A SAFETY FUEL ELEMENT ASSESSMENT BY USING NEW MATERIALS AND ADVANCED MODELLING TOOLS

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Abstract

After the accident occurred at Fukushima Daiichi nuclear power plant (NPP), a widespread awareness emerged in the nuclear field community that revealed the urgent need to develop fuel designs much more resistant to extreme accident conditions and capable to limit the magnitude of its harmful consequences. In order to fulfil these requirements, a worldwide initiative is underway, aimed to develop and implement the needed Accident Tolerant Fuel (ATF) concepts. The BaCo code was developed to simulate the nuclear fuel rods behaviour under irradiation and dry storage conditions including statistical analysis and 3D extensions. Research on new fuels and cladding materials properties based on \textit{ab initio} and Multiscale Modelling of Materials (M³) are developed to be included in the BaCo modelling. This work studies the whole life of fuel elements, during and after irradiation, and the benefits of implementing the M³ calculations in the behavioural code. The evaluation of an ATF with a silicon carbide (SiC) cladding is the considered example with a focus on the analysis on the fuel safety.

1. INTRODUCTION

As a consequence of the accident produced in the Fukushima Daiichi nuclear power plant, leading to huge radioactive release in 2011; it was considered essential to implement mayor improvements in nuclear safety not only during operational conditions but also in case of a less frequent accident occurrence. In order to do that, the Accident Tolerant Fuels (ATF) initiative arose as an answer to develop more resistant fuel elements to support the first steps of a severe accident.

Simulation and modelling of the fuel behaviour at different operational conditions are essential in order to improve fuel designs and nuclear security. They allow evaluating critical conditions without expending too much time and money in the performance of expensive experiments. First principles methodology, based on density functional theory is implemented through the QuantumEspresso code, as a theoretical complement to support these calculations. It is also meant to determine the predictive ability of this code to adequately describe the studied material’s mechanical and thermal properties.

This paper aims to show how a computational tool as the BaCo code could help improve the everyday management of a nuclear power plant (NPP).

2. THE BACO CODE

The BaCo code ("BArra COmbustible" -Spanish expression of "Fuel Rod"-) was developed at CNEA ("Comisión Nacional de Energía Atómica" – Atomic Energy National Commission of Argentina) to simulate...
nuclear fuel rods behaviour under irradiation conditions [1, 2] and at dry storage conditions [3, 4]. BaCo focus on PHWR fuels as the CANDU [4, 5] and Atucha [6] ones but we keep a full compatibility with PWR, BWR, WWER and PHWR MOX, plus advanced, experimental, prototypes and/or non-usual fuels. At present CNEA are developing the CAREM reactor where the fuel element has a hexagonal array as the WWER fuels. BaCo was strongly involved in the original design and test of that innovative Argentinean PWR fuel and advanced PHWR fuels (as PHWR MOX and the CARA ones).

BaCo assumes azimuthal bi-dimensional symmetry in cylindrical coordinates for the FR (“Fuel Rod”) [1]. Although angular coordinates are not considered explicitly, angular dependent phenomenon, as well as radial cracking, are simulated through the angular averaging method [7]. Also axial pellet cracking and relocation are included in BaCo. The hypotheses of axial symmetry and modified plane strains (constant axial strain) are used in the numerical modelling. The fuel rod is separated in axial sections in order to simulate its axial power profile dependence. Rod performance is numerically simulated using finite time steps (finite differential scheme). The modular structure of the code easily allows the description of phenomena observed in the UO₂ pellet and the Zry cladding behaviour. The current version of BaCo can be applied to any geometrical dimensions of cylindrical fuel rods mainly with UO₂ pellets (either compact or hollow, with or without dishing) and a Zry cladding.

In addition, it is suitable for the inclusion of advanced structural and fuel materials of the fuel rod, at least for the present version of the code, with an illustrative and comparative purpose. At present, the modelling of materials as metallic uranium, uranium carbide, uranium nitride (for pellets) and silicium carbide, FeCrAl (for claddings) are under development [8, 9].

The details of the mechanical and thermal treatment and the pellet, cladding and constitutive equations are available in reference (1) and an extended description of the code is included in reference [2].

The BaCo code was a participant of a series of Coordinated Research Projects (“CRP”) of the International Atomic Energy Agency (“IAEA”) generically named FUMEX (“Fuel Modelling at Extended Burnup”). The D-COM and the CRP FUMEX I, II & III were a series of comparison among experimental irradiations and code calculations. At present, we are involved in the IAEA’s CRP ACTOF (CRP on “Analysis of Options and Experimental Examination of Fuels for Water-Cooled Reactors with Increased Accident Tolerance”).

The code includes additional tools as the software package for finite elements 3D calculations and the statistical analysis for advanced fuel designs by taking into account the as fabricated fuel rod parameters and their statistical uncertainties. BaCo allows the calculation of a complete set of irradiations as for example the calculation of a full reactor core. BaCo 3D tools [10], statistical analysis [11], full core calculations [6] and graphical data post-processing improve the code performance and the analysis of the calculations [2].

Although the BaCo code uses a quasi-two-dimensional approach, the use of several three dimensional (3D) finite element features allow a complementary analysis of 3D properties, as for example the stress-strain state at a specific period of time during the irradiation [10]. The BaCo code results were enhanced by using “ad hoc” tools developed at the MeCom and SiM³ Divisions (Bariloche Atomic Centre, CNEA) [12].

For a better understanding of the uncertainties and their consequences, the mechanistic approach must, therefore be enhanced by the statistical analysis [11]. BaCo includes a probability analysis within their code structure covering uncertainties in fuel rod parameters, in the code parameters and/or into the fuel modelling taking into account their statistical distribution. As consequence, the influence of some typical fabrication parameters on the fuel cycles performance can be analyzed. It can also be applied in safety analyses and economics evaluation to define the operation conditions and to assess further developments. These tools are particularly valuable for the design of nuclear fuel elements since BaCo allows the calculation of a complete set of irradiations.

3. FIRST PRINCIPLES METHODOLOGY

For non-traditional fuels, in particular ATF and Generation-IV fuels, available data for the development of new materials can be obtained through the Multi-Scale Model of Materials (or M³), a methodology that provide the theoretical approximation to the modelling of the properties of the Materials through ab initio methods, molecular dynamics, Monte Carlo Kinetic and finite element calculations.
Silicon carbide is presented as a large family of crystalline structures called "polytypic". This manifests the ability of this compound to crystallize in numerous modifications that can be described as different stacking sequences of the same unit layer. Most of the polytypic are modifications of the alpha phase (hexagonal structure). Nevertheless, for reasons of nuclear application, we were interested in starting with the study of the $\beta$ phase (cubic structure) that forms below 1700°C.

3.1. Methodology applied to $\beta$-SiC

3.1.1. Elastic Constants

The silicon carbide beta phase ($\beta$-SiC) corresponding to the 3C polytypic has the cubic zinc blende crystal structure (F-43m space group).

The elastic constants determine the elasticity and mechanical stability of the crystals. For small deformations, within the elastic range, one expects a quadratic dependence of the energy of the crystal $E$ with the deformation (Hooke's Law).

Table 1 compares the results of the constants and elastic modulus obtained in our work in comparison with those previously published by other authors.

<table>
<thead>
<tr>
<th></th>
<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$C_{44}$</th>
<th>$B$</th>
<th>$Y$</th>
<th>$G$</th>
<th>$\nu$</th>
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<tbody>
<tr>
<td>Present work</td>
<td>376.35</td>
<td>121.38</td>
<td>257.65</td>
<td>206.37</td>
<td>317.15</td>
<td>194.26</td>
<td>0.142</td>
</tr>
<tr>
<td>Ref.[13]</td>
<td>390.1</td>
<td>142.7</td>
<td>191.0</td>
<td>225.1</td>
<td>313.6</td>
<td>123.7</td>
<td>0.268</td>
</tr>
<tr>
<td>Ref.[14] (CAST EP)</td>
<td>420</td>
<td>132</td>
<td>267</td>
<td>228</td>
<td></td>
<td>208</td>
<td></td>
</tr>
<tr>
<td>Ref.[15] (FP-LMTO)</td>
<td>420</td>
<td>126</td>
<td>287</td>
<td>223</td>
<td></td>
<td></td>
<td>0.231</td>
</tr>
<tr>
<td>(Exp.) Ref.[16]</td>
<td>390</td>
<td>142</td>
<td>256</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Exp.) Ref.[17]</td>
<td>379</td>
<td>141</td>
<td>252</td>
<td></td>
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3.2. Thermal properties

3.2.2. Phonon density of states

The phonon dispersion curves have no imaginary frequencies, showing its dynamic stability, in agreement with the results obtained by the analysis of the elastic constants. In Fig 1, it can be observed that at low frequencies (below 20THz) the partial density of phononic states is dominated by the Si atoms as the acoustic modes originate mainly from the heavier elements, and above 20THz the phononic modes originate mainly by C atoms. It can also be noticed the presence of a phonon gap around 18-22 THz which is in good agreement with previous results [18].
3.2.3. **Specific heat**

For many purposes, an efficient method to calculate the temperature dependent properties of an anharmonic crystal is the quasi-harmonic approximation (QHA). Within this approach, the anharmonic lower order corrections are considered, allowing the interatomic force constants and phonon frequencies to depend on the volume. In order to calculate the specific heat at constant volume, only the harmonic frequencies are needed, which were determined for the lattice in equilibrium.

![Figure 2: Entropy (SV), Helmholtz free energy (FV) and specific heat (CV) as a function of temperature.](image2)

![Figure 3: Specific heat calculated as a function of temperature. The red line corresponds to the dimensionless quantity CV/3Nk_B while the blue line CP/3Nk_B, where N is the number of Avogadro and k_B is the Boltzmann constant. The horizontal line indicates the Dulong-Petit limit.](image3)

Fig. 2 and 3 show the calculated thermal properties CV, SV, and FV, using the harmonic approximation, while in order to calculate CP the quasi-harmonic approximation (QHA) was used. The results obtained for the vibrational entropy and specific heat at constant pressure are in good agreement with the work Refs. [18], [19], [20] and [13], but the work of Lee et al. [19] who used the VASP code, and Thakore et al. [20] who used the Quantum Espresso code, have a difference between the specific heats that appears to be a little more pronounced. Since the solids are assumed practically incompressible, values for CP and CV are almost equal and therefore a single value for the specific heat is used. However, with increasing temperature, their difference is no longer small.

3.2.4. **Linear thermal expansion coefficient**

The linear thermal expansion coefficient, denoted \( \alpha_L \), is obtained theoretically as:

\[
\alpha_L = \left( \frac{\partial a}{\partial T} \right)_P \tag{1}
\]

where \( T \) is the temperature and \( "a" \) is the lattice parameter. Therefore, a very good approximation is given by:

\[
\alpha_L \approx \frac{1}{a_0} \left( \frac{\partial a}{\partial T} \right)_P \tag{2}
\]

where \( \alpha_L \) is the lattice parameter and \( a_0 \) is the lattice parameter at the reference temperature \( T_0 = 300K \).

![Figure 4: Temperature dependence of \( \beta \)-SiC phase linear thermal expansion coefficient vs. temperature.](image4)
4. BASIC DEVELOPMENT OF ATF’S (ACCIDENT TOLERANT FUELS)

Currently, the nuclear industry has begun to require the development of new fuels that are resistant to severe accidents and do not imply a reduction in their performance. New materials and new designs of fuel elements converge in the ATF initiative. We analyse the physical properties of the proposed materials for fuels and claddings. In addition, the parameters of these materials are obtained by "ab initio" and M³ methods, in particular for the SiC, which are incorporated into the BaCo and BaCo3D codes for a first approximation of the ATF simulation under irradiation.

4.3. Performance Analysis of an ATF

The relevant calculations were performed with a version of BaCo which includes the properties of SiC calculated by ab initio methods in the previous section in order to obtain a new approximation about its use as cladding material (i.e. a type SiC/UO₂ fuel rod) and continue the comparison of the behaviour relative to Zry-4/UO₂ and FeCrAl/UO₂. The results obtained in this new version with the support of the basic physics associated with the modelling, are equivalent with the parameters from bibliography.

As an example of these calculations, in FIG. 5, is represented the central temperature of the fuel pellet of UO₂ as a function of the burnup, where is reached the maximum temperature recorded in the FE (“Fuel Element”) at a steady irradiation at 250 W/cm. The reduction in temperature for the case of Zry-4 from ~3 MWd/kgU is due to the PCI (“Pellet-Cladding Interaction”) which improves the thermal conductivity and the transfer of heat from the pellet to the coolant. In FIG. 6, we see the evolution of the pellet and clad radii using a SiC cladding, obtaining PCI to EOL (“End Of Life”).

A more realistic example corresponds to the "hoop stress" in FIG. 7, where the irradiation corresponds to a realistic simplification (generic, conservative PWR power history, already very high burnup) of a commercial irradiation that was provided by FANP ("Framatome Advanced Nuclear Power, Inc.”) for its study (Case 27.2d).
within the FUMEX II (Coordinated Research Project on Fuel Modelling at Extended Burnup II) of the IAEA, where BaCo participated [21, 22]. The modelling starts at an average power of 350 W/cm followed by 4 power cycles at a decreasing level. The irradiation time is from ~1700 days to an extraction burn of ~70 MWd/kgU. FIG. 8 shows the curves of the internal pressure of the free gases in the sheath (filling gases - He and gaseous fission products, Xe and Kr). Here we have coupled all the aspects of calculation with the code BaCo, i.e., mainly, the thermal and mechanical aspects because the temperature and evaluation of the free space inside the sheath are relevant. The highest-pressure value is obtained with the use of Zry due to the PCI that eliminates the free spaces between the pellet and the cladding mainly due to the creep down of the cladding and thermal expansion of the fuel pellet.

To illustrate, we repeat the simulation calculation of a fuel under dry storage conditions by using a WWER 440 fuel rod type used as a control case in the FUMEX II CRP (Case 9), irradiation in the Kola 3 NPP, BC 007 of the FA 222) [22] and assuming a SiC cladding. FIG. 9 shows the same agreement with the previous calculations and the same general observations. It is evident that, if using a SiC cladding, it will be necessary to ensure that its gap remains open, to avoid PCI. In order to prevent a failure of a solid tube of SiC we use a cladding type SiC/SiC (inner solid tube covered with fibres). FIG. 10 shows the evolution of the gap in which PCI is reached to EOL and we find the gap opened when passing to storage conditions.

The FIG. 11 shows the gas pressure inside the fuel rod during the irradiation. In order to enhance the previous analysis of the behaviour under dry storage conditions it is included in the FIG. 12 the pressure outside the fuel rod. That pressure is the coolant one during the irradiation at the nuclear reactor. The fuel rod after irradiation is stored at normal pressure (1 atm) and a temperature of 200°C is estimated. The gas pressure is lower than the pressure of the coolant during all the time of the irradiation as it is required for safety reason.

The calculations plotted on Figures 5 to 11 were performed by using the data input corresponding to the nominal or as fabricated parameters of the fuel rod and the reactor. The as fabricated tolerances of the fuel parameters usually are not included in the code calculations. BaCo allows the calculation including the statistical dispersion produced during the fuel fabrication that is compatible with the tolerances defined for the fuel designers among other parameters. Several running codes are automatically programmed into BaCo in order to perform a statistical evaluation of the fuel behaviour. Each running code is using a random input data set. Each parameter of each individual data set is estimated by taking into account the as fabricated tolerance and its statistical dispersion. Then each individual data set is representing a possible and real fuel rod.

FIG. 12 is including the previous nominal calculation and the statistical evaluation. The set of points plotted in FIG. 12 and its density shows the dispersion of the calculation. A smooth increment on time of the gas pressure was found including the stage at dry storage due to the diffusion and release of the fission gases retained in the fuel pellets. The main difference is that the external pressure (the coolant pressure) during irradiation is compressive and a tensile state is done at dry storage conditions. The tensile stress on the SiC cladding could produce an unexpected crack or failure during storage.

FIG. 13 shows the hoop stress of the cladding (tangential stress at the inner surface of the cladding). A compressive and conservative tensile stress is found during irradiation and stress reversal is done at EOL. The positive value of stresses at dry storage shows a non-desirable condition.
FIG. 14 is the statistical evaluation with BaCo of the pellet centre temperature. The big dispersion at high burnup up to EOL is done due to the PCI or non PCI event during that step time.

The calculations were using the most demanding conditions and it is expected a more conservative result during an experiment environment.

5. FINITE ELEMENT METHOD (FEM) EXTENSIONS

5.4. BaCo (~2D) and BaCo3D (FEM)

5.4.5. Optimization of a WWER fuel pellet with a central hole

One of the characteristics of the WWER fuel pellets is the presence, by design, of a central hole. This hole produces a reduction of tensions, which induces a reduction of the radial deformations and, finally, a reduction of the stresses between the pellet and the cladding. Finally, a conservative design result is obtained. On the other hand, this design causes a decrease in the inventory of fissile material which can approximately be compensated for by a somewhat larger pellet radius or possibly with a slight increase in enrichment (if this is possible and, the extra technical problems that this would entail).

FIG. 15. includes the comparison of the radial deformation axial profile of a fuel pellet. The reduction of this deformation is verified experimentally in FIG. 16., where it is shown how effectively the presence of that central hole causes a decrease in the radial deformations. It is interesting to remark that the data provided by the Halden Reactor Project used for the validation is not a specific experimental irradiation [23].

Currently, designers of fuel element of the WWER have the possibility of producing solid pellets, without a central hole and with a "dishing" [24]. This design change is still under discussion for the security reasons already discussed. Another parameter that is taken into account is the proposed diameter for the central
hole of this type of pellets (see FIG. 15.). FIG. 17. includes a set of maps representing the radial deformation of a WWER fuel pellet as illustrative purposes of the BaCo3D capabilities.

![FIG. 15. Radial deformation of irradiated WWER type pellets in equivalent situations by varying the diameter of their central hole (Ø = 1.2, 1.8 and 3.0 mm).](image1)

![FIG. 16. The power profile of an experimental FR of the Halden Reactor. At the top of the FR are the pellets pierced with thermocouples. (HRP-305/8, [23]).](image2)

![FIG. 17. Radial deformation of a WWER fuel rod with a central hollow in the pellet. Different graphical ways to represents them with an "ad hoc" software developed in junction with BaCo3D.](image3)

6. **NEUTRONIC CONSIDERATIONS ABOUT ATF DEVELOPMENT**

In agreement with previous results, all new materials proposed for the ATF initiative need to revaluate its neutronic behaviour. First studies performed at unit cell level, showed that the reactivity of the core is highly reduced when using FeCrAl alloy for example [25]. A more detailed analysis of this subject needs to be done to estimate reactivity coefficients and a complete core calculation to see which other parameters are affected.

7. **CONCLUSIONS**

Different capabilities of the BaCo code to perform a complete analysis of the fuel element were shown. The importance of these types of analyses relies on the feedback that an operation authority could have in case of different engineering problems that could appear during the life plant.

In accordance with the analysis, the methodology of M³ is essential to support behavioural codes to obtain proper values of the properties of new materials. The experimental data needed for fuel simulation and ATF designs can be partially supported by using M³. The results obtained for the SiC has enough accurate as to be included in the BaCo modelling. As a conclusion of the presented calculations of SiC it was shown the needs to prevent PCI and the formation of microcracks during irradiation which could grow and present fission products leakage during storage. In the same way, it was clearly shown how a fuel pellet designed with a central hole will reduce stresses and deformations in the pellet. Those results were validated by using data provided for
the Halden Reactor Project and it is interesting to mention that the experimental data were extracted from an experiment of irradiations not related with the main issue of our study.

Improvements on the tools that support BaCo code, as FEM method, statistical analysis and the inclusion of material parameters obtained with M³, could increase the information that it can be obtained and can help to visualise in a simpler manner the fuel rod and the fuel element as a hole during irradiation and dry storage. Once all these tools are tuned and benchmarked, new improvements can be done and new designs that can enhance a safe operation of the NPP.

8. ACKNOWLEDGEMENTS

To the IAEA that give us the opportunity to present our work, Halden Reactor Project that kindly provided the valuable data used in this paper and the MeCom Department of CNEA that provided the basic engine for the FEM calculations included in BaCo3D.

REFERENCES


