high<sup>87</sup>. It has also highlighted, however, how difficult it is to rule out that these conditions can be reproduced in operation and the fact that oxide protection is not guaranteed to be a solution, especially in connection with fracture toughness. Austenitic steels seem to be much less affected by this issue, at least below 400°C. Yet it has been demonstrated, in contact with liquid Na, that these steels also see their mechanical behaviour affected if intimate contact is achieved. Moreover, the use of some type of filler material to reduce the susceptibility to hot cracking may lead to  $\delta$ -ferrite contents up to 5% in the welds. LME could therefore have a serious impact on the mechanical properties of welded joints of austenitic steels exposed to HLM. The role of unstable austenite having a martensitic transformation under mechanical strain is also a possibility that would render austenitic steels more susceptible to LME.

A **clear understanding** of the mechanisms behind LME in F/M steels and the synergy between LME and irradiation hardening is **still missing**, as well as a model to rationalize the whole body of available experimental data. The lack of understanding based on experimental observations calls for the use of advanced modelling tools in order to explore hypotheses and mechanisms from a physical standpoint. It is widely recognized that the complexity of the phenomenon, and the challenges in controlling the experimental details, make it difficult to discriminate the relevant processes to LME from those that are not. **The identification of a proper framework for materials codification and licensing whenever LME is involved is expected to be especially complex.**

### Ferritic/martensitic steels

Despite their high thermal conductivity and excellent dimensional stability under neutron irradiation, the core applications of F/M steels in fast reactors have been limited to the hexagonal cans in the French SFR's. Their **use as cladding or core material in other systems is at present prevented by:**

- Loss of strength at high temperature ( $T > 550^\circ\text{C}$ ), including softening under cyclic operation;<sup>88</sup>
- Neutron irradiation embrittlement at low temperature ( $T < 350^\circ\text{C}$ ), including plastic flow localisation with subsequent drastic reduction of uniform elongation;<sup>44</sup>
- Susceptibility to liquid metal embrittlement in contact with HLM.<sup>89</sup>

<sup>83</sup> P. Lorusso et al, Progress in Nuclear Energy, Volume 105, 2018, Pages 318-331.

<sup>84</sup> Goodfellow et al., Metall. & Mater. Trans. A, 49 (2018) 718; Bowman, Superalloys: A Primer and History, <https://www.tms.org/Meetings/Specialty/Superalloys2000/SuperalloysHistory.html>;

<sup>85</sup> Boothby, Radiation Effects in Nickel-Based Alloys, in: Comprehensive Nuclear Materials 4.04 (2012) 123.

<sup>86</sup> D. Gorse, et al J. Nucl. Mater., 415 (2011), pp. 284-292.

<sup>87</sup> X. Gong et al Journal of Nuclear Materials Volume 509, October 2018, Pages 401-407.

<sup>88</sup> Raj & Vijayalakshmi, Ferritic Steels and Advanced Ferritic-Martensitic Steels, in: Comprehensive Nuclear Materials 4.03 (2012) 97.

<sup>89</sup> Ersoy et al., J. Nucl. Mater. 472 (2016) 171.



These phenomena and the different material laws of F/M steels with respect to austenitic steels, complicate the codification of the former, which cannot be a simple adaptation of the design rules used for the latter. For this reason, even existing F/M steels such as T91 (or Eurofer for fusion) have not been fully codified yet in RCC-MRx.<sup>90</sup>

Nevertheless, in the long term the use of F/M steels is very desirable in a context of optimal use of resources (high-burn up), given that, at present, these are the only available industrial materials that **can bear the promise of withstanding neutron doses in excess of 150 or even 200 dpa.**

Their use in future GenIV commercial power reactors will require a number of actions:

- Improvement of **high temperature mechanical behaviour**;
- Improvements of **ductility after low temperature irradiation**, including **slip localization with loss of uniform elongation**;
- Improvement of the **compatibility with HLM**, by investigation the mechanisms responsible for corrosion and, even more importantly, **liquid metal embrittlement.**

Any **strategy proposed to mitigate these problems** requires not only an effort in terms of improved material development, based on appropriate understanding of the relevant mechanisms and models of the underlying ageing processes, but also of qualification and pre-normative research, in view of the relevant codification.

The two proposed alternatives included here to improve high temperature strength of F/M steels, ODS and CSE, should also include the assessment of the potential cyclic softening, low temperature irradiation (slip localization) and compatibility with coolants. Both classes of steels have potential application also in other energy technologies where high temperature and temperature gradient effects are an issue, e.g. concentrated solar power.

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### ***Oxide dispersion strengthened steels***

Increasing the operating temperature as well as the mechanical strength of F/M steels by a fine dispersion of oxide particles in the matrix is considered a viable path towards improved properties for cladding applications. Indeed, ODS F/M steels offer indeed improved strength at temperatures above 550°C, potentially up to 700°C or even above, while keeping the outstanding swelling resistance of the ferritic (or ferritic/martensitic) matrix. ODS cladding tubes have been produced and their qualification is included already in the early phases of the ASTRID SFR<sup>91</sup>.

The homogeneous dispersion of oxide particles in the nanometre range is obtained thanks to a powder metallurgy fabrication procedure, where the atomized steel particles of the desired chemical composition are milled with oxide particles (usually yttrium oxide) by mechanical alloying. Post-processing steps involve degassing, consolidation (by HIP or extrusion) and annealing treatments. The **nano-sized oxide particles in high number density not only improve creep strength, but also act as sinks for irradiation induced defects and thus improve radiation resistance**<sup>92</sup>. This effect is more or less strong depending on the chemical composition of the ODS and the nature of the nano-oxides, which therefore play an important role.

The main issue affecting ODS cladding is certainly the **high cost** associated with the multiple fabrication steps needed in a classical powder metallurgy route, which is also a **bottleneck for upscaling to industrial production**. In addition the microstructural details associated with the individual steps of the processing route are not yet well understood: this currently **limits the reproducibility of the quality of the end product**. In particular, the resulting microstructure after tube production is highly anisotropic. Removing this **anisotropy** is not trivial and require the still open development of suitable intermediate thermal treatments leading to controlled

<sup>90</sup> Eurofer is present in RCC-MRx in the probatory section.

<sup>91</sup> P. Dubuisson, Journal of Nuclear Materials, Volume 428, Issues 1–3, September 2012, Pages 6-12.

<sup>92</sup> M. Klimenkov, et al, Journal of Nuclear Materials, Volume 493, September 2017, Pages 426-435.

recrystallization<sup>93</sup>. So there is a **need to optimise and standardize the fabrication process, guaranteeing reproducibility and industrial scalability**. Possible **alternative and cheaper fabrication routes** should also be identified. **Additive manufacturing** presents itself as a promising way forward. Finally, an open issue is the development of **appropriate welding procedures**<sup>94</sup>. Conventional fusion welding destroys the distribution of the fine oxide particulates which give the steel its strength and creep resistance. Presently no established welding methodology has been devised, yet, and further research is needed in this direction.

**The TRL of ODS steels remains low:** the priority here is therefore to improve the production routes, to guarantee reproducibility and industrial production, in partnership with steel-makers (although currently hardly any is equipped for ODS production, especially in Europe), together with analysis of deformation and fracture mechanisms and stability of the microstructure under irradiation and at high temperature (again, physical models are expected to be of use for these purposes).

### ***TMT/composition-tuned creep-strength enhanced F/M steels***

A promising alternative, or at least complement, to the ODS obtained by mechanical alloying are conventionally produced (i.e. by casting in crucible) F/M steels, the **creep performance of which is enhanced by particle (carbide) strengthening, via conventional metallurgical techniques**, based on fine tuning of the chemistry and suitable post-casting thermomechanical treatments<sup>95</sup>. These are referred to as CSE F/M steels. It has been shown that compositional adjustments on conventional 9-12%Cr steels and thermo-mechanical treatment optimization, guided by computational thermodynamics, produces a better distribution of the reinforcing carbides and a more stable microstructure at high temperature. Notably, **F/M steels operating up to 650°C have been produced for use outside nuclear energy**. While it is unlikely that this operating temperature can be further exceeded by pursuing this development path, no nuclear grade steel of this type currently exists, thus the **production of nuclear grade CSE F/M steels** would be certainly a valuable step forward.

The current use of these classes of steels for cladding (but a priori also for other core components) requires the elaboration of the corresponding design criteria and assessment procedures, which, in turn, requires dedicated testing for data collection and physical modelling including the stability of the optimised microstructure under irradiation. The codification of conventional CSE F/M steels is expected to be more simple than for ODS, which exhibit significantly different deformation and fracture behaviour, by being linked to the more general issue of the elaboration of design rules for existing F/M steels. It is also considered that **compositional tuning and thermomechanical treatments** may also enable the **minimisation of the problem of low temperature radiation embrittlement**, by reducing both the DBTT before irradiation and the DBTT shift after irradiation<sup>96</sup>. The former seems to be achievable by working on grain size reduction through both thermomechanical treatments and composition changes; the latter mainly through compositional changes, removing or minimising the elements that are responsible for embrittlement.

### ***Improvement of compatibility with coolants***

F/M steels are prone to **corrosion by dissolution and erosion** when in contact with flowing liquid metals. While this problem is shared with other steels and classes of materials, this specific class of steels, as well as many other alloys with body centred structure, is reported to suffer from **liquid metal embrittlement** when exposed to heavy liquid metals.

The problem of **corrosion by dissolution and erosion** can be faced either by applying suitable **surface protections** (coatings), or by aluminization **surface treatments**, which would promote the spontaneous formation of a stable aluminium oxide ( $Al_2O_3$ ) layer, or by producing corrosion resistant steels that by themselves, through **addition of Al**, produce such a protective layer. Steels of this type, denoted as **FeCrAl**<sup>97</sup>, which are the equivalent of AFA for F/M steels, exist already and are also industrially produced: they may contain up to 20-30 wt% Cr and between 4 and 7-10 wt% Al. However, they still need composition optimisation in order to minimise side-effects of the addition of

<sup>93</sup> E. Vakhitova et al Journal of Nuclear Materials, Volume 494, October 2017, Pages 20-28.

<sup>94</sup> Suk Hoon Kang et al, Fusion Engineering and Design, Volumes 109–111, Part A, 1 November 2016, Pages 182-185.

<sup>95</sup> R.L. Klueh, et al, Scripta Mater., 53 (2005), p. 275.

<sup>96</sup> D. Strok et al, Fusion Engineering and Design, Volume 89, Issues 7–8, October 2014, Pages 1586-1594.

<sup>97</sup> Jun Lim et al Journal of Nuclear Materials, Volume 441, Issues 1–3, October 2013, Pages 650-660.

Al (decrease of ductility despite a fully ferritic microstructure) while guaranteeing the formation of a stable and continuum oxide layer, which requires reducing the content of both Cr (towards the content typical for F/M steels in nuclear applications, i.e. 9-12 wt%) and especially Al. This objective can be reached by adding small quantities of suitable reactive elements, for example Hf or Zr.<sup>98</sup>

F/M steels are especially prone to embrittlement when exposed to LM (see above). It is possible - although it remains to be demonstrated - that these alumina-protected steels may also provide protection against LME. A clear understanding of the mechanisms behind LME in F/M steels is still missing, and with the synergy between LME and irradiation hardening this prevents clear conclusions to be drawn in this respect.

### SiC<sub>f</sub>/SiC composites

Increasing the temperature of operation beyond 800°C, such as in the case of GFR and VHTR, requires the use of refractory materials that allow access to temperatures well beyond the current limits of the most heat-resistant super-alloys. Of all the possible material classes considered in the past, the main and conceptually most advanced candidates are those based on SiC<sub>f</sub>/SiC composites, i.e. SiC fibers in a SiC matrix. Sublimating into vapour directly from the solid at 2830°C, this is indeed one of the most temperature resistant materials available on earth. These composites are also proposed in the commercial deployment of the SFR for the realization of the hexagonal cans, as well as for cladding and structural materials for high temperature HLM cooled systems, due to their good corrosion and erosion resistance,<sup>99</sup> while being considered also for advanced accident tolerant fuel cladding for GenIII+ reactors.<sup>100</sup>

Thin-walled SiC<sub>f</sub>/SiC tubes are fabricated using textile methods where fine filaments (~ 10 μm) combined into fibre tows, are woven or braided into a tubular cladding preform and bound into place by an additional bulk SiC matrix, by chemical vapour deposition (CVD), designed to prevent the weave from moving under stress. An interlayer of pyrolytic carbon (PyC) is deposited on the fibers by chemical vapour infiltration (CVI). The PyC interlayer transfers the mechanical loads within and through the ceramic fiber reinforced composite, allowing the material to strain without catastrophic brittle failure. At the macroscopic level composites of this type emulate a plastic behaviour, but at the microscopic level they accommodate the deformation by micro-cracking damage. The **main weakness** of these composites as cladding materials is that, both as produced and especially after deformation, they are **not hermetic** and therefore **cannot retain the fission products**.

A potential solution to this issue is the “sandwich” concept recently proposed and patented by the French CEA, where a metallic liner is interposed between two structural SiC<sub>f</sub>/SiC shells to secure hermeticity against fission gas release. SiC<sub>f</sub>/SiC composite thin-walled, small diameter tubes with one closed end and internally lined with a refractory metal are thus the pin type fuel elements envisaged by the present design for the high temperature configuration of the GFR. An alternative design, the triple-layered (triplex) structure, features a fully ceramic component made of high-density monolithic SiC tube to ensure the gas tightness and an outer layer made of SiC<sub>f</sub>/SiC to prevent brittle fracture.

Various processes produce the same local chemical bond between the silicon and carbon atoms, but different porosity, crystallography (beta or alpha phases) and Si/C stoichiometric ratios. These influence strongly the overall mechanical and thermal behaviour of SiC<sub>f</sub>/SiC, as well as its performance under irradiation.

### SiC behaviour under irradiation

<sup>98</sup> R.Gao et al Journal of Nuclear Materials Volume 444, Issues 1–3, January 2014, Pages 462-468.

<sup>99</sup> A. K. Rivai, M. Takahashi, J. of Pow. and Ener. Sys. 1 (2007) 134-142; M. Takahashi, M. Kondo, Prog. in Nucl. Ener., 53 (7) (2011) 1061-1065; M. Kondo, M. Takahashi, J. Nucl. Sci. and Tech 43(2) (2006) 174-178; A.K. Rivai, M. Takahashi, Prog. Nucl. Energy 50 (2008) 560-566; M. Takahashi, S. Uchida, Y. Kasahara, Prog. Nucl. Energy 50 (2008) 197- 205.

<sup>100</sup>

<http://www.westinghousenuclear.com/Portals/0/Technovation%20Stuff/Accident%20Tolerant%20Fuel%20Brochure%20.pdf>; <http://www.ga.com/accident-tolerant-fuel>.

Nuclear-grade SiC<sub>f</sub>/SiC composites have shown to be stable to extremely high irradiation doses after the initial swelling and changes in strength and thermal conductivity have saturated at relatively low irradiation dose ( $0.1 \cdot 10^{25} \text{ n} \cdot \text{m}^{-2}$ ,  $E > 0.1 \text{ MeV}$ ).

The **swelling** behaviour of high purity beta SiC with stoichiometric composition ( $\text{Si}/\text{C} = 1$ ) is relatively well-known in the temperature range of interest ( $400^\circ\text{C}$ - $1600^\circ\text{C}$ ): it decreases with increasing temperature to a minimum at  $\sim 1000$ - $1200^\circ\text{C}$ , and increases above this with the onset of void formation, but **remains less than 1-2% in the whole range**.<sup>101</sup> Deviations with excess Si or C may induce local volume changes, and non-stoichiometric SiC-based fibers are known to undergo substantial volume contraction under exposure to neutrons. Here the **main issues to be addressed concern swelling in the actual component** (fuel cladding tube): for this reason SiC<sub>f</sub>/SiC sandwich composites irradiations have been performed in the BOR60 reactor in 2012-2015, in contact with sodium at  $550^\circ\text{C}$ , with doses up 105-120 dpa.

The **loss of thermal conductivity** of SiC<sub>f</sub>/SiC under irradiation by one order of magnitude or more poses the **most severe limitation** for its application as fuel cladding. A major role in this is played by the evolution during irradiation of the PyC interphase, through the formation of a network of microcracks, due to amorphization and dimensional changes, which induces radial tensile stress at the interface between the PyC and SiC. If the stress exceeds the bond strength between layers, debonding of PyC from SiC occurs.<sup>102</sup> Work should be therefore devoted to understand the mechanisms that drive the microstructural evolution of the PyC with increasing dpa. Ion irradiations can provide only limited information because, contrarily to metals, covalent compounds are severely damaged by electronic excitations, making it especially difficult to extrapolate their behavior under neutron irradiation.

### SiC corrosion Issues

Chemical compatibility in reactor environments is highly dependent not only on the thermodynamic stability of SiC in the pure coolants, but also in contact with the possible impurities contained therein. The coolant may contain small amounts of gas impurities such as  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{H}_2\text{O}$ ,  $\text{H}_2$ ,  $\text{CH}_4$ ,  $\text{O}_2$ , as well as solid particles coming from a variety of sources throughout the reactor system.<sup>103</sup> However, the lack of graphite moderation in the GFR, compared to HTR systems, will significantly reduce the concentration of C-based impurities. Therefore the corrosion of SiC<sub>f</sub>/SiC cladding in a GFR will likely occur primarily by oxidation, rather than by any other chemical process. The key oxidising impurities to consider in the He coolant will be oxygen and water vapour, the latter being of greatest importance. The **reaction of SiC with O<sub>2</sub> and moisture** at elevated temperatures leads to three typical oxidation features, passive oxidation, active oxidation and bubble formation. In an environment with sufficient high oxygen concentration, pure SiC is expected to form a  $\text{SiO}_2$  passive layer protecting the surface according to the reaction  $\text{SiC} + \text{O}_2 \rightarrow \text{SiO}_2 + \text{C}$ . But under oxygen deficient conditions the reaction proceeds as  $\text{SiC} + \text{O} \rightarrow \text{SiO} + \text{CO}$ , where the SiO is a gas and the reaction proceeds until the SiC is consumed.<sup>104</sup> Furthermore, in environments containing water vapour at very high temperature ( $1900\text{K}$ ) the formation of SiO and CO vapours at the SiC/SiO<sub>2</sub> interface leads to bubble formation, growth and rupture of the oxide. In the active oxidation and bubble formation regions, SiC<sub>f</sub>/SiC therefore undergoes significant loss of mass and non-protective  $\text{SiO}_2$  layer formation. Thus, the use of SiC composites requires extensive testing to evaluate the impact of non-passivating oxidation and corrosion in relevant environments. Barrier coatings to hinder or limit corrosion may have to be envisaged.

### SiC Joining

SiC<sub>f</sub>/SiC tubes are initially fabricated with one closed end; the open end must then be hermetically sealed after loading the fuel pellets with an end-cap that should withstand the pressure of the fission gasses and the neutron radiation field, while being chemically stable in the coolant environment. A crucial technology gap is the **lack of a reliable, reproducible technique to join and hermetically seal**

<sup>101</sup> L.L. Snead et al. JNM 371 (2007) 329.

<sup>102</sup> Gregory K. Miller, David A. Petti, John T. Maki Journal of Nuclear Materials 334 (2004) 79–89.

<sup>103</sup> Estimation for ALLEGRO from HTR-10 experiments.

<sup>104</sup> This active oxidation could be an issue for the use of SiC in Na or HLM that contain little oxygen.

**the tubes.** A number of techniques for the joining of ceramic composite materials to themselves or dissimilar materials (e.g. metal components) have been developed. These include spark plasma sintering, laser-based joining, brazing, diffusion bonding, transient eutectic phase routes, glass-ceramic joining, adhesion, pre-ceramic polymer routes and mechanical fastening. Presently there is insufficient information pertaining to the compatibility with coolants and stability under irradiation of the joints and additional studies are needed for the assessment of reliable joining technologies

### ***Standardization of testing procedures for SiC<sub>f</sub>/SiC***

The qualification and eventual codification of SiC<sub>f</sub>/SiC requires a vast effort of pre-normative research, starting from the problem of the standardization of tests as a pre-requisite to characterize the behaviour of the material in environments (tightness against fission products, contact with flowing He, irradiation). **Standards for mechanical testing of nuclear grade SiC<sub>f</sub>/SiC are thus necessary** to produce accurate and reliable data, based on well-defined test methods, detailed specimen preparation, comprehensive reporting requirements, and commonly accepted terminology. In particular, standard test methods are critically important to achieve a predictive capability for the failure probability of SiC<sub>f</sub>/SiC tubular structures under given loading conditions. Testing is performed at several laboratories to progress the development of the GFR fuel element, so it is mandatory that common and well-recognized sets of testing and evaluation technologies are followed. Lacking these, the work would become inconsistent and difficult to accept by the design community.

A number of standard tests for continuous fiber ceramic composites have already been developed by relevant technical committees of existing National/International Organizations for Standardization: ASTM (C28-07), CEN (TC184-SC1), ISO (TC206/WG4), AFNOR (B43-C). However, not only there are no commonly accepted design methodologies for tubular components made of advanced composites, but there are also no mechanical test standards for any of the properties of tubular geometry ceramic composite components. Also, the temperature range of interest for the clad case is not addressed.

Work on standards for ceramic composites is deliberately separated from that on monolithic ceramics, due their heterogeneous microstructure, unique properties and behaviour, requiring appropriate specimen geometry, experimental procedures and approaches to data analysis. As a consequence, current standards for design purposes are inadequate because they ignore the fundamental issue of whether the specimen they employ is fully representative of the entire composite structure. The failure behaviour of SiC<sub>f</sub>/SiC components having tubular geometries is anticipated to be significantly different from that observed on flat two-dimensional architectures. Developments in this direction are therefore required, with the contribution of qualified laboratories having experience in continuous fiber ceramic composite testing and investigation, and possessing the necessary equipment.

### ***Other ceramics***

The design of the high temperature gas cooled reactor concept (GFR, V/HTR), imposes a need for materials solutions to be devised for the purposes of **thermal shielding and insulation**, as well as **control rods and seals**. These materials must be gas-tight, corrosion resistant, and exhibit high fracture and creep strength, while being inexpensive and preferably fabricated in a net-shape design, with the ability to be joined. Achieving reasonable leak tightness is one of the most important issues, since He gas permeates easily through most materials. While also some metallic materials are being explored, (Inconel/Ag) and ceramics are especially investigated and tested for O-ring seals. In many cases the material chosen also should be resistant to shocks related to thermal transients, which might occur when the flow of the process fluid or the coolant is interrupted. Therefore, next to the superalloys, ceramics with improved fracture toughness are being developed, i.e. for intermediate heat exchangers. At the moment, in addition to SiC (deposited using CVI), also other materials such as carbon composites, mullite, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, ZrC, ZrN, B<sub>4</sub>C, graphite or graphene are being considered for this function.



## Refractory alloys

The requirements of safe operation at very high temperature calls for materials with melting temperature above 1500°C. **Refractory metals (V, Nb, Mo, Ta and W)** have melting points above 2000°C and are thus an obvious choice for high temperature structural applications, as a **metallic alternative to SiC<sub>f</sub>/SiC** and also as competitors to advanced steels for in-core applications. They are potential candidate materials for the commercial deployment of SFR and HLM cooled systems and serve as a backup solution for GFR fuel cladding to provide a risk mitigating measure in case of failure of the SiC<sub>f</sub>/SiC option. The high melting temperature implies several beneficial properties such as high elastic modulus, high microstructural stability, high limiting creep rate and low thermal expansion coefficient. However, these alloys exhibit **high affinity for oxygen, hydrogen, nitrogen and carbon**, which easily diffuse in the bulk causing **hardening and embrittlement** during both the production process and the service life.

The upper temperature limit of applicability of these materials is thus mostly determined by oxygen corrosion and interstitial impurity embrittlement. This restricts their high temperature applicability to vacuum or high purity inert atmospheres, unless they are properly protected by suitable coatings to prevent oxidation and mass transfer issues. Canning under inert atmosphere is mandatory for high temperature processing and welding need appropriate processing to avoid contamination.

The low temperature limits for their use under neutron irradiation are determined by irradiation embrittlement and DBTT shift at or above the operational temperatures. V, Nb, Mo, Ta and W all have a DBTT at or above room temperature and radiation induced defects shift the DBTT to around 400°C in V4Cr4Ti and above 700°C for Mo ZTM.<sup>105</sup>

Another key challenge at present is to re-establish large-scale production capabilities and to recapture former expertise on processing, joining, brazing, cold work in various shapes, irradiation effects, and weldability.<sup>106</sup>

Several refractory metals and alloys have been considered in fusion for first wall and blanket structural materials applications. Here only two alloys of this type are further discussed as prominent examples, both were studied to some extent in the past and are recently experiencing renewed interest for cladding: V and Mo alloys.

### **Vanadium alloys**

Advanced V-alloys with an increased operating temperature window are considered in the US and in Japan for fusion reactor blankets.<sup>107</sup> High quality manufacture of V-4Cr-4Ti heats of high purity, with state-of-the art properties required for blanket application, has been demonstrated in a variety of engineering-relevant shapes, including small diameter thin wall tubes for creep studies. These materials have attractive features also for application as fuel cladding materials in fast reactors. Research on V-alloys as clad materials for the previous generation SFR was initiated in Europe during the 1970s but was soon discontinued. A programme of fabrication of V-4Cr-4Ti alloy (CEA-J57 grade) cladding tubes for SFR was started in 2008 by CEA and INR,<sup>108</sup> but was interrupted in 2015 due to budget constraints. V-alloys remain on the list of potential materials for GenIV applications.

The V-4Cr-4Ti alloy has a design temperature window of about 430°C-700°C, for an allowable stress of 100 MPa, which is bounded on the lower side by irradiation embrittlement and on the upper side by thermal creep and recrystallization. An essential condition to make V-alloys potentially suitable for core applications is to enlarge this reference window, by expanding both the low and high operation temperature limits. The issue to be addressed is therefore to find the most appropriate approach (composition, fabrication process, ...) to improve high temperature strength and low temperature ductility of V-alloys, similarly to F/M steels.

Another element of concern is the high vulnerability to oxidizing species and susceptibility to embrittlement by the impurities that unavoidable in fast reactor coolants. Compatibility issues with fuel or fission products could also arise. These are very significant feasibility issues imposed on the

<sup>105</sup> Zinkle and Ghoniem, Fusion Engineering and Design, 51-52 (2000) 55-71

<sup>106</sup> Zinkle et al., Proc. Space Technology & Applications International Forum, STAIF-2002, AIP Conf. Proc. No. 608, vol. 1, American Institute of Physics, Melville, M.S. El-Genk (Ed.), NY, 2002, p.1063.

<sup>107</sup> Muroga, Vanadium for Nuclear Systems, in: Comprehensive Nuclear Materials, 4.12 (2012) 391.

<sup>108</sup> FP/ MATTER Report D11.2.

development of adequate protective barriers to ensure protection against oxidation and corrosion. Attention should be focused on suitable fabrication and processing technologies that: avoid contamination; are compliant with operative requirements and nuclear environment; and are applicable to complex surfaces, including tubes and their internals.

### **Molybdenum alloys**

With a melting temperature in excess of 2600°C, molybdenum is the most versatile among the refractory metals and an excellent structural material for applications requiring high strength and rigidity at temperatures up to 1650°C, (60–80 MPa at 1500°C), where it can operate in vacuum, under inert or reducing atmospheres or with a coating. Mo alloys also have excellent thermal conductivity (138 W/m K) and an acceptable neutron capture cross section.<sup>109</sup> They have been extensively investigated for their application as cladding of fast spectrum high temperature gas cooled reactors for space.<sup>110</sup> In fusion, TZM (Titanium Zirconium Molybdenum) and Mo-Re alloys have been studied for application to the first wall and blanket structures. More recently, after the Fukushima accident, an ATF cladding design is under development that utilizes molybdenum's high strength at temperatures up to ~1500°C to maintain fuel rod integrity and core coolability during a severe accident. Sufficient experience on fabricability and joinability exists for molybdenum alloys such that component construction is not a major concern.

Like the other refractory metals, molybdenum and its alloys have a great affinity for oxygen, carbon and nitrogen, which easily diffuse in the lattice occupying the interstitial sites and cause embrittlement. Its application needs therefore a clean environment, with very low oxygen activity. Thus, rather than the loss of mechanical properties, the temperature limit of applicability is mostly determined by oxygen corrosion and interstitial impurities embrittlement. In addition, Mo and Mo based alloys are susceptible to oxidation corrosion in oxidizing environments where the low melting temperature (795°C) and the formation of volatile oxides (sublimation is not negligible even at temperature as low as 400°C) give rise to catastrophic oxidation.<sup>111</sup>

Despite suffering oxidation corrosion in oxidizing environments, several authors report an outstanding resistance to HLM corrosion after testing in both stagnant and fluent conditions, at several temperatures and up to 800°C, in the presence of temperature gradients and with different oxygen concentrations. The features responsible for this outstanding corrosion resistance in HLM have been little investigated and the negligible solubility of Mo in lead surely plays a major role. Some authors report the formation of a lead molybdate  $\text{MoO}_4\text{Pb}$  film that may render the surface passive.

Molybdenum and its alloys have good swelling behaviour under neutron irradiation, with volumetric swelling less than 3% ( $D\phi/\phi < 1\%$ ) for irradiation up to 110 dpa. Due to its neutron capture cross section, Mo and its alloys have been excluded from the list of candidate materials for the high temperature evolution of the SFR. However, the lower neutron absorption cross section of HLM and He cooled systems makes their use viable in LFR's and GFR's cores. The main drawback for their use in core applications is their limited ductility and irradiation induced embrittlement with, depending on the dose, a DBTT shift of up to 800°C for unalloyed Mo. Impurities influence the mechanical properties, particularly through grain boundary weakening, but strong improvements in ductility have been achieved through grain refinement, impurity control, and alloying<sup>112</sup> to modify the grain boundary precipitates and solute distribution. Creep performance is, however, a potential issue to be evaluated for applications as cladding material, so oxide dispersion strengthening is a possible way to be pursued to improve this point.

Rare-earth oxide particles in ODS Mo-La alloys have been found particularly effective in reducing the grain size and gathering the oxygen in solution. These materials show a DBTT at -100°C in the unirradiated state, largely due to the grain size reduction and the presence of the oxide particles that

<sup>109</sup> Shmelev and Kozhahmet, *Journal of Physics: Conf. Series* 781 (2017) 012022.

<sup>110</sup> Cockeram et al. *J. Nucl. Mater.* 382 (2008) 1; Byun et al., *J. Nucl. Mater.* 376 (2008) 240.

<sup>111</sup> Simnad and Spilners *JOM* 7 (1955) 1011. <https://doi.org/10.1007/BF03377603>; Lai, *High-Temperature Corrosion and Materials Applications*, ASM Intl. Materials Park Ohio; Smolik et al., *Oxidation and Volatilization of TZM Alloy in Air*, 9<sup>th</sup> International Conference on Fusion Reactor Materials ICFRM-9 10 - 15 Oct 1999, Colorado Springs, CO, USA.

<sup>112</sup> Leonard, *Radiation Effects in Refractory Metals and Alloys*, in: *Comprehensive Nuclear Materials* 4.06 (2012) 181.



strengthen the grain boundaries.<sup>113</sup> They also show an outstanding reduction in the DBTT shift after irradiation even at low temperatures: the DBTT increases to 25°C for irradiation with fast neutrons to 13.1 dpa at 833–882K, and for irradiation to 13.1 dpa at 1143–1209 K remains unchanged from the non-irradiated material. The reduced susceptibility to irradiation embrittlement of ODS-Mo at low irradiation temperatures, at which defect mobility is limited, is considered to be due to the increased density of interfaces acting as sinks that is associated with the reduction of grain size and the introduction of oxide particles.<sup>114</sup>

The corrosion behaviour of Mo-based alloys in oxidising environments is an issue, due to the formation of MoO<sub>3</sub>, which volatilizes easily above 400°C, leading to rapid oxidation that is potentially catastrophic at higher temperatures. However, Mo has good corrosion resistance in several liquid metals including Bi, Li, K, Pb and Na. Corrosion data in lead and lead bismuth are few and sparse and nothing is known about the mechanical performance in contact with HLM. Mo alloys have high corrosion resistance and poor solubility in liquid Pb and LME up to 800°C.<sup>18,115</sup> So, depending on the mechanisms leading to the observed corrosion resistance, their application as cladding may require the deposition of suitable diffusion barrier to prevent oxidation corrosion and mass transfer issues for high temperature applications. Beyond the compatibility with liquid metal coolants, the neutronic performance of the material must be evaluated, considering the reactions that take place during the fast neutrons irradiations, the attenuation that follow and the compositional changes that may occur.

The arguments above call for a careful and systematic evaluation of these alloys to verify whether they meet the challenging requirements for application in the core of HLM cooled systems. The feasibility of Mo-alloy fuel cladding for HLM cooled reactors should be thus demonstrated via: (a) Characterization of the corrosion in HLM with varying [O] and temperature; (b) characterization of the creep performance of ODS-Mo alloys; (c) mechanical testing to investigate LME issues; (d) development of welding procedures; (e) development of barrier coatings to prevent gathering of the oxygen contained in the coolant; and (f) irradiation at dpa doses relevant for GenIV, to evaluate swelling and embrittlement issues.

### Modified surface layers for protection against coolant attack

The adoption of corrosion-resistant materials and the mitigation strategies set in place to limit the effects of aggressive media and wear may not be sufficient or applicable, thereby calling for protection via coatings or other surface treatments. The underlying idea is that **protective surface layers should not alter the mechanical properties of codified materials with known properties, but would impart the desired corrosion (and/or wear) resistance.** The previous sections show that this need is common to (a) steels in HLM operating at temperatures above 400-450°C; (b) SiC<sub>f</sub>/SiC in oxygen deficient high temperature environments; and (c) Refractory alloys, (d) parts and components subject to fretting wear (stellite replacement).

There exist five main criteria for the development of an optimized surface layer (SL):

- 1) The SL should form thermodynamically stable passivating phases by reaction with the environment.
- 2) These phases should be slowly growing in order to keep SL reservoir depletion rates low.
- 3) Interdiffusion between the layer and substrate should occur as slowly as possible, suggesting the introduction of an interdiffusion barrier or substrate where the diffusion rate of the SL species is high.
- 4) The values of the coefficients of thermal expansion of SL and substrate should be very similar, to minimize cooling and reheating stresses during temperature oscillations.

<sup>113</sup> Cockeram et al., J. Nucl. Mater. 346 (2005) 165.

<sup>114</sup> Cockeram, Metall. Mater. Trans. 40A (2009) 2843; Mueller et al., Inter. J. Ref. Met. Hard Mater. 18 (2000) 205; Liu et al., Nature Materials 12 (2013) 344.

<sup>115</sup> Rivai and Takahashi, Progress in Nuclear Energy 50 (2008) 560-566; Fazio et al., J. Nucl. Mater. 318 (2003) 325; Hata and Takahashi, Corrosion study in direct cycle Pb-Bi cooled fast reactor, Proc. GLOBAL, Paper No. 446, Tsukuba, Japan (2005).

- 5) If deposited, the SL deposition processes should be carried out at low temperature to avoid the degradation of the substrate performance, in general and especially under neutron irradiation.

The last constraint rules out the adoption of diffusion coating techniques for the core components that imply exposure to temperatures  $> 800^{\circ}\text{C}$  (for steels) for times of the order of hours.

Most work on protective SL for core applications in ESNII prototypes involves two systems:

- alumina-forming metallic layers produced by GESA;
- ceramic  $\text{Al}_2\text{O}_3$  barrier coatings produced by pulsed laser deposition (PLD).

These two are discussed here as prominent examples, together with detonation gun spraying as an especially straightforward solution, and atomic layer deposition as an emerging technique.

Other processes are of course open and possible: among others, coatings based on refractory alloys, alumina forming alloys, silica forming alloys and MAX phases have been considered and tested. FeCrAl coatings produced by pack cementation are being considered for their application to the steam generator of ALFRED.

### ***GESA process***

This process consists of the deposition of an Al-containing metallic layer on the steel surface, subsequently melting the layer, together with a few  $\mu\text{m}$  from the substrate, using intense pulsed electron beams.<sup>116</sup> During exposure to an oxygen-containing medium, if the concentration is sufficient, a thin, continuous, slowly growing, adherent and stable alumina layer is formed at the surface, which protects the components from corrosion attack.

No sign of dissolution attack or exfoliation was observed during corrosion tests performed at temperatures between  $400\text{--}650^{\circ}\text{C}$  for up to 10000 h. During creep-to-rupture, cycling fatigue, bending, tube pressurization and erosion tests, the alumina layer protected the steel components with modified SL against the negative influence exerted by the HLM on the mechanical properties. The modified SL has shown also promising irradiation tolerance.

However, for qualification of this material concept, R&D is needed on: (a) optimization of the SL composition; (b) optimization of the deposition method; (c) exposure to HLM in normal and off-normal conditions; (d) ageing in HLM; (e) welding of steel components with modified SL (e.g. end caps); and (f) irradiation tests at relevant doses in presence of HLM.

### ***Pulsed lased deposition (PLD)***

In this case, an  $\text{Al}_2\text{O}_3/\text{FeCrAlY}$  system that includes barrier coating and buffer layer is deposited using a pulsed laser. The buffer layer accommodates the stresses due to the differences in the thermomechanical behaviour. Coatings deposited on F/M steels have shown, after testing, no need for a buffer layer. PLD is a relatively simple technology that allows thin films to be grown on a wide range of materials. The deposition conditions can be adjusted to obtain different coating microstructures, varying from fully dense and compact to columnar and porous. Fully dense and compact  $\text{Al}_2\text{O}_3$  coatings, grown by PLD on various steels, have been obtained by tailoring the deposition process to attain an advanced nanocomposite which consists of a homogeneous dispersion of ultra-fine nanocrystalline domains, with size in the range  $6\pm 4$  nm in an amorphous alumina matrix. The material gives an ensemble of metal-like mechanical properties ( $E=195\pm 9$  GPa,  $\nu=0,29\pm 0,02$ ) enhanced plastic behaviour, relatively high hardness ( $H=10$  GPa), full compactness and strong adhesion.  $\text{Al}_2\text{O}_3$  barriers have been tested up to  $600^{\circ}\text{C}$  in stagnant HLMs with outstanding results and their performance under heavy ion irradiation investigated up to 150 dpa at  $600^{\circ}\text{C}$ ; the energy of the ions having been chosen to obtain the maximum displacement damage at the interface with the substrate. The irradiation did not induce any loss of adhesion or delamination effects. Complete characterization with pre-normative purpose will require evaluation of: (a) performance

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<sup>116</sup> Patent: EP1896627; cladding tubes made of ferritic/martensitic or austenitic steel for nuclear fuel elements/fuels and method for subsequently treating a FeCrAl protective layer thereon that is suited for high temperatures.

under neutron irradiation; (b) fracture toughness by micro-indentation; and (c) resistance to erosion by high speed HLM flow.

### ***Detonation gun spraying***

Detonation gun spraying is a thermal spray process to deposit and overlay thick, hard coatings on steels, with an extremely good adhesive strength, low porosity and with compressive residual stresses, for parts subjected to wear and erosion/corrosion. The combustion of a gas mixture, ignited by a spark plug in the detonation chamber, generates high pressure shock waves that propagate through the gas stream. The hot gases travel through the barrel at a high speed and high temperature, carrying the material to be deposited in a plastic state with a velocity of 1200 m/s. The particles impacting on the surface form a splat and the coating results from several splats. The temperature and speed of the particles lead to dense and adherent layers. Due to the size of the splats and the residual stresses generated during cooling, the minimum applicable protective layer is around 10  $\mu\text{m}$ . Although the powders reach temperatures up to 4000°C, the deposition does not affect the substrate temperature, which remains cold. The residual stresses can be reduced by interposing a buffer layer on the substrate. Work is needed, here too, to optimise the process and to evaluate the performance of the coating under service conditions. Because of the step variation of the mechanical properties between steel substrate and ceramic coating, cyclic loads are expected to be especially damaging.

### ***Atomic layer deposition (ALD)***

ALD is a well-known deposition technique, largely used for depositing thin films for a variety of applications, mainly: organic electronics; semiconductor processing; and nano-mechanics coating. More and more miniaturization has produced very high aspect ratio structures that need to be coated accordingly. Moreover, the need for continuous and pinhole-free thin films (i.e. the well-known ultra-low gas diffusion barriers) is matched perfectly only by atomic layer deposited films. ALD is based on binary (or more) reaction sequences where two surface reactions occur and deposit a binary (or more) compound film. Since there is a finite number of surface sites, the reactions can only deposit a finite number of them. This allows a precise thickness control by a self-limiting reaction behaviour and an excellent step coverage and conformal deposition on high aspect ratio structures. To be defect free and compact are fundamental features required for barriers to tritium permeation and also for anti-corrosion coatings for future nuclear fission reactors. Currently, an advanced ALD process is under development; the process is suitable to produce a high performance  $\text{Al}_2\text{O}_3$  coating as a barrier against tritium permeation and Pb-Li corrosion. Preliminary results are related to the optimization of the morphology and controllability of the process for the most common oxides such as  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$ . By tuning the process parameters, such as deposition temperature, precursor temperatures, purging time, pumping time and reaction time, an ALD regime can be obtained. More studies should be performed in order to qualify the coating.

### ***Qualification of coated materials***

The qualification of coated materials is especially delicate because, bearing in mind the licensing of the component, it will be necessary to prove that no major safety-threatening consequences will ensue in the event of (local) failure of the SL protection. Essentially this implies foreseeing not only tests aimed at verifying the stability of the protective SL and standard qualification procedures, but also to include the qualification of the substrate material. These are needed in order to know which corrosion-rate or general effects should be expected in case of SL failure. Worst case condition testing is also needed, according to criteria that will necessarily depend case by case.

**Welding** coated components may also pose issues because of the likely risk that in the process the protection is lost.

### ***Prospective materials***

None of the materials solutions listed in the previous sections can be truly called new or innovative. These are all materials that have been long considered, but that still require significant work in terms

of composition, manufacturing processes and property screening before a final codifiable material emerges and reaches sufficiently high TRL. Here two truly new classes of materials with promising features, but for the moment never used in any technological application, are discussed, namely high entropy alloys (HEA) and MAX phases.

### **High-entropy alloys**

High entropy alloys (HEA) are a fundamentally new metallic material concept proposed in recent years.<sup>117</sup> Elements are combined in roughly equimolar concentrations so that, in theory, the high entropy of mixing stabilizes simple solid-solution phases with relatively simple crystal structures, hindering the formation of conventionally expected complex intermetallics. Thus, while these alloys may be compositionally complex, they can be microstructurally simple. HEAs exhibit high strength due to their compositional complexity (solute strengthening), and are thus considered promising for high temperature applications. HEAs containing passivating elements, such as Cr, Al, etc., have shown equivalent or superior resistance to corrosion compared with conventional alloys in aggressive environment.<sup>118</sup> These good mechanical and corrosion properties make HEAs attractive wherever extreme service environments exist, such as in nuclear. Relatively little is known, however, about their stability under neutron or ion irradiation. The metastability associated with the energy stored in the lattice distortions could ultimately lead to the evolution toward the amorphous state under irradiation, or induce segregation and precipitation against thermodynamic forces. In some cases, the existing results indicate that they can be excellent irradiation-resistant materials.

The concept of a crystalline phase stabilized by the entropic contribution of the free energy is extremely attractive and offers the possibility to tailor via a suitable alloy design the desired thermo-mechanical, corrosion and radiation resistance properties, by modifying the composition without a priori incurring in precipitation and segregation issues. The field offers therefore wide opportunities to explore, discover, and develop new classes of alloys for structural and functional applications. Alloying element combinations previously perceived as questionable, due to microstructure instability, may now become a possibility, suggesting completely new families of light metal alloys, high strength metals, and high-temperature metals. Further research could be devoted to explore their applicability to the GenIV systems. Beyond the necessary efforts required to develop a totally new class of alloys, that involves their synthesis, mechanical testing, corrosion testing, irradiations etc., it is important to emphasise that in this case alloy design cannot currently be supported by CALPHAD type models: databases need to be developed, opening a wide field also for modelling as guidance for an accelerated development (section TA.1.1.2).

### **MAX phases**

The MAX phases are layered solids with hybrid metallic-ceramic behaviour and properties that depend on stoichiometry, given by the general formula  $M_n+1AX_n$ , where M is an early transition metal, A is an A-group element (Al-S, Ga-Se, Cd-Sb, Tl-Bi), and X is C or N, while n is typically 1, 2 or 3.<sup>119</sup> They are versatile materials, whose properties (deformability, thermal stability, oxidation resistance, etc.) can be tailored by forming solid solutions on the M, A and X sites, that often exhibit better properties than the 'parent'.<sup>120</sup> The machinability of the MAX phases is similar to that of graphite, which makes them suitable materials for the production of geometrically complex components. Finally, these materials are characterized by unusually high –for ceramics– damage tolerance (e.g.  $K_{IC}$  values of up to  $18 \text{ MPa}\cdot\text{m}^{1/2}$  were reported for textured  $\text{Nb}_4\text{AlC}_3$ ), due to various toughening mechanisms. In terms of their response to irradiation MAX phases seem to have a remarkable capacity for self-annihilation of neutron-induced defects at elevated temperatures.<sup>121</sup>

<sup>117</sup> Yeh et al. *Adv Eng Mater* 6 (2004) 299; Huang et al. *Adv Eng Mater* 6(1-2) (2004) 74; Yeh *Ann Chim Sci Mat* 31(6) (2006) 633; Zhang et al. *Adv Eng Mater* 10 (6) (2008) 534.

<sup>118</sup> Liu et al. *Transactions of Nonferrous Metals Society of China* 25 (4) (2015) 1341.

<sup>119</sup> Barsoum, *MAX Phases: Properties of Machinable Ternary Carbides and Nitrides*, 2013 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.

<sup>120</sup> Lapauw et al., *Inorganic Chemistry* 55 (2016) 5445; Tunca et al., *Inorganic Chemistry* 56 (2017) 3489.

<sup>121</sup> Tallman et al. *J. Nucl. Mater.* 468 (2016) 1; *ibidem* 484 (2017) 120; Ang et al. *Scripta Materialia* 114 (2016) 74; Ang, et al., *Journal of the European Ceramic Society* 37 (2017) 2353.

They are not, however, refractory materials, i.e. they will have a limitation in terms of operating temperature, dictated by their stability in the specific working environment.<sup>122</sup>

Because of their excellent compatibility with HLM they are promising core materials for HLM-cooled GenIV systems. For example, exposing  $\text{Ti}_3\text{SiC}_2$  to HLM with [O] in the  $10^{-8}$ - $10^{-6}$  mass% range, between 550-750°C range and up to 4000 h showed the formation of a thin oxide ( $\text{TiO}_2$ ) scale and no liquid metal attack.<sup>123</sup> Moreover, screening mechanical tests on selected MAX phases in oxygen-poor LBE at 350°C showed no mechanical property degradation.

As with every innovative nuclear material, the MAX phases need to be optimised for the envisaged application. Optimisation involves:

- Selection of appropriate composition followed by microstructural tailoring, playing with the possibility of forming solid solutions M, A and X sites, so as to meet the property requirements of the targeted end application. The appropriate composition can also be determined in order to limit the end-of-life component activation.
- Phase purity: it is often challenging to produce MAX phase materials without ‘parasitic phases’, such as binary carbides and intermetallics. Phase purity can be seriously improved by making solid solutions or by the addition of critical dopants, but the right approach to making phase-pure MAX phases is chemistry-specific and labour-intensive. Phase purity is likely to affect both the radiation tolerance of the MAX phase materials (possible cracking due to differential swelling) and their corrosion resistance.
- Collection of statistically-relevant experimental data: the design and optimisation of MAX phases for selected Gen-IV applications involves application-driven material processing, mechanical testing in both inert and liquid metal media, corrosion/erosion testing and irradiation. The collection of statistically-relevant data could be accelerated with appropriate design/production/performance assessment strategy.

## TA.2 Commonalities with GenII/III nuclear reactor materials

### TA.2.1 Integrity of structural materials

The first common area between GenII/III and GenIV nuclear technologies concerns the integrity (performance and ageing) of structural materials. Four topics for collaboration have been identified:

- Development and qualification of welding procedures, including the analysis of residual stresses  
Failure in metallic components often occurs in welds, so the integrity of welds is critical for the safe performance of nuclear components. Among the different factors that impact on the integrity of welds, residual stresses play an important role. Residual stresses in turn depend among others on the basic material properties and on the welding process. This issue is of major interest for both Gen II/III and Gen IV communities and austenitic steels are of interest as reference materials.<sup>124</sup>
- Testing and qualification procedures for miniaturised specimens for both mechanical characterization and crack growth under environmental conditions  
Miniaturised specimen testing would be beneficial to characterise mechanically neutron irradiated materials to limit activity handling issues and to optimize the use of the limited amount of neutron irradiated material that is generally available.<sup>125</sup> The issue as to whether these specimens can be used to predict real component behaviour is addressed in both communities. Validation of specimen geometry and testing procedure, undertaken by inter-laboratory exercises to achieve the standardization of the procedures, will benefit from an enlarged number of laboratories involved. The determination of the fracture toughness and crack growth rate is also of common interest.

<sup>122</sup> Low, Thermal Decomposition of MAX phases, <https://www.azom.com/article.aspx?ArticleID=6711> .

<sup>123</sup> A. Heinzl, et al., Journal of Nuclear Materials 392 (2009) 255.

<sup>124</sup> See section TA.1.1.1, page 107.

<sup>125</sup> See section TA.1.1.1, page 108.

- Advanced characterisation and multi-scale modelling of microstructural evolution under irradiation  
Radiation hardening of steels may compromise component integrity. Hardening appears already after fractions of dpa round 300°C and increases further with dose, generally saturating only after several dpa. In steels, hardening is the primary driver of embrittlement. In both RPV steels and F/M alloys, impurities and solute atoms form radiation induced clusters that are rich in Si, Mn, P, Ni (and Cu). The nature, formation and evolution of these features depends on many variables, chiefly composition (influencing thermodynamic but also diffusion mechanisms of chemical species, point defects and their clusters) and irradiation conditions (temperature, dose and dose rate). Developing models to describe the formation and evolution of these nano-features is accordingly the goal of several Euratom projects of both Gen II/III and Gen IV communities and could be addressed jointly.<sup>126</sup>
- Ion irradiation as a neutron irradiation surrogate to gain better understanding of microstructural evolution under irradiation and improve identification of radiation resistant materials.  
Due to the high cost of neutron irradiations and to the scarcity of facilities, irradiation experiments using alternative irradiation sources such as ions and charged particles are clearly of interest for both communities. These contribute to materials screening and improvement, as well as to model development/validation. The issues to be addressed are common, in terms of transferability of results between charged particle and neutron irradiation environments, i.e. specific PIE, development of models, etc.,.<sup>127</sup> Collaboration will thus benefit both communities

### TA.2.2 Fuel and cladding materials

A second area where close collaboration will be especially profitable for both GenII/II and IV communities is research on nuclear fuels and claddings.

As far as qualification and assessment of procedures for safety and integrity are concerned, the following topics are of special common interest.

- Safety of oxide fuels  
In the short term, research on oxide fuels and in particular MOX is of significant common interest. Needs are identified in: (i) measurement of safety relevant properties as a function of composition and burn-up, (ii) assessment of behaviour during irradiation, including post irradiation examinations (PIE), and (iii) continued improvement in safety in conventional synthesis technology.
- Innovative fuels and synthesis routes  
In the longer term, increased sustainability can be reached through increased nuclear fuel recycling and MA transmutation, which require the development of innovative fuels and fuels allowing the burning of Pu and MA.
- Fuel performance codes development and validation  
To meet future requirements it is essential that the fuel performance and safety codes are continuously improved and validated by reducing uncertainties and extending experimental data.

In addition, the further development of advanced mechanistic and multiscale modelling tools and the execution of separate effect experiments and detailed materials characterization, used in complement to technological research, are all areas of common interest for both existing and innovative fuel designs fuel types.

The materials and coatings that are considered as accident tolerant claddings for Gen II/III reactors, because of their high temperature and corrosion resistance, are in some cases common to Gen IV future reactors e.g. silicon carbide composites (SiC<sub>r</sub>/SiC), innovative steels (FeCrAl including ODS grades), etc. The main issues are standardisation, joining, and qualification for the reactor

<sup>126</sup> See section TA.1.1.2, page 111.

<sup>127</sup> See section TA.1.1.2, page 116.



environment. Advanced fuel types (e.g. nitrides) and advanced fuel synthesis technologies for accident tolerant fuels are also areas to be addressed together.

Finally, the experimental facilities enabling the manufacturing and characterisation of fuels need to be maintained and expanded to support both the current fleet and the development of future reactors. It is therefore essential that support is provided jointly to irradiation facilities, hot cells and laboratories.

### TA.2.3 Innovative LWR Designs and Technologies

Three potentially common research areas are found also with the field of design of innovative LWR:

First, even though the Supercritical Water Cooled Reactor (SCWR) is not part of the ESNII portfolio and materials for the SCWR are not included per se in the EERA-JPNM activities, the SCWR is a Gen IV concept. Material testing in contact with this type of coolant is consistent with the general need to study the corrosion behaviour of advanced materials, including welds and joints, developed for other environments, as well as to develop materials that are resistant to corrosion in hostile environments. In particular, it is likely that similar advanced material mitigation strategies will have to be envisaged, i.e. ceramic coatings, alumina-forming steels, etc.<sup>128</sup>. Therefore, it is possible to foresee future joint projects in which the qualification of materials, in particular new material solutions, is extended to all coolants of interest for GenII/III and Gen IV, from liquid metal to gas, water and SCW.

The development and application of advanced or novel materials manufacturing processes constitute important subjects for collaboration. Powder metallurgy offers advantages for the production of components for Gen II/III reactors and is also the technique used for the production of ODS alloys: thus, there are opportunities for cross-fertilisation. Moreover, 9Cr ODS steels have improved resistance in SCW compared to conventional 9Cr F/M steels. There may also be the opportunity for involvement of industry in ODS alloy production. Another advanced/novel manufacturing method that could be of interest for NUGENIA and EERA-JPNM is additive manufacturing (3D printing). The latter allows manufacturing of components of complex geometry and is in particular interesting for small quantities. Projects devoted to similar issues that are cross-cutting through NUGENIA and EERA-JPNM could thus be considered in the future.

Finally, the development of new materials is a cross-cutting issue throughout the Gen IV concepts. Materials more resistant to corrosion, in particular austenitic steels and Ni-base alloys, may be of interest for application not only in the SCWR, but also in water-cooled SMRs, given that in several of these concepts the fuel is envisaged to remain in the reactor until the end of its exploitation, without intervention for fuel replacement or reshuffling. This implies that sufficient guarantees need to be provided in terms of the capability of the core materials, which keep together the fuel assemblies, to not fail for the whole reactor lifetime due to, for example, irradiation-assisted stress corrosion cracking. In this framework, coatings, surface treatment (e.g. shot peening) or alumina forming steels may be an interesting option to be explored.

### TA.3 Commonalities with materials for (V)HTR

The existence of subjects of common interest and common challenges related to materials between NC2I and EERA JPNM stems in particular from the strong similarities between GFR (ESNII system) and (V)HTR, which is the core business of NC2I. Except for the fact that the latter includes the use of graphite as moderator, many components are similar or subjected to the same requirements, namely high temperature operation in contact with flowing pressurized helium. Therefore the materials of interest are largely the same.

High temperature resistant materials are being developed within the EERA JPNM for use either in the second phase of prototypes/demonstrators or in commercial GenIV reactors.<sup>129</sup> For intermediate heat exchangers, insulating structures, control rods, and other internal structures such as core

<sup>128</sup> See section TA.1.1.3, page 129.

<sup>129</sup> Issues mainly addressed in section TA.1.1.3.



restraints, core belts, core barrel and fuel cladding, these materials include ODS alloys and composite ceramic materials like SiC<sub>f</sub>/SiC, as well as, to a lesser extent, refractory materials and Ni-based alloys. The latter (Inconel 617, Haynes 230, including here also Alloy 800 as high-Ni stainless steel) are the reference candidate materials for out-of-core high temperature gas-cooled primary system components of both GFR and V/HTR (intermediate heat exchanger). Some advanced grades such as Inconel 718 and 738, or advanced ODS alloys, are considered to match the requirements of the high temperature helium environment, as well as the severe mechanical load of gas turbine blades. For fuels, materials of common interest would be actinide carbides and nitrides.

In addition, all Generation IV reactors should be designed for a design life of 60 years. Thus the development of design rules and design curve and codification into design codes is a shared problem. The ASME BVPC code Section III Division 5 is now being revised to support the design of the US (V)HTR project and dedicated test programmes and development of associated design rules for 60 years design life are being implemented for a number of materials such as alloy 800H and 316 (austenitic stainless steels), Grade 91 (F/M steel) and Alloys 617 and 709 (Ni-based alloys).

The problems to be addressed jointly concern thus the degradation of the properties of the above materials in operation, due to the synergistic effect of high temperatures, mechanical stresses, radiation and gas coolant environments, accumulated over a long exposure time.

Among them:

- Development and qualification of welding and joining procedures of high performance structural materials, including the analysis of residual stresses;
- Development of fabrication technologies for critical components;
- Investigation of mechanisms related to long term operation due to high neutron dose, such as swelling and irradiation creep, and/or under high temperature: thermal creep, synergies with dynamic strain ageing, etc.;
- Constitutive modelling and description of materials behaviour, from physics-based to engineering models, and damage development at high temperature, as a basis for codification improvements and qualification;
- Micro/nano-sample testing for model validation and condition monitoring;
- High-temperature stability for metals and refractory ceramics (thermal aging effects);
- High temperature materials' property database (tensile strength, creep, creep-fatigue, ...);
- Development and nuclear grade codification of composite and ceramic materials and the high temperature alloys.

## TA.4 Commonalities with fusion materials

Despite the significant differences that exist in terms of functioning principles and therefore design between GenIV fission and fusion energy reactors, a number of materials issues of common interest have been identified.

### TA.4.1 Classes of materials

**Commonalities on materials almost exclusively concern steels.** Fusion imposes stricter conditions than fission, due to the reduced activation requirement to minimise the impact of waste. Specifically, the high Ni content of austenitic steels, materials of choice for the design and construction of GenIV prototypes, raises insurmountable concerns in terms of activation for fusion applications. Austenitic steels are also much more prone to swelling and He production than F/M steels, especially under fusion conditions. Thus **F/M steels are the only possible choice as fusion reactor breeder blanket structural materials.**<sup>130</sup> They can withstand the high neutron flux and energy of fusion (14 MeV), that leads to He production and swelling, up to >100 dpa, and can be produced in reduced activation grades (e.g. EUROFER in the EU, F82H in Japan, CLAM in China, ...). In the long-term improved F/M

<sup>130</sup> D. Stork and al.: "Materials R&D for a timely DEMO: Key findings and recommendations of the EU Roadmap Materials Assessment Group", Fusion Engineering and Design 89 (2014) 1586–1594.

steels with wider operational window have a high potential as structural and cladding materials also for GenIV, in order to increase burnup.<sup>131</sup> Therefore, **F/M steels are the only possible cross-cutting metallic material between fission and fusion.**

**In terms of refractory materials,** ceramic composites such as SiC/SiC have been long considered as structural materials for fusion, due to their especially low activation and excellent resistance to high radiation dose and temperature. They continue to be studied in some countries (e.g. Japan) as a necessary choice for advanced breeding blanket designs, beyond DEMO, so they could potentially be also a topic of joint research. At the moment, however, this material is not in the focus of fusion materials research in Europe.

#### TA.4.2 Compatibility with heavy liquid metals

The use of reduced-activation F/M steels in the blanket of a fusion reactor raises **corrosion** concerns because liquid PbLi eutectic is used in many blanket designs as a flowing tritium breeder and in some designs also as coolant. To mitigate the effect of corrosion, temperature and liquid velocity need to be limited, but even in this case it is considered that **alumina or other protective surface layers are going to be needed.** Even without contact with HLM, in fusion the surface treatment of steel structures is essential to reduce tritium permeation through large thin-walled steel surfaces that separate the coolant and breeding loops. Tritium permeation has to be avoided for tritium extraction efficiency and safety considerations. Rupture of the thin protecting layer that forms on the surface leads to increased tritium permeation, so its functioning and integrity is crucial. However, **the need for protective surface layers raises several questions** regarding the stability of the protection at the temperature of operation, especially under neutron irradiation, the possibility or not of self-healing (very limited or non-existent in the case of contact with PbLi), and the consequences of local rupture of the coating during thermal cycling. These problems are extremely similar to those faced by HLM-cooled systems in GenIV.<sup>132</sup> Thus **protective surface layers for steels and their characterisation, including suitable tests and standardization of corrosion tests, are clearly an issue of interest both for fission and fusion applications.** Moreover, the GenIV fission community has identified **LME of F/M steels** as a major concern, which is **expected to be a serious concern also for fusion,** although to date the fusion community has not given significant attention to this problem. Addressing the problem of LME, with a view to better understanding it and mitigating it, is thus a topic of high value for both the fusion and the fission communities, the latter being in this respect more advanced than the former.

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#### TA.4.3 Codification of F/M steels

The use of **F/M steels** for the design of both fusion and GenIV fission systems is hampered by the fact that these steels **are not yet fully included in the design codes** (e.g. RCC-MRx), except at probatory level. Problems that are either specific to F/M steels (e.g. softening under cycling loads, low temperature embrittlement...), or common to all steels but especially serious for F/M steels (e.g. plastic flow localisation and subsequent loss of elongation after irradiation at low temperature) need to be addressed, based also on the understanding of the relevant physical processes. Different approaches from the case of austenitic steels, which are already codified, are needed. The codification of EUROFER is currently ongoing in the fusion community in view of the licensing of the test blanket for ITER by F4E. The fission community had also been involved in the codification of F/M steels, until these became materials of lesser priority. Many results obtained in the FP7/MATTER project turned out to be very useful for the fusion materials community. Hence **design rules for F/M steels, specifically cyclic softening and plastic flow localisation, can be considered as common ground for the fission and the fusion materials communities.**<sup>133</sup> Linked with the process leading to

<sup>131</sup> In the short-term, the Korean SFR (sodium fast reactor) considers the use of a F/M steels (HT9) for the cladding and another steel of this class (EM10) will be used for the wrapper in ASTRID.

<sup>132</sup> See section TA.1.1.1, page 104, and section TA.1.1.3, page 129.

<sup>133</sup> See section TA.1.1.1, pages 102-107; section TA.1.1.2, page 113; and section TA.1.1.3 page 121.

the codification of F/M steels in general (and specific types of steel in particular) is also the usefulness of a **joint fission-fusion materials database**.

#### TA.4.4 Welding procedures and characterization

In fusion components as well as in any other system component, welds are potential weak spots that require attention and are an integrating part of pre-normative materials research programme. Although currently most attention on welds in GenIV is focused on austenitic steels and GenIV relevant environments, while in fusion F/M are the key class of materials, the qualification methodology can be common. **The issues of detecting defects in welds and evaluating their consequences, taking into proper account the presence of residual stresses, are certainly common.**<sup>134</sup> Moreover, any result concerning weldments on F/M steels and their response to irradiation and/or exposure to HLM will be equally valuable for the fusion and GenIV fission communities.

#### TA.4.5 Small specimen testing

Establishing an accepted standardized methodology to extract component-relevant mechanical properties from sub-sized and miniaturized specimens is absolutely crucial for fusion. Fusion reactions release 14 MeV neutrons, thereby producing a spectrum that is significantly different from the fission spectrum and totally irreproducible in fission irradiation facilities. The impossibility of testing materials under fusion-relevant spectra constitutes a serious drawback for the establishment of fusion relevant design rules and for the licensability of fusion breeding blanket components. **Suitable neutron sources reproducing fusion-relevant spectra need to be built**, but these are costly facilities and so far no source of this type is operative. Even when such a source becomes operative, **the volume of irradiated material will be extremely small** (how small depends on the design and the ambition, and therefore the cost, of the source). Therefore **the use of sub-sized and miniaturized specimens becomes mandatory and their standardization represents another ground for collaboration between fission and fusion materials communities.**<sup>135</sup>

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#### TA.4.6 Development of F/M steels with better high and low temperature properties

In the He-cooled DEMO design, as well as in commercial GenIV fission reactors, temperatures in excess of the current limit of 550°C for F/M steels are targeted to increase thermal efficiency and so optimise the use of resources. For this purpose, the creep resistance of F/M steels becomes the property of concern that needs to be improved. Moreover, low temperature embrittlement under irradiation is an unavoidable concern since the blanket of the water-cooled DEMO design is planned to operate between 250 and 350°C. This will make the problem of low temperature embrittlement of F/M steels a key issue to be resolved, even more than for GenIV systems. Thus, **both advanced GenIV systems and fusion share the important goal of improving the creep resistance of F/M steels to above 650°C and of reducing their susceptibility to low temperature (<350°C) embrittlement.** This implies the development and qualification of industrially-scalable innovative materials such as ODS F/M steels or conventional F/M steels with enhanced creep strength. The thermomechanical treatment (TMT) and composition tuning are also expected to improve the ductility, with an impact on reducing low temperature embrittlement.<sup>136</sup> While fusion is subject to the stricter requirement of reduced-activation, the cross-fertilisation in what concerns the criteria used for the tuning of the composition and the choice of TMT is certainly going to be fruitful.

#### TA.4.7 Tools to screen among radiation-resistant materials

Irrespective of the issue of the very specific fusion neutron spectrum, most of the characterization of fusion materials can only be done using fission facilities. However, such irradiation experiments are

<sup>134</sup> See section TA.1.1.1, page 107.

<sup>135</sup> See section TA.1.1.1, page 108.

<sup>136</sup> See section TA.1.1.3, page 121.

costly and the irradiation facilities are scarce. Moreover, for fusion even more than for fission there is a need to produce optimized materials. To date the materials options for DEMO remain very open, given that even Eurofer, which is the current reference reduced activation F/M steel, is likely not to be the final material that will be used for the construction of DEMO if F/M steels with better high and low temperature properties are developed (see above). Therefore, tools to screen among different materials in terms of, in particular, their response to irradiation, are as much needed for fusion as for fission applications. These rely on the **combination of charged particle irradiation, coupled with suitable PIE and, in particular, mechanical property probing techniques on small volumes** (see above). The **establishment of a methodology** to achieve the goal of characterizing materials using these tools is thus yet **another ground for collaboration between fission and fusion materials research communities**.<sup>137</sup>

#### TA.4.8 Advanced modelling

The elaboration of robust rules defining materials behaviour, the identification of appropriate mitigation strategies and the improvement of materials properties depend crucially on the precise understanding of the physical processes that govern the evolution of materials exposed to extreme conditions. Thus, **the continuous improvement of the level of understanding of materials behaviour by exploiting advanced microstructural characterization and evolving physical models is clearly a common issue and goal both for fission and fusion applications**. Modelling activities of specific common interest may concern: microstructure evolution under irradiation; low temperature hardening and embrittlement; irradiation creep and swelling; liquid metal corrosion and liquid metal embrittlement; ODS fabrication processes; precipitation coarsening and recrystallization of oxides or carbides in advanced steels.<sup>138</sup>

### TA.5 Commonalities with materials for other energy technologies

#### TA.5.1 General methodological common patterns

An analysis of materials needs and materials science approaches through energy technologies in EERA, including nuclear, led to the identification of the following common patterns<sup>139</sup>:

- **Ageing and degradation mechanisms studied by combining advanced experimental characterization with multiscale modelling.** This approach is ubiquitously used and perceived as the only way to really advance towards extending the lifetime of materials and/or improving their performance. Even if the problems addressed differ with the energy technology, in most cases the (experimental and modelling) techniques and the approaches used will be similar, as similar methodological problems need to be addressed and solved, irrespective of the specific type of materials or issues studied. In particular, a very general problem that can be jointly addressed concerns the establishment of protocols for the application, analysis and comparison with simulation results of microstructural examination techniques<sup>140</sup>. The problem of accelerated testing (key to address long-term ageing) partly enters this topic, too, because it may be solved by combining specific testing techniques with models that allow the safe extrapolation to much longer exposure times.
- **Characterization of energy materials and devices: contribution of large scale facilities, as well as in situ and operando techniques.** The use of large scale facilities for materials exposure and testing/characterization is a need through all energy technology materials. In particular, in situ and operando characterization techniques have been identified as key for studying materials in working devices, although the requirements and possibilities to actually do so heavily depend on the energy technology. In the case of nuclear materials, operando techniques have currently

<sup>137</sup> See section TA.1.1.2, page 116.

<sup>138</sup> See section TA.1.1.2.

<sup>139</sup> [http://www.eera-set.eu/wp-content/uploads/EERA-JP-workshop-Materials\\_for\\_Energy\\_report.pdf](http://www.eera-set.eu/wp-content/uploads/EERA-JP-workshop-Materials_for_Energy_report.pdf)

<sup>140</sup> See section TA.1.1.2.

limited application but it may be very interesting to interact with other energy technologies in connection with the issue of materials controllability.

- **Rational design of materials supported by modelling.** For all technologies there is a need to find new or alternative materials to improve the performance and the durability of the devices, to decrease their costs or to avoid critical or hazardous elements. Examples are non-noble metal based catalysts, or photovoltaic cells free of hazardous elements, such as cadmium or lead. This calls for an approach based on rational design supported by modelling, which is common to all energy technologies. In particular, high throughput approaches are strongly pursued. The extent of applicability of these approaches will be different in each case and will depend on the complexity of the phenomena involved and how the adequate functionality of specific materials can be quantified, but the trend towards innovation remains defined.

### TA.5.2 Materials for high temperature applications

In addition to the above common general patterns, resistance to high temperature has been recognised as a requirement for materials in a wide spectrum of energy technologies. First, irrespective of the origin of the heat, the efficiency of thermodynamic cycles operating between two heat reservoirs is improved by increasing the temperature of the hotter one. Moreover, some energy production systems inherently require high temperature in order to function because of the physical-chemical processes involved. Thus, **low carbon energy technologies as different from each other as fuel cells and hydrogen, concentrated solar power, bioenergy, geothermal, GenIV nuclear fission, and fusion, find commonalities in the need to operate at temperatures above, and sometimes well above, 400°C.** During the transition to a fully low-carbon economy, this problem affects also clean fossil fuel plants, which also will need to operate at the highest temperature possible to increase efficiency and minimize emissions. In this context it is worth noting the European Creep Collaborative Committee (ECCC)<sup>141</sup>, which was formed in 1991 to co-ordinate Europe-wide development of creep data for high temperature plants. ECCC has probably the largest and most complete set of creep data in Europe for parent materials and welds, and regularly issues guidelines on data generation and assessment methods. In 2017, France decided to rejoin after 10 years absence and ECCC started a new activity for nuclear applications focusing on 316SS and long-term creep.

Temperatures in excess of 400°C, approaching and sometimes even exceeding 1000°C, together with thermal cycles imposed by power fluctuations, inflict severe thermomechanical stresses on the plant components. This requires the use and development of materials that should be proven to maintain their integrity and properties for a sufficiently long time at high temperature and/or when subject to thermomechanical fluctuating loads, within reasonable costs. Moreover, high temperature operation requires efficient cooling, leading to the production of superheated steam or to the use of alternative coolants like liquid metals, molten salts or gases, to which materials will have to be exposed. Environmental degradation due to salt water also affects other technologies, such as off-shore wind turbines or ocean energy. Thus materials need to be also resistant to the attack of these environments.

The components that are mainly affected by high temperature and environmental degradation problems are, quite clearly, those where heat exchange and transfer take place, either physically or chemically, such as boilers, turbines and pipes, in addition to absorbers, receivers, electrodes and catalysts ... Some of these components have structural functions, their integrity therefore also having safety implications. Others need to maintain their properties mainly for system efficiency, and therefore also economic, reasons.

The materials that are selected and qualified to demonstrate they are able to maintain their properties at high temperature and in aggressive environments, also need to be joinable or weldable, fabricable, formable and - importantly for many renewable forms of energy - low cost.

**Almost irrespective of the energy technology considered, only a few classes of materials can potentially meet all these requirements,** with different levels of quality in their response:

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<sup>141</sup> <http://www.ommi.co.uk/etd/eccc/advancedcreep/com.html>



- Some materials are currently used or are already commercially available, but need to be demonstrated to be suitable for the application. These are: creep-resistant F/M steels; austenitic steels; Ni-base alloys; and ceramics, mainly silicon carbide composites ( $\text{SiC}_f/\text{SiC}$ ) and alumina based ceramics. Often, they need appropriate coatings as protection from environmental aggression.
- Other are incremental improvements on the above materials that offer potential for better efficiency or functionality of the system/component. These are developed to different TRL and are: CSE F/M steels that may operate up to 670°C and ODS steels for operation above 700°C; protective and durable coatings suitable to withstand cyclic operation or alumina-forming austenitic (AFA) or ferritic ( $\text{FeCrAl}$ ) steels; advanced composites ( $\text{SiC}_f/\text{SiC}$  with improved oxidation/corrosion stability,  $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ , mullite/mullite...). Importantly, for these new materials joinability and industrial production should be ensured.
- Finally, prospective materials are: oxide ceramics with infiltrated nano-catalysts; refractory metallic alloys (V, Mo, W...); MAX phases.

**This list of materials proves once again to have much in common with GenIV structural materials.**

The qualification of materials in service environments and the development of new material solutions to enhance system efficiency or increase component availability and lifetime are costly activities. In the nuclear field, however, the strict safety requirements imposed by regulatory bodies and the harsh service conditions have obliged nuclear research to devote large efforts to materials science and the nuclear industries to devote substantial resources for materials research. This has enabled advanced material science competences to be developed and sophisticated facilities and infrastructures for materials research to be built in the nuclear field. These competences and facilities can become now useful and beneficial also for other energy technologies, especially renewables, where there is no room for investment in new material solutions, due to the cost constraints related to low energy density or intermittent energy production, and therefore low profitability. Thus, cross-cutting projects on high temperature materials represent, in principle, win-win situations for different energy technologies where nuclear and renewables collaborate.

### TA.5.3 Specific commonalities with concentrated solar power

**Specific commonalities can be identified and targeted between materials issues for GenIV fission energy and concentrated solar power (CSP).** Current large CSP solar tower systems, which are at a commercial stage, are still affected by the limited operation of the ceramic solar receivers, which would largely benefit from the use of  $\text{SiC}_f/\text{SiC}$  to extend life and working temperatures above 800°C (air temperature).<sup>142</sup> Different CSP systems are being developed, from "large tower" systems to small domestic ones: it is the limited reliability of ceramics materials for the receiver that hinders the commercial success of these systems, especially large "solar tower" systems (>5MW<sub>e</sub>), which are otherwise at an advanced maturity level after over a decade of intense R&D and are poised to achieve 20 years of maintenance-free operations. Moreover, new receivers, made of composite ceramics (or ODS steels) with increased resistance to oxidation and thermal shock, would boost innovative smaller "dish systems" that should withstand working temperature of about 900°C and can be coupled directly to a gas micro-turbine. Finally, solar thermal energy storage is a crucial need to reduce the energy supply fluctuation that is inherent to intermittent sources like the Sun. A credible solution rests on the deployment of liquid lead, to be heated and stored in insulated tanks at temperatures as high as possible, up to 700 or even 800°C. These tanks will suffer from very similar environmental attacks from the flowing lead as the inner vessel wall of lead-cooled reactors. The materials and material solutions considered for GenIV applications, and the relevant research, are therefore expected to be of use for concentrated solar power; in turn, solar thermal energy

<sup>142</sup>  $\text{SiC}_f/\text{SiC}$  is also experiencing renewed interest in the aeronautical industry amid technological advances in jet turbines that could substantially reduce its costs.



environments offer extreme conditions to test materials solutions that may become useful also for GenIV nuclear applications.<sup>143</sup>

## TA.6 Infrastructure for nuclear materials R&D

The research activities described above have as an essential prerequisite the availability of suitable facilities and infrastructures for materials qualification through exposure to conditions representative of service and subsequent characterization, both mechanical and microstructural. Modelling, on the other hand, implies the availability of suitable computational facilities.

The three service conditions of interest for GenIV reactor nuclear materials are **high temperature, contact with coolants and a high level of irradiation**. The first two can be of interest and application also for non-nuclear technologies (e.g. facilities for tensile and fracture toughness tests, standard creep and corrosion tests, slow strain rate tests in environment...) and may be in some cases even redundant. Moreover, microstructural studies on materials exposed to corrosive environment and/or high temperature can be performed in many laboratories. In contrast, **irradiation is quite specific of nuclear materials and irradiation facilities are scarce**, as well as are scarce the infrastructures that allow **testing and examination of irradiated materials**. Therefore, this section focuses mainly on facilities for exposure to irradiation and handling of irradiated materials. Notwithstanding the focus of this section, it is clear that equipment in **furnaces** to perform for example very long-term creep and creep-fatigue testing, or **loops** for exposure to flowing coolants, are **crucial facilities** that are in fact not so common and may not be available in sufficient quantity for a full qualification of materials for sustainable nuclear energy.

### TA.6.1 Irradiation facilities

#### TA.6.1.1 Fast neutron facilities

Neutron irradiation facilities and associated 'hot' cells laboratories are a central necessity for performance and safety testing of all types of structural and fuel materials. Since the ESNII demonstrators are all fast neutron spectrum systems, materials for their construction should be qualified in fast neutron irradiation spectra. Unfortunately, there is currently no fast neutron power or testing reactor operating in Europe. Worldwide, fast neutron testing reactors are limited to BOR-60<sup>144</sup> in Russia, CEFR<sup>145</sup> in China, FBTR<sup>146</sup> in India and JOYO<sup>147</sup> in Japan, the first one being the only one exhibiting significant availability; the last one currently not in operation. Russia also hosts two power SFRs, BN-600 and BN-800, running on MOX fuel, but these are in principle not available for materials qualification experiments, even if some fuel tests could be envisaged. To redress the situation, **a fast system should be built in Europe**, but this leads to a vicious circle because fast neutron data are needed for this purpose. This means that **at the moment Europe is totally dependent on non-European countries to have access to fast neutron flux facilities**. The construction of at least one of the ESNII demonstrators within the coming two decades is thus essential in order to provide Europe with a facility to qualify structural and fuel materials for commercial GenIV reactors.

#### TA.6.1.2 Material Testing Reactors and associated 'hot' cells

Materials testing reactors (MTR) are high power research facilities that can be used to expose materials to operation-like conditions. Flexibility and ability to adapt to changing needs is a fundamental principle for such reactors. MTR can perform a variety of test irradiations

<sup>143</sup> The H2020 project NEXFLOW is an example where materials technologies and competences of nuclear origin are used for concentrated solar power applications.

<sup>144</sup> <http://www.niiar.ru/eng/node/224>

<sup>145</sup> <http://www.ciae.ac.cn/eng/cefr/index.htm>

<sup>146</sup> <http://www.igcar.gov.in/romg/fbtrdesc.htm>

<sup>147</sup> <https://www.jaea.go.jp/04/o-arai/joyo/english/joyo/roshin.html>

simultaneously, the number of these depending on their design and on the nature of the tests. In specific cases, MTR can be augmented by loops emulating the coolant flow conditions in power reactors, although in practice devices of this type are very limited in existence and availability – and none exist in a fast spectrum. Almost all MTR have on-site ancillary ‘hot’ cells to make preliminary or detailed examinations, as well as to enable packaging and distribution of samples to other laboratories. Among the approximately 40 MTR currently operational in the world, Europe hosts seven (see Table TA.1 **Error! No se encuentra el origen de la referencia.**), the most prominent ones being the High Flux Reactor (HFR)<sup>148</sup> at NRG, Petten (The Netherlands) and the BR2 reactor<sup>149</sup> at SCK•CEN in Mol (Belgium).<sup>150</sup> These MTR can be used for fuel testing and have appropriate ‘hot’ cells available. Other reactors that currently do not have a license to handle MOX fuel, but are available for structural materials’ investigations, are LVR-15<sup>151</sup> at CVR in Řež (Czech Republic), including the experimental loops for in-pile material testing under PWR/BWR, but also SCWR and high temperature He conditions, MARIA<sup>152</sup> at NCBJ in Świerk-Otwock (Poland), and the TRIGA II reactors at RATEN ICN in Pitesti (Romania)<sup>153</sup> and at IJS in Ljubljana (Slovenia)<sup>154</sup>. These centres are equipped with ‘hot’ cells, as well. In addition, materials still remain to be analysed from experiments performed at the Osiris reactor (Saclay, France), which was shut down at the end of 2015. The Jules Horowitz Reactor (JHR) is under construction at the CEA Cadarache site (France) to replace the capacity lost due to the shut-down of Osiris. **Europe’s neutron irradiation capacity is currently very limited** and even with the soon to start JHR, should no further new construction occur, the **opportunities to test new materials in reactors will be restricted to the extreme.**

For irreplaceable component materials, the doses expected at the end of life can be reached in existing MTR, although in order to have exposure times much shorter than the real ones the fluxes used are much higher. While this may be a problem especially when the operating temperatures are high and the synergy between mild irradiation and thermal ageing and/or thermal creep may lead to unknown effects, MTR available in Europe are at least usable to reach end-of-life doses, with the only caveat of the different neutron spectrum. However, for fuel and replaceable component materials, especially cladding, the doses expected in service are much higher than those accessible in thermal MTRs. The only way round this problem to reach doses up to or in excess of 100 dpa is currently the use of charged particles, as described in the next section.

**Table TA.1:** Materials Testing Reactors currently operational in Europe and their irradiation characteristics.

Name	Location	Maximum fast neutron flux (>0.1 MeV) [ $10^{14}$ n /cm <sup>2</sup> s] (from 155)	Dose rate [dpa <sub>Fe</sub> /fpy]	Accessible temperatures [°C]
BR2	Mol, Belgium	7	Up to 5	50-1200
HFR	Petten, The Netherlands	5.1	<7	80-1100
LVR-15	Řež, Czech Republic	3	~1	50-850
MARIA	Świerk-Otwock, Poland	1	~1	50-100
Triga Pitesti	Pitesti, Romania	1.8	2.5	80-300 (→500 future)
Triga Mark II	Ljubljana, Slovenia	0.06	< 0.01	20-50 (→300 future)

<sup>148</sup> [http://iet.jrc.ec.europa.eu/sites/default/files/documents/brochures/hfr\\_mini\\_blue\\_book.pdf](http://iet.jrc.ec.europa.eu/sites/default/files/documents/brochures/hfr_mini_blue_book.pdf)

<sup>149</sup> <https://www.sckcen.be/en/Research/Infrastructure/BR2>

<sup>150</sup> The HBWR at IFE, Halden (Norway), has been recently definitively shut down.

<sup>151</sup> <http://cvrez.cz/en/infrastructure/research-reactor-lvr-15/>

<sup>152</sup> <https://www.ncbj.gov.pl/en/o-nas/maria-research-reactor>

<sup>153</sup> <https://www.nuclear.ro/en/departments/triga.php>

<sup>154</sup> <http://www.rcp.ijs.si/ric/description-a.html>

<sup>155</sup> <https://nucleus.iaea.org/RRDB/RR/ReactorSearch.aspx>

### TA.6.1.3 Charged particle irradiation facilities

Charged particles can be electrons, protons or ions of different mass. Although the penetration of protons and light ions can be significant, all these particles are obviously efficiently stopped by electrons and ions inside the materials, thereby affecting only limited volumes, insufficient to produce standard specimens for e.g. mechanical property testing. Moreover, there are spectral differences with respect to neutrons in terms of damage that is produced and the progressive slowing down results in damage production gradients, while chemical species initially absent in the material and/or atoms in excess are injected in the target material, including sometimes unwanted impurities that are difficult to control, e.g. carbon. Despite these shortcomings and limitations, which certainly prevent their use for full qualification purposes, charged particle irradiation is useful to get insight into the behaviour of materials under irradiation.

**A significant number of facilities permitting charged particle irradiation, which can be exploited for modelling and screening purposes, exist in Europe at nuclear research centres and universities, for example the JANNuS facility at CEA Saclay /U. Orsay and the ion beam centre at HZDR:**

- The JANNuS multi-ion beam irradiation platform consists of two facilities: (1) JANNuS-Orsay, located in Orsay at the Centre for Nuclear Sciences and Material Sciences CSNSM, a joint research unit of Centre National de la Recherche Scientifique (CNRS) and Université Paris-Sud; (2) JANNuS-Saclay, located on the CEA Paris-Saclay as part of the Nuclear Materials Department of the Nuclear Energy Division. The capabilities combine a total of five electrostatic accelerators for single beam, dual beam and triple beam ion experiments and a transmission electron microscope for in situ studies.
- The ion beam center of HZDR, Dresden is a European user facility that provides a large variety of ions for materials modification and probing (energy range from 1 eV to 50 MeV, temperature range -175 ... +1000°C); dual beam irradiation experiments (e.g. Fe<sup>+</sup>, He<sup>+</sup>) are possible.

Concerning fuel, while ion irradiation is used to study uranium-bearing fuel behaviour, no charged particle irradiation facility exists for plutonium-containing materials in Europe, nor in the world, except the Casimir facility at CEA Saclay (France)<sup>156</sup>. This facility is dedicated to the ion beam analysis of radioactive materials, but can only be used, with some limitations, to irradiate materials using light ions (H, He, N...). In practice, fuel pins are only qualified for a limited amount of time sufficient to license the first core. Further qualification or the qualification of alternative fuel pins are then performed in the reactor itself, when built.

## TA.6.2 Handling of irradiated materials

### TA.6.2.1 'Hot' Cells and shielded facilities<sup>157</sup>

Materials exposed to neutron irradiation need to be handled, tested and examined in dedicated 'hot' cells or similarly shielded facilities. In particular, capsules taken out of the reactors need to be safely dismantled and 'hot' cells on the sites of all European MTRs are an integrated component of any experiment. For specimen fabrication and large specimen testing, e.g. mechanical testing, dedicated 'hot' cells are absolutely necessary. For microstructural specimen fabrication and characterization, or in other specific cases where the quantity of material to handle is small, simpler types of shielding are often sufficient. But invariably these are very costly infrastructures, both to build and maintain, and are available and employed only in a few equipped and licensed research centres, under severe safety rules for the operators, and only very rarely in universities.

The main 'hot' cell facilities in Europe are located next to the European MTRs (see section TA.6.1.2). In addition some centres are equipped with 'hot' cells even without possessing an MTR, such as the

<sup>156</sup> [http://iramis.cea.fr/Phoce/Vie\\_des\\_labos/Ast/ast\\_sstechnique.php?id\\_ast=361](http://iramis.cea.fr/Phoce/Vie_des_labos/Ast/ast_sstechnique.php?id_ast=361) (in French only)

<sup>157</sup> In addition to those associated with MTRs.

CEA centres of Saclay, Cadarache and Marcoule (France), the Sellafield site of the UK National Nuclear Laboratory (NNL), the Joint Research Center in Karlsruhe (European Commission), Studsvik in Sweden and HZDR in Germany. The largest part of this 'hot' cell capacity was built in the 1970s and 1980s and, despite regular refurbishments, might not be able to be operated much longer, because of the increasing maintenance costs and increasingly demanding safety regulations for this type of facilities. For example two 'hot' cell facilities dedicated to fuel located in Cadarache (LEFCA and LECA) will close in the next few years for these very reasons, while only one full scale replacement is planned to be built: the MOSAIC 'hot' laboratory, which is planned in Cadarache (France) to replace both the LECA fuel 'hot' lab (Cadarache) and the LECl laboratory for structural materials (Saclay) facilities when these close - the funding of this project, however, is not yet guaranteed. JRC is modernising part of the infrastructure with the construction of a new laboratory space (Wing M), to replace partially the aged facilities. The Studsvik 'Hot' Cells are divided into two separate facilities, the HCL-Laboratory with 7 concrete cells, of which two are large enough to allow reception and handling of full length LWR fuel assemblies and pins, and the Active Metal Laboratory, with 11 lead and 8 steel cells primarily for mechanical testing. At the Active Metal Lab, test can be performed at both cladding and structural materials. HCL is primarily used for fuel investigations, failure analyses, material studies and corrosion-related work as well as for refabricating test fuel rods. In addition to this, the facility has access to a chemical analysis laboratory. The site also maintains a pool-type storage and inspection facility. Finally, the 'hot' cell facilities at HZDR, Dresden, can accommodate a total activity of 8 TBq. Besides the preparation and testing of standard samples (tensile, hardness, Charpy impact and fracture toughness), devices for small specimen technology techniques (mini-CT and small punch) are available. The preparation of samples for microstructural techniques (SANS, TEM, APT) is possible.

The number of sites where new 'hot' cell facilities have been recently built or refurbished is limited. The UK has known a recent significant increase of its hot cell capacity, with the complete refurbishment of the Windscale Laboratory and the construction and commissioning of the Phase 1 and part of the Phase 2 of the Central Laboratory, both at Sellafield. In addition, new hot cell complexes have been recently completed at CVR (Czech Republic) and VTT (Finland). The CVR facility contains 10 individual hot cells interconnected by the transport system. Two types of cells are utilized, gamma cells designed for testing the fuel cladding and those for irradiated nuclear power plant structural materials. They can accommodate up to ~300 TBq activity converted to  $^{60}\text{Co}$  isotope. The alpha cells are designed for R&D on radioactive waste processing. VTT has 8 hot cells and a shielded glovebox, designed for PIE of reactor structural materials (specimen fabrication, mechanical testing, analytical microscopy). The VTT 'hot' cells can receive external transports of 3.7 TBq  $^{60}\text{Co}$  activity. Finally, the GENESIS platform in France enables characterisation of irradiated materials: at CEA/DMN Saclay (for highly active materials –in the CEA LECl laboratory, so long as it operates) and at CNRS/GPM Rouen, France (where new installations have been built, able to host activity up to 200MBq/sample). GENESIS is thus a new instrumental platform for nanoanalysis effects of radiation in materials (FIB-SEM, APT, HRTEM).

The limited availability of 'hot' cell facilities, especially those licensed for fuel handling, whose availability has declined steadily over the decades, limits enormously the number of tests, measurements and examinations that can be performed on irradiated materials. This determines the fact that complete post-irradiation examination (PIE) of samples from irradiation experiments may take several years to be completed. This problem is exacerbated by the ever rising costs and difficulties of irradiated materials transport and the fact that current PIE include advanced microstructural examination with new techniques that did not exist when most 'hot' cell facilities were built in Europe. In the case of fuel, the limited 'hot' cell capacity also has important negative consequences on Europe's capability of fuel procurement, since these facilities are indispensable for the preparation and safety testing of new fuel.

Despite fair availability, Europe's aged 'hot' cell capacity is already insufficient today and, as they age further, their replacement will be imperative. Given the time and budget necessary to build and commission a 'hot' cell facility, new replacements should be planned right now at national and/or European level.

## TA.6.2.2 Transportation of samples for dedicated analysis

Suitable, flexible transport flasks and containers for irradiated test specimens are essential to make an effective use of Europe's currently geographically separated test reactors, irradiation facilities and 'hot' cells, as well as of the specificities of the various facilities. The capability to ship irradiated materials to and between appropriate facilities is an essential component of European infrastructure. Currently, in addition to the inherently high costs of this type of shipments, the existence of differences in the laws and rules applied to radioactive material transport through European countries often causes this type of transports to suffer from unduly very large delays, which effectively increase the overall costs, reduce the effectiveness of the research and prevent the timely completion of projects. **Improved coordination and standardisation of regulations and transport containers are here essential to reduce transport times and costs, and to break the strangle hold of ever increasing costs for fewer transports.** A parallel effort can be done in miniaturising the size of the samples to be transported for specific analysis, for example by employing FIB.

## TA.6.3 Computational facilities

Even if the need for computing facilities is not specific to the investigation of nuclear materials, it is worth stressing that the **ICME approach**, as well as the simulations using design and fuel performance codes, are **computationally intensive** and **call for access to world class high performance computing (HPC) systems.**

More generally, this access is essential for international competitiveness in science and engineering. The importance of developing HPC capacity has been recognized by countries such as USA, Japan or France since the 1990's. Europe has recently acknowledged that HPC was a strategic resource for Europe's future and the need for a European-level policy to optimise national and European investments and adopted its HPC Strategy in 2012. It combines 3 elements:

- Developing the next generation of HPC technologies, applications and systems towards exascale computing,
- Providing access to the best supercomputing facilities and services for both industry (including SMEs) and academia complemented with training, education and skills development in HPC,
- Achieving excellence in HPC applications in domains that are most important for Europe.

Concerning the access to supercomputing facilities, the vision is that of a pyramidal ecosystem, with at the top 3 to 5 European-level supercomputing centres (Tier-0), supported by a Tier-1 composed of national centres, in turn supported by a Tier-2 of regional or "meso"-centres.

Industry and SMEs are increasingly relying on the power of supercomputers to come up with innovative solutions, reduce cost and decrease time to market for products and services. This is also true in the development of nuclear systems. Therefore, in addition to the development of the global European HPC capacity, **efforts must be made in the nuclear community to have the investigations on nuclear materials recognized as a top priority subject in Europe and guaranteed access to significant computational resources.** Currently, while fusion research can rely, as part of the EURO-fusion consortium, on a funded HCP facility, **no equivalent exists for fission research.** This is a foremost condition for advanced modelling to bring the appropriate support to the development and qualification of the materials needed for, especially, GenIV systems.

## TA.6.4 General considerations on nuclear infrastructures in Europe

### TA.6.4.1 Costs related to crucial infrastructures build and use

The construction and maintenance of infrastructures and facilities such as MTRs and 'hot' cells is beyond the scope of the EERA JPNM activities and capabilities. It is important, however, to emphasise the importance of this point. In particular, if it is out of place to attempt estimates of infrastructure investment costs here, it is useful to remind that they have been detailed in European

projects such as ADRIANA,<sup>158</sup> and vary considerably, with material testing reactors lying in excess of 1000 M€, and ‘hot’ cells, depending on scope, in the 100-300 M€ range. Typical single fuel irradiation experiments cost 1-2M€, depending on complexity, instrumentation required, and PIE effort. The cost of the transport of irradiated fuel can reach 150 k€, although in a well-planned oriented approach can drop to as little as 15 k€; active structural material specimens can be transported with costs varying between 5-30 k€. The advent of micro sampling techniques (e.g. FIB) for dedicated examination on ultra-small samples can reduce these costs even further.

In addition, Tier-0 HPC resources, which do not serve the whole European scientific and industrial community, have to be renewed every 2-3 years, with construction costs between 200 and 400 M€ and annual running costs around 100 M€/yr.

#### TA.6.4.2 Renewal of nuclear infrastructures

A research agenda can only be fulfilled if it is matched by appropriate and timely available infrastructures. Therefore, in the present context, **in order to maintain its competences and progress further in the field of nuclear materials, Europe should**, through its research organisations located in different MS, and thus with the full commitment of the latter:

- Ensure that a **fast neutron flux facility** comes available soon (e.g. ESNII demonstrator);
- Construct at least one **new MTR in addition to JHR** (i.e. PALLAS, MYRRHA, the latter with the added value for GenIV applications to offer a fast neutron spectrum);
- Renew and possibly **extend its ‘hot’ cell capacity**, taking due account of **transportation issues**;
- Ensure that ‘hot’ laboratory capabilities, in particular for fuels, **do not decrease** further;
- Ensure appropriate access to **high performance computing for the fission community**;

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In addition, three recommendations for policy-makers, that are further developed in the next section, are in order:

1. **Plan** major infrastructures judiciously at a **pan-European** level in a **harmonised** way;
2. Implement consequent **open access** and infrastructure **sharing** initiatives;
3. Foster **joint programming** (fusion energy provides an example to follow).

#### TA.6.4.3 Sharing and joint programming of nuclear infrastructures

**Access to and sharing of facilities for nuclear materials** exposure, testing and examination, especially controlled zones where radioactive materials are manipulated, **can be problematic** for legal, security, safety and financial reasons, namely:

- Legal: Protection of know-how & expertise: there is often reluctance to give full open access as this may reveal details on protected know-how (this attitude is however often inconsistent even within the same organisation);
- Related to security: Access to ‘hot’ cells requires clearance from authorities for security reasons: this takes weeks or months and a significant administrative burden;
- Related to safety: Only trained & skilled operators can safely use some equipment, especially in ‘hot’ labs (manipulators...);
- Financial: Availability of specific equipment has a high cost and opening it for access to external users might limit the profitability of facilities.

Notwithstanding these difficulties, **solutions can be envisaged**, for example a scheme of mutual compensation between organisations within a “virtual research centre”, in-kind or in-cash. An in-kind type of compensation is for example by seconding employees from A to B in a stable way. This could happen under specific bilateral, as well as multilateral, agreements concerning non-disclosure of know-how, use of manpower for host purposes, etc.

<sup>158</sup> [http://ojs.uif.cas.cz/~wagner/transmutace/erinda/presentations/05\\_ADRIANA\\_ERINDA.pdf](http://ojs.uif.cas.cz/~wagner/transmutace/erinda/presentations/05_ADRIANA_ERINDA.pdf).



It should be stressed that **mobility scheme** of this type provide also a **key motor to drive education and training** of not only researchers, but also operators, enabling Europe to maintain and actively manage key competences currently dwindling in the nuclear field, for the benefit of all nuclear research centres.

To facilitate both sharing and mobility, in a context of scarcity of infrastructures and financial means, and in order to foster pan-European harmonised infrastructure planning , **joint programming is key**. In particular, it is believed that the creation of a **permanent joint European planning forum and management scheme of nuclear infrastructures** is a timely need.<sup>159</sup>

The **joint forum** should mainly agree upon **irradiation future needs**, in order to plan and propose **harmonised European investments**. It may also express views on the rules for financial coverage for the use of nuclear infrastructures.

The **joint scheme for the management of nuclear infrastructures** should fulfil the following conditions:

- 1- Be a **single entry point**, i.e. a single interlocutor whom users should address for the use of available infrastructures;
- 2- **Coordinate the best use of infrastructures** for a given experiment, by pooling facilities and experts in Europe, to design the best experiments possible (also the cheapest ...), establishing if and when there are advantages to access non-EU infrastructures;
- 3- Benefit from the **harmonised commitment of MS to pay the costs of the infrastructures and use**, as part of the engagement to innovation, but develop flexible management attracting collaborative projects and also industrial users;
- 4- **Distinguish between R&D users** (low cost open access, selection based on merit of proposal, open access data and results, ... also open to non-MS) **and industrial users** (charged services, data protection, ...).

Within such a scheme, tenders could be launched and/or differences between available facilities could be optimally used to explore specific effects, including, whenever suitable, fundamental studies and the use of ion irradiation. In particular, such a scheme would allow the **best use of available space in reactors** to be made, through the design of joint campaigns. For instance, while it is true that MTR operation is expensive and cannot be offered for free, irrespective of the open access nature of the facility, under some conditions it is possible to perform less expensive "piggy-back" irradiation experiments, in a concerted framework, especially if experiments funded by industrial users can be partially used for this purpose at no detriment of the customer.

Of course, for this scheme to see the light the **willingness of sharing and joint programming must exist at all levels**, including high management of research centres as well as MS policy-makers. This should also be properly fostered by Europe-driven actions. It remains to be seen whether the conditions for this approach to materialise actually exist.

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<sup>159</sup> It could be a European version of the American NSUF: <https://nsuf.inl.gov/>.