

ON THE SEARCH FOR A SAFER NUCLEAR FUEL: CIEMAT'S APPROACH AND RECENT ACCOMPLISHMENTS



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RESEARCH APPROACH

Safety assessment of nuclear fuel necessarily goes through the analysis of its thermo-mechanical performance. This requires a detailed understanding of the physical and chemical properties describing pellet and cladding materials behaviour, as well as of the synergistic effects among thermal, mechanical, and other relevant phenomena such as the release of fission gases in the pellet and corrosion of the cladding. Such an understanding allows the identification of degrading mechanisms that may jeopardize the fuel rod integrity. From a safety point of view, the research focuses primarily on preserving the integrity of the cladding as the first confinement barrier of the fission products.

In this context, the research approach followed by the Unit of Nuclear Safety Research of CIEMAT is based on the understanding and modelling of the fuel rod thermo-mechanical performance from which methodologies for cladding integrity assessment are built. The scope encompasses conventional fuel (UO_2 pellets with Zirconium alloy cladding) and evolutionary ATF's (Advanced Technology Fuels), under in-reactor operating conditions and design basis accidents; in case of traditional fuels, conditions related to interim dry storage are also studied.

Therefore, reliable predictive capabilities of the fuel rod thermo-mechanical behaviour (i.e., fuel performance codes) are indispensable as a basis for the methodologies aimed at this research. This entails activities along three axes:

- Verification and validation of fuel performance codes.
- Model development and enhancement.
- Extension of codes beyond their design domain.

The reference analytical tools used for this purpose are the so-called FRAP family codes developed by PNNL [1], [2]: FRAPCON (latest version 4.0) for steady state (slow ramps included) and FRAPTRAN (latest version 2.0) for transients (design basis accidents such as loss of coolant, LOCA, and reactivity initiated accidents, RIA).

RECENT RESULTS

This section compiles some of the latest accomplishments based on the approach outlined above. By no means, it should be seen as a comprehensive compilation, but just an illustration. For a more thorough insight into every contribution made over the last fifteen years since the described

approach was adopted, specific references may be reached out [3-11].

Codes verification and validation

Changes in fuel rod designs and operation conditions highlight the need for continuous verification and validation of fuel performance codes. CIEMAT contributes to the FRAPCON/FRAPTRAN assessment reporting back to the developers and drawing attention to any rising issue that might happen. The research recently performed by CIEMAT to assess the FRAP-family codes reliability can be classified according to the scenarios addressed: normal operation, power ramps, and LOCA. The following subsections show the assessment carried out in each case.

Normal operation

Among the fuel safety criteria under normal operation conditions [12], the one concerning the limitation of the rod internal pressure (RIP) is relevant not only in reactor to avoid cladding lift-off, but also in dry storage to limit the cladding stress as a driver of degrading mechanisms such as creep or hydride radial reorientation (related to cladding embrittlement). Therefore, RIP should be properly predicted by fuel performance codes, for which they should reliably estimate fission gas release (FGR) and rod void volume along irradiation. The FRAPCON assessment was mostly based on an experimental database focused on FGR [13]. Under the frame of the EJP-EURAD and OCATS projects (funded by EURATOM HORIZON-2020 and ENRESA, respectively), CIEMAT has been working on extending the FRAPCON assessment concerning RIP with an alternative database built on data coming from commercial operation of fuel rods.

The CIEMAT's database consists of 16 data sets available through the open literature or participation in international projects [14], [15]. In addition to End-Of-Life (EOL) RIP measurements (post-irradiation examinations, PIEs), in all the cases the primary variables RIP depends on (i.e., FGR and rod void volume) are reported too. The burnup range covered goes from 45 to 70 GWd/tU, approximately.

Figure 1 shows the code-to-data comparison in terms of RIP at EOL represented as a function of the rod average burnup (FRAPCON's simulations with the recommended FGR model, called MASSIH). The code is found to be accurate for burnups lower than 60 GWd/tU (close to the maximum burnup FRAPCON has been validated so far, 62 GWd/

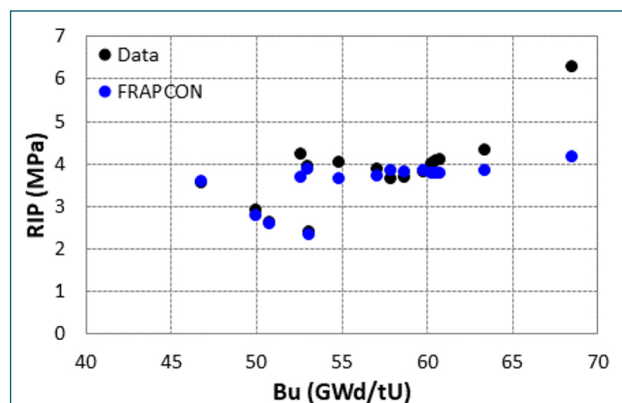


Figure 1. RIP as a function of burnup (Bu).

tU), with an average deviation of around 5% and a maximum one of around 10%. For higher burnups, though, deviations grow progressively up to nearly 30% at 69 GWd/tU. Most importantly, such deviations underpredict measurements; in other words, in case of extrapolation beyond the FRAPCON validation limit, the estimate would not be conservative, which is inconvenient in terms of safety. An analysis of the results pointed to FGR as the main source of discrepancy. Hence, in case FRAPCON is intended to be used for higher burnups than 60 GWd/tU, the FGR modelling of the code might need a proper extension.

Power ramps

The interest in flexible modes of NPP operation has drawn attention to fuel rod responses to "frequent" controlled power changes. Power ramps foster pellet-cladding mechanical interaction (PCMI) and transient (sometimes called "burst") FGR. Both of them are potential precursors of cladding failure. CIEMAT's recent efforts have been oriented towards chromia-doped pellets concerning PCMI, and traditional UO_2 fuels when dealing with FGR.

PCMI

By benchmarking FRAPCON against PIEs data from SCIP-II Aa2 rod (fast power ramp on a fuel with chromia-doped UO_2 irradiated up to 28 GWd/tU) [15], it has been found that the rigid pellet assumption adopted in the code leads to a systematic overprediction of the cladding deformation (Figure 2). This observation has a general nature, but it becomes more significant in chromia-doped UO_2 fuels given the enhanced viscoplasticity of this material with respect to traditional fuel [16]. Therefore, in the case of modelling this type of fuel removing the rigid pellet approximation and considering chromia-doped UO_2 fuel viscoplastic behaviour, would result in less conservative and more accurate results.

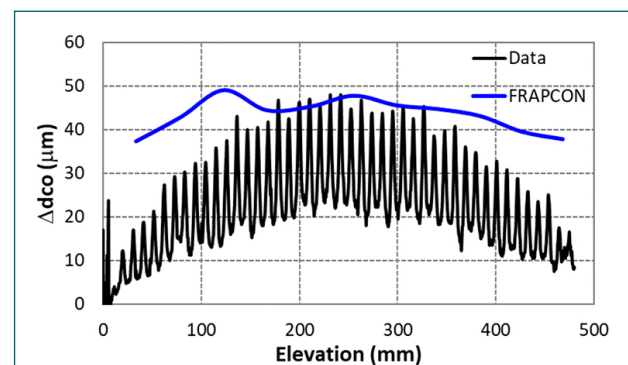


Figure 2. Rod Aa2 cladding outer diameter increase, Δd_{co} , as a function of the fuel rod axial elevation.

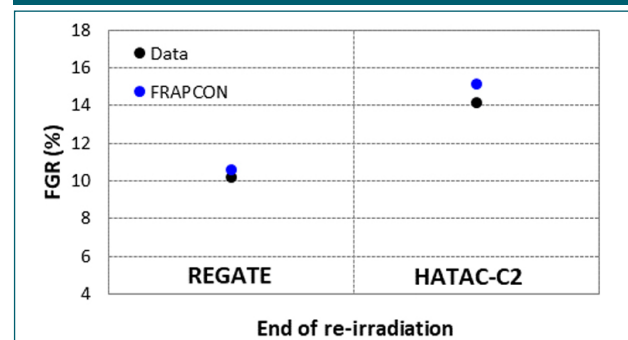


Figure 3. FGR for tests simulated.

This work is framed under the IAEA ATF-TS Coordinated Research Project.

Transient FGR

Simulation of the REGATE and HATAC-C2 fuel rods (both around 47 GWd/tU during base irradiation, followed by re-irradiations where multiple power ramps were imposed) from the IFPE database [17], led to FRAPCON's deviations of less than 20% in relative terms (Figure 3). The MASSIH's model for FGR, which does not distinctly account for transient FGR, was used. Further benchmarking is to be necessary to broadly assess FRAPCON performance under ramp conditions.

This work is being done under the frame of the Expert Group of Reactor Fuel Performance of the OECD/NEA.

LOCA

Under LOCA, cladding might balloon due to high temperatures at local spots, which would affect its coolability and, most importantly, could threaten its integrity (cladding burst). FRAPTRAN deviations in these scenarios had been already reported [11], but no specific study on the impact of key uncertainties, mostly embedded in the creep model and failure criteria, had been addressed. By simulating the Halden IFA-650.10 experiment (LOCA test on fuel irradiated up to 61 GWd/tU) [18], it has been proven that high-temperature creep law plays a fundamental role in time-to-failure FRAPTRAN inaccuracies; contrarily, failure limits uncertainties seem not to have a significant impact. These observations are displayed in Figure 4, where the fast pressure drop indicates the time-to-failure.

This work is framed under the R2CA project of the EURATOM HORIZON-2020 Program.

Modelling extension/development

The continuous evolution of the nuclear fuel context unavoidably gives rise to the need for modelling "stretching" beyond the fuel performance codes design. Two activities are worth emphasizing: the FRAPCON extension to dry storage, called FRAPCON-xt; and the FRAPCON/FRAPTRAN extension to evolutionary ATFs. Furthermore, an in-house model for in-clad hydrogen distribution (called HYDCLAD) has been developed to be coupled with FRAPCON/FRAPTRAN.

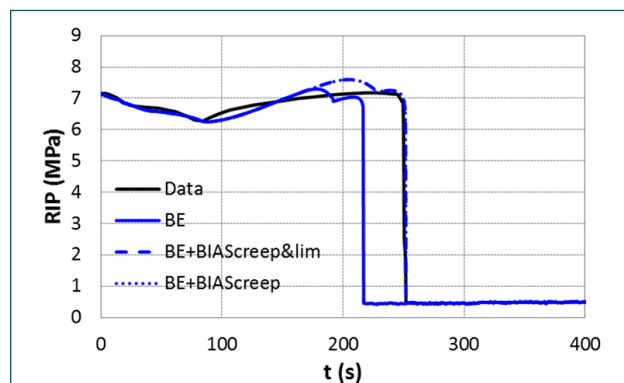


Figure 4. RIP vs. time in IFA-650.10. Code's predictions: best estimate, BE, best estimate plus bias of both creep and failure models, BE+BIAScreep&lim, and best estimate plus bias of creep model, BE+BIAScreep.

FRAPCON-xt

FRAPCON-xt was born with the intention to provide a sound thermo-mechanical characterization of spent fuel rods during the predisposal stage [6], as this is an essential input for later handling/transport operations and potential unexpected events.

The most recent work has been related to the initial thermo-mechanical characterization of high burnup spent fuel previous to dry storage (i.e., in-reactor EOL). This responds to the deviations reported above for RIP at burnup higher than 60 GWd/tU. The alternative FRAPFGR model, which offers a more factorized formulation of FGR, has been recalibrated through the intergranular resolution term in the fission gas diffusion equation. Figure 5 shows an enhancement of the RIP prediction accuracy with respect to the modelling by default (relative error reduction from 33% to 12% in the highest burnup fuel rod). Further validation, though, still needs to be done to confirm the trend observed.

This work is framed in the EJP-EURAD and OCATS projects.

FRAPCON/FRAPTRAN extension to evolutionary ATFs

Safety assessment of evolutionary advanced technology fuels (ATFs) under normal and off-normal conditions calls for the extension of fuel performance codes to the modelling of novel materials. CIEMAT is contributing to this field by identifying and addressing key requirements for modelling and simulation. Based on that, FRAPCON and FRAPTRAN have been extended to FeCrAl cladding and chromia-doped pellets through the implementation of material models and correlations.

The FGR and the PCMI behavior of chromia-doped UO_2 fuel under normal operating and power ramp conditions have been assessed against experimental data from Halden (IFA-716 and IFA-677 tests) and Studsvik (SCIP-II Aa2 test). The outcome of the simulations provided valuable insights that contributed to improving the understanding of these phenomena.

A comparative analysis of FeCrAl and Zircaloy-4 claddings has been conducted under steady-state conditions [19] and LOCA conditions (IFA-650.9 and IFA-650.10 tests) for rods burned up over 60 GWd/tU. Major thermo-mechanical differences have been noted between both materials and the

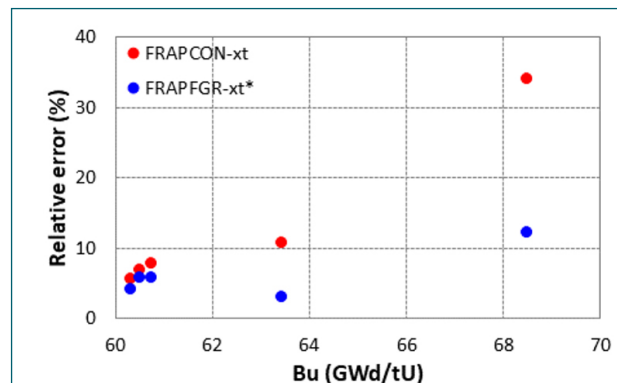


Figure 5. RIP relative errors (absolute value) above 60 GWd/tU. Predictions with FRAPCON-xt (before calibration) and FRAPCON-xt* (after calibration).

key properties responsible for it have been identified. However, the lack of experimental data limits the ability to validate the FRAPCON/FRAPTRAN extensions to FeCrAl cladding. Figure 6 illustrates this comparison under LOCA conditions; the differences observed between the two cladding materials are consistent with the superior mechanical strength of FeCrAl.

This work is being carried out under the frame of the IAEA ATF-TS Coordinated Research Project.

HYDCLAD

Hydrogen distribution and precipitation in the cladding is a key aspect for cladding integrity, as it is directly related to cladding embrittlement (i.e., ductility reduction due to hydride rim at the cladding outer side in high burnup fuels and potential radial reorientation in dry storage).

HYDCLAD is a model developed to simulate hydrogen migration and precipitation [8], as well as the radial hydrides formed under dry storage conditions [10]. It is coupled with FRAPCON and FRAPCON-xt, from which it gets the boundary conditions needed for the simulation of the irradiation period (cladding temperature, hydrogen absorbed, and oxide thickness) and the interim dry storage (cladding temperature and hoop stress).

HYDCLAD has been recently upgraded with a new and more phenomenological approach to precipitation modelling, which considers nucleation and aggregation to previous hydrides as precipitation mechanisms [21]. The upgraded HYDCLAD has been assessed under two irradiation scenarios:

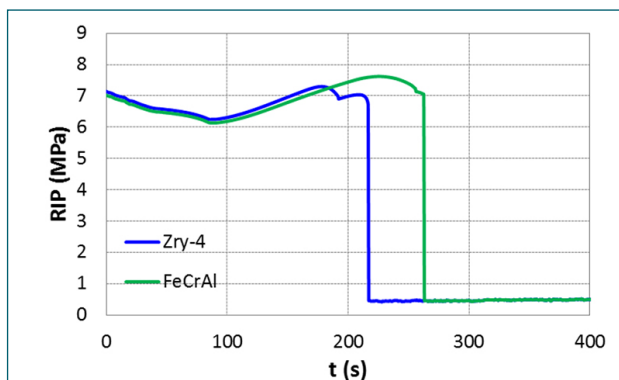


Figure 6. Estimated RIP of UO_2 -FeCrAl vs. UO_2 -Zircaloy fuel rods under the Halden LOCA test IFA-650.10 conditions.

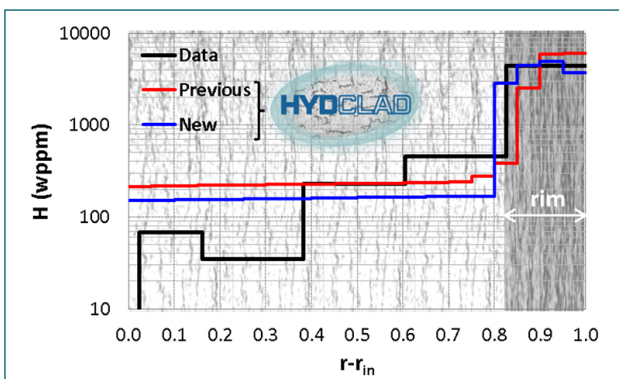


Figure 7. Distribution of the hydrogen content (in logarithmic scale) across the cladding thickness (normalization of the radius, r , minus the inner radius, r_{in}).

ios: hydriding in non-defective fuel rod (comparison against EOL PIE data from a 69 GWd/tU fuel rod) and secondary hydriding in defective fuel rods (hypothetical scenario with massive pickup in the cladding fuel side at the beginning of life). The former seems to indicate that the newest HYDCLAD version is more accurate in the prediction of the hydride rim (Figure 7). In the defective fuel rod scenario, the new precipitation modelling is shown to be capable of estimating the hydrides blisters expected under massive hydrogen uptake; however, the lack of open data prevents a true validation of this estimate.

This work has been developed under the frame of the R2CA project.

Methodologies

Beyond the progress in simulation capabilities of nuclear fuel, the research activities explained above are expected to be fed into methodologies for fuel rod integrity assessment in different conditions. Two main activities have been recently developed: an analytical toolkit for uncertainty and sensitivity analysis (UaSA) of predictions; and a protocol to characterize remaining cladding ductility under potential mechanical loads (oriented to dry stored spent fuel).

Uncertainty and sensitivity analysis

In the context of analytical tools like the fuel performance codes, the uncertainty analysis quantifies the code's precision (dispersion of the estimations) in predicting the figures of merit, FOMs, selected (e.g., peak cladding temperature, rod internal pressure, etc.), whereas the sensitivity analysis identifies the most relevant input uncertainties for these FOMs.

The objective of bringing UaSA methodology into fuel performance simulations is to assess the uncertainties associated with important variables in fuel performance characterization and to identify the most influential thermo-mechanical phenomena. The uncertainty propagation applied uses the statistical uncertainty analysis tool DAKOTA [22]. Recently, a runtime environment has been developed to couple FRAPCON and FRAPTRAN with DAKOTA for those cases in which the uncertainties associated with the input parameters must not only be propagated through the base irradiation (FRAPCON), but also the transient (FRAPTRAN). The program only requires the following inputs to perform the best estimate plus uncertainty calculation in a single run:

- FRAPCON input with the base irradiation specifications.
- FRAPTRAN input with the transient specifications.
- DAKOTA input with the uncertain input parameters and the associated probability density function.

Figure 8 shows a scheme of this methodology applied to LOCA test simulation with a rodlet refabricated from irradiated fuel.

Analysis of spent fuel cladding ductility

In the handling/transport of spent fuel after interim dry storage, the remaining ductility of the cladding must be able to withstand mechanical loads that may occur. In this context, the cladding characterization in terms of radial hydrides content (related to embrittlement) is of utmost interest to assess the integrity under the anticipated mechanical loads [23].

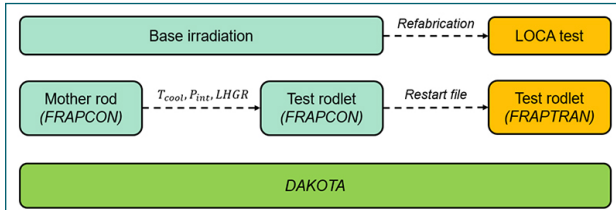


Figure 8. Scheme of UaSA for a LOCA test simulation.

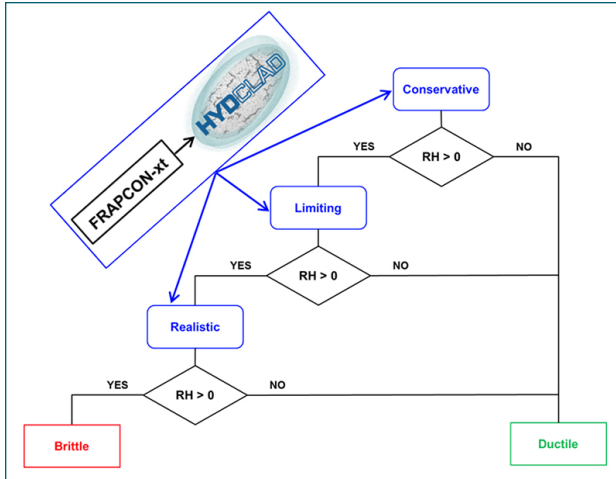


Figure 9. Scheme of the methodology for spent fuel cladding characterization (RH: Radial Hydrides).

A methodology for the characterization of the cladding ductility during dry storage has been developed [24]. It is based on the prediction of the radial reorientation of hydrides with HYDCLAD (fed with FRAPCON-xt). For this purpose, three levels of prediction are established depending on the thermal boundary conditions applied during storage:

- Conservative. Maximum cladding temperature of 400°C with isothermal axial profile.
- Limiting. Maximum cladding temperature of 400°C and axial temperature profile.
- Realistic. Maximum cladding temperature of 350°C and axial temperature profile.

Regarding the criterion applied for the ductility characterization, since there is no embrittlement criterion related to the radial hydrides content, a conservative criterion has been assumed where the cladding is considered embrittled if the radial hydrides content prediction is higher than zero.

Figure 9 shows a scheme of the methodology explained. Note that this methodology has been used so far taking into account the best estimate of the analytical tools, but it can be extended with the uncertainty and sensitivity analysis capabilities previously shown.

FUTURE OUTLOOKS

The strategy set years ago by CIEMAT to support even safer fuel rod designs and operations still stands. Nonetheless, a look ahead allows identifying several sources of innovation to be brought in:

- Foreseen changes in the own research domain, the fuel rod (design and operation) during its irradiation in commercial nuclear power plants and during the pre-disposal stage. With no intention to be comprehensive but just to

mention a few: new designs called advanced fuels (beyond ATFs), coming with new reactor systems expected to be deployed even within the decade; more challenging operating conditions searching for a better and still profitable fit of nuclear power to evolving electricity grids; and, no less important, search for optimizing back-end fuel management till its final disposal.

- New generation of analytical tools (their way of use included) and methodologies. The FRAP family evolution to FAST (the reference USNRC analytical tool for steady and transient irradiation of nuclear fuel) is already a fact, but there is still a path to make it as sound and robust as its predecessors have become. Additionally, and without abandoning the mainstream of analytical modeling approach, other tools like TRANSURANUS, SCIENTIX, or even multi-scale multi-dimensional codes (like BISON or OFFBEAT) are to be considered as potential sources of insights into fuel rod behavior that might be embedded in the analytical capabilities used.
- Expansion of the safety domains to be investigated. Recently, an extension of the FRAP family to address DEC-A accident scenarios (i.e., core degradation with no significant fuel melting) has been attempted to achieve a more mechanistic assessment of potential radiological threats to the environment. There is no reason to exclude any further extension, which will come along with in-house model developments that will be later fed into the analytical tools being used.

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REFERENCES

- [1]. Geelhood, K.J., Luscher, W.G., Raynaud, P.A., Porter, I.E. FRAPCON-4.0: A Computer Code for the Calculation of Steady-State, Thermal-Mechanical Behavior of Oxide Fuel Rods for High Burnup. PNNL-19418, Vol.1 Rev.2 (2015).
- [2]. Geelhood, K.J., Luscher, W.G., Cuta, J.M., Porter, I.A. FRAPTRAN-2.0: A Computer Code for the Transient Analysis of Oxide Fuel Rods. PNNL-19400, Vol.1 Rev2 (2016).
- [3]. Herranz, L.E., Tijeras, A. Solid-solid conductance in nuclear fuel: An assessment of it in-code models performance. Progress in Nuclear Energy, 52, 435-441 (2010).
- [4]. Herranz, L.E., Vallejo, I., Khvostov, G., Sercombe, J., Zhou, G. Assessment of fuel rod performance codes under ramp scenarios investigated within the SCIP project. Nucl. Eng. Des. 241, 815-825 (2011).
- [5]. Sagrado, I.C., Herranz, L.E. Modeling RIA benchmark cases with FRAPTRAN and SCANIR: A comparative exercise. Nuclear Engineering and Design, 278, 150-162 (2014).
- [6]. Feriá, F., Herranz, L.E., Penalva, J. On the way to enabling FRAPCON-3 to model spent fuel under dry stor-

- age conditions: The thermal evolution. *Annals of Nuclear Energy*, 85, 995-1002 (2015).
- [7]. Fera, F., Herranz, L.E. Critical review of data and correlations describing key clad thermo-mechanical processes under SFR transient conditions: Alternative modelling. *Progress in Nuclear Energy*, 97, 90-98 (2017).
- [8]. Fera, F., Herranz, L.E. Effect of the oxidation front penetration on in-clad hydrogen migration. *Journal of Nuclear Materials*, 500, 349-360 (2018).
- [9]. Fera, F., Herranz, L.E. Evaluation of FRAPCON-4.0's uncertainties predicting PCMI during power ramps. *Annals of Nuclear Energy*, 130, 411-417 (2019).
- [10]. Fera, F., Aguado, C., Herranz, L.E. Extension of FRAPCON-xt to hydride radial reorientation in dry storage. *Annals of Nuclear Energy*, 145, 107559 (2020).
- [11]. IAEA. Fuel Modelling in Accident Conditions (FUMAC). IAEA-TECDOC-1889 (2019).
- [12]. NEA. Nuclear fuel safety criteria technical review. ISBN 978-92-64-99178-1 (2012).
- [13]. Geelhood, K.J., Luscher, W.G., 2015. FRAPCON-4.0: Integral Assessment. PNNL-19418, Vol.2 Rev.2.
- [14]. Aguado, C., Fera, F., Herranz, L. E. FRAPCON-xt internal pressures validation for the characterization of spent fuel behaviour at dry storage. Reunión Virtual de la Sociedad Nuclear Española (2020).
- [15]. NEA. https://www.oecd-neo.org/jcms/pl_25445/studies-vik-cladding-integrity-project-scip#toc_1_2
- [16]. NNL. Provision of Information on Chromium Doped Fuel for Use in Light Water Reactors (2020).
- [17]. NEA. https://www.oecd-neo.org/jcms/pl_36360/international-fuel-performance-experiments-ifpe-contents
- [18]. Lavoil, A. LOCA testing at Halden, the tenth experiment IFA-650.10. HWR-974, OECD Halden Reactor Project (2010).
- [19]. Aragón, P., Fera, F., Herranz, L.E. Modelling FeCrAl cladding thermo-mechanical performance. Part I: Steady-state conditions. *Progress in Nuclear Energy*, 153, 104417 (2022).
- [20]. Fera, F., Herranz, L.E. Assessment of hydride precipitation modelling across fuel cladding: Hydriding in non-defective and defective fuel rods. *Annals of Nuclear Energy*, 188, 109810 (2023).
- [21]. Passelaigue, F., Lacroix, E., Pastore, G., Motta, A.T. Implementation and Validation of the Hydride Nucleation-Growth-Dissolution (HNGD) model in BISON. *Journal of Nuclear Materials*, 544, 152683 (2021).
- [22]. Adams, B.M. et al. Dakota, A Multilevel Parallel Object-Oriented Framework for Design Optimization, Parameter Estimation, Uncertainty Quantification, and Sensitivity Analysis: Version 6.16 Theory Manual (2022).
- [23]. Billone, M., Burtseva, T.A., Einziger, R.E. Ductile-to-brittle transition temperature for high burnup cladding alloys exposed to simulated drying-storage conditions. *Journal of Nuclear Materials*, 433, 431-448 (2013).
- [24]. Fera, F., Herranz, L.E. Análisis de integridad de barra de CN Almaraz en reinundación tras almacenamiento en seco en ENUN32P mediante estimación de reorientación de hidruros. DFN/SN-01/OP-23 (2023).■