Modelling of Materials under Irradiation in Inertial Fusion Reactors: Damage, Tritium and Activation

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Abstract. Neutron intensities and energy spectra in structural support materials versus time after target emission are presented for two IFE protections (LiPb, Flibe); these data are strongly required for evaluation of pulse effect. A multiscale modelling (MM) study of pulse irradiation in metals (Fe) has been extended up to the microscopic scale; we explain physics effects and remark differences with continuous irradiation. Static and dynamic validation of a new tight-binding molecular dynamics code to accurately determine defects energetic in SiC is presented. The effect of HT is remarked together with that of HTO when studying tritium releases; it is shown that HT contributes 90-98% to the total dose from ingestion of natural agriculture and meat and the rest comes from inhalation by the re-emission to the atmosphere. A Monte Carlo procedure estimates the effect of activation cross section uncertainties in the accuracy of inventory calculations and final materials consequences, which is based on simultaneous random sampling of all the cross sections and it is implemented in the activation code ACAB. The procedure is applied in collaboration with LLNL to the analysis at the National Ignition Facility gunite chamber shielding under a reference pulsing operation. Preliminary results show that the 95 percentile of the distribution of the relative error of the contact dose rate can take values up to 1.2. Model is also promising when applied to the uncertainty analysis of activation in IFE power plants, by using a continuous-pulsed model to represent the IFE real pulsed irradiation.

1. Time-dependent Neutron Intensities in IFE Structural Walls and Multiscale Modelling of Radiation Damage.

A full set of calculations have been performed [1,2], using 3D MonteCarlo, to determine neutron intensities as a function of time after the target emission in Fe samples representing the structural material in IFE environments. Energy spectra have also been generated for each layer and time interval. Spherical and cylindrical simulations have been performed considering a central target emission surrounded by vacuum (4 m), and protections of LiPb and Flibe of 60 and 100 cm thickness. The Fe layer is behind the protection with a total thickness of 10 cm. A first part of the study gives the intensity and energy spectra of neutrons emitted from the compressed and burning DT target using different assumptions from uniform to distributed compression and the source is $2x10^{20}$ n/pulse. Figure 1 represents the total intensity versus time for the inner and the outer Fe layer (10 cm thickness) and the two types of protection. We remark the different behaviour depending on the protection. In the case of Flibe the maximum is attained in the inner layer at the time interval 0.09-0.10 μs, while that maximum arrives at 0.10-0.11 μs in the outer layer; and the intensity decreases very rapidly in both layers. On the contrary, in the LiPb protection the intensity get its maximum at 0.2 μs, similar in both studied layers, with a new maximum at 1 μs much more pronounced in the inner layer. The interpretation of these results comes from the very different behaviour of the multiplicative nuclear reactions in the components of the liquid protections; in particular, (n,2n) and (n,3n) are very significant in the case of Pb, increasing the intensity but with neutrons of lower energy (a few MeV from those reactions).

Along the last years, Multiscale Modelling simulation of the pulse radiation damage in Fe has been conducted at DENIM [3]. The goal has been to determine the microscopic structure of irradiated Fe with time-dependent neutron intensity in terms of the concentration of defects (and type and size) with increasing accumulated doses. The simulation procedure has been described elsewhere in the context of lower fluence simulations which results have been extended recently[2]. We consider frequencies of 1 – 10 Hz and dose rates of 0.01 and 0.1 dpa/s. We obtain cluster accumulation in those rate conditions by injecting 10^4 and 10^5 pulses of 150 keV Fe ions in a 300 nm cubical box. Upgrading previous calculations the final dose, so far reached, is 10^{-3} dpa, being the time deposition of the pulse 1 μs and the dose accumulated per pulse in the two cases respectively of 10^{-7} and 10^{-8} dpa. A clear effect of the frequency is observed, with the same dose rate, indicating that the larger is the frequency the higher is the accumulation. This "macroscopic" result cannot be explained without consideration of the "microscopic" features of the species considered in the Kinetic MonteCarlo simulation, which certainly plays an important role in the evolution of the microstructure. The higher is the velocity of injection of defects in the simulation box the lower is the probability of migration for a vacancy or a di-vacancy, which are the mobile species in the simulation. We can conclude that more time elapses between pulses the more probable is for a di-vacancy (with lower migration energy) to migrate. The results of this migration will be the formation of bigger cluster, raising in this way the average cluster size and lowering the cluster concentration.

All these simulations will be clearly representative if a good agreement of experiments and calculations are previously demonstrated at the microscopic level. To obtain such goal there are some National and International Projects (REVE). In particular, a Project named VENUS-II is carried out in Spain under Nuclear Regulatory Commission and Electric Utilities sponsorship. VENUS-II implies the irradiation of pure and ultra-pure Fe samples using 150 keV Fe ions at 600 K. From those results a clear dependence on the diffusion parameters will clearly envisioned that needs more study and the final establishment of a reliable data base. An important work has been concentrated in the detailed calculation of such diffusion parameters and the physics understanding of cluster defects types and their inter-transformation [4].

Amorphization and cascade effects of irradiated SiC has been published [5]. A new step is being the development of a semiempirical Tight Binding molecular dynamics scheme (TBSiC) applied to Silicon Carbide (ß-SiC), which has been successfully validated with static and dynamics magnitudes and the electronic and crystalline structure [2, 3].

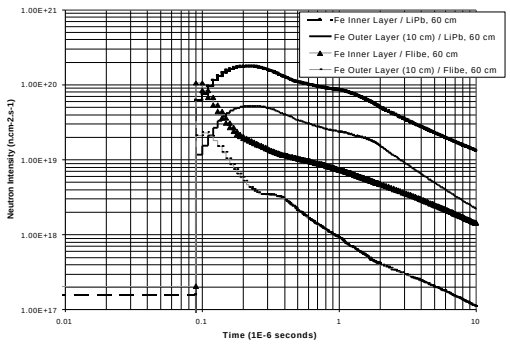


Fig. 1. Neutron Intensities vs. Time

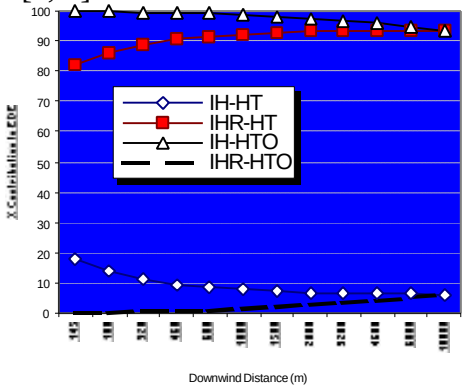


Fig. 2. Inhalation (IH) and Re-emission (IHR)

2. Elementary Tritium in Emissions of IFE Reactors

The behaviour of tritium in the biosphere and its radiological impact due to the accidental emission and normal operation may play an important role in the design of fusion reactors. We consider necessary to compare HT with HTO because both forms differ in their behaviour in the environment. The pathway of tritium after the liberation to the air have three differential and chronological phases that have been well described in previous works [6, 7, 8].

From our studies we remark the different behaviour of the two forms depending of ingestion and inhalation. A remarkable case is the internal dose by inhalation and skin absorption from the atmosphere during the re-emission phase (in the secondary phase), Figure 2. That process occurs when the elementary tritium deposited on the ground, rapidly converts into the HTO, 48-100 hours after the emission. For emissions of 100% HT the re-emission is higher than for emissions of 100% HTO. A fraction of HT in the leaves can be converted to HTO, but the most of oxidation (HT into HTO) is on the surface soil. The contribution to the total doses by re-emission is in the order of 80-90% in HT emissions, but only 1,5% in the HTO case. In normal conditions of operation the difference of the range between HT and HTO is smaller (13% and 3% respectively). The doses for inhalation of the primary plume can contribute 20-25% of total dose, and only the HTO has a significant contribution. For the first re-emission to the atmosphere, the tritium-gas has larger importance than the HTO. Therefore the tritium-gas has its larger contribution in the cases of ingestion and re-emission to the atmosphere of the tritium while the HTO is important through inhalation process and internal ingestion of tritium.

3. Monte Carlo Uncertainty Analyses Of Pulsed Activation In The Nif Gunite Shielding

The need to estimate the effect of activation cross section uncertainties in the accuracy of isotopic inventory calculations is an issue that is drawing more and more attention. Concerning this problem, the cross sections improvement is an important issue. Original cross sections uncertainty files have been successively improved and as a result of this work the uncertainty file FENDL UN/A-2.0, was generated [9]. To estimate the effect of such uncertainties in the isotopic inventory some calculational procedures have been proposed. In previously published work, we developed a method based on the first order Taylor [10, 11] series. One of the drawbacks of the method is that its application to pulsed scenarios is difficult. The other and most important limitation is that it is impractical to deal with the synergetic/global effect of the uncertainties of the complete set of cross sections.

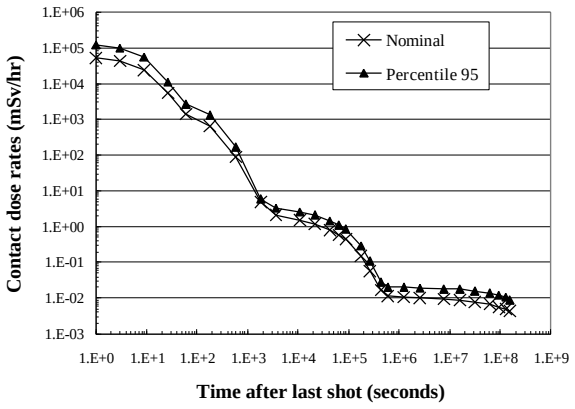
To overcome these limitations, we have developed a Monte Carlo procedure based on simultaneous random sampling of all the cross sections probability density functions (PDF) involved in a problem. The PDF for each cross section is assumed to be lognormal. This means that $\log(\sigma/\sigma_0)$ follows a normal distribution $N(0,\Delta)$ with σ_0 being the best-estimate cross section value contained in FENDL/A-2.0 and Δ being the square root of the variance included in FENDL UN/A-2.0. Using a set of collapsed cross sections values for all the reactions involved in the problem, the activation code ACAB2000 is used to obtain the corresponding nuclide inventory and inventory response functions. This process is repeated until a sample with the sufficient number of histories is obtained.

3.1 Contact Dose Rate for the NIF Gunite Shielding

We have applied the methodology presented above to the uncertainty analysis of the radionuclide inventory and contact dose rate at the National Ignition Facility (NIF) gunite chamber shielding. We have used the detailed material composition obtained from recent on-site measurements. Activation calculations assume thirty years of NIF operation. Each year consists of sixty shots each with 20 MJ of fusion yield; these shots are separated by approximately 6.1 days.

The neutron flux in the gunite is calculated by the TART Monte Carlo transport code. Activation calculations are performed with the ACAB2000 code by exactly representing the pulsed irradiation agenda. We obtain the probability distribution for the random variables contact dose rate (D) and relative error of the contact dose rate (E), where $E = (D - D_o)/D_o$ and D_o is the nominal contact dose rate obtained in the standard activation calculations, i.e., using the best estimate cross sections and not considering uncertainty data. Figure 3 shows the nominal contact dose rate and the 95 percentile of the contact dose rate distribution after the last shot. A result with more practical significance is the 95 percentile of the distribution of E (E_{95}), shown in Figure 2 (Base case). It is seen that for some times of interest the value of E_{95} can reach levels up to 100%.

A few radionuclides have been identified as important for the contact dose rate and for the contact dose rate uncertainty: ^{16}N (7 seconds half-life, total $\Delta^2 = 2.97\ 10^6$) shortly after shutdown, ^{24}Na (15 hour half-life, total $\Delta^2 = 1.73\ 10^1$) at times of relevance to maintenance activities and ^{60}Co (5.3 years half-life, total $\Delta^2 = 3.61\ 10^{-1}$) which dominates the decommissioning dose.



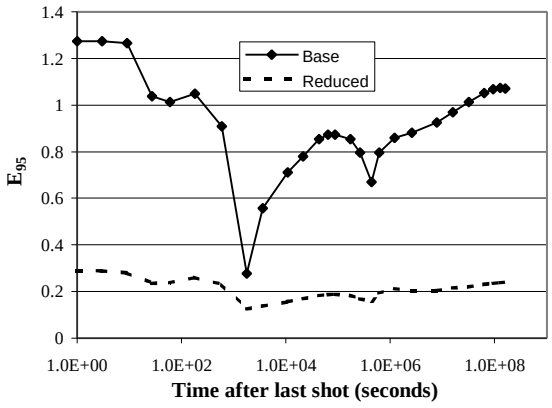


Fig.3. Contact dose rate and 95% confidence Interval. *Fig.4. Relative error of the contact dose rate.*

We have obtained that a significant reduction of the total contact dose rate uncertainty could be achieved by reducing the uncertainty in the concentration of only the critical radionuclides [12], which in turn depends only on the uncertainty in the cross sections for the three reactions: $^{16}O(n,p)^{16}N$, $^{27}Al(n,\alpha)^{24}Na$, $^{59}Co(n,\gamma)^{60}Co$. Assuming as a reasonable cross section improvement to reduce the variances to 1/10 of their current value, the problem has been reanalyzed. This approach allows us to evaluate the required reduction in the uncertainties of the cross sections in order to obtain a required uncertainty in the total contact dose rate. The new resultant values of E_{95} are shown in Figure 4 in dashed line (Reduced case).

The importance of these results may be found in the determination of the workers stay-out time before performing maintenance activities near the NIF chamber after 30 years of operation. If we consider that the maximum acceptable worker dose is 20 mSv/year (10 μSv/hr), and assuming that only the gunite shielding contributes to the worker dose, the contact dose rate would be below that limit after: 3 months based on best estimate calculations, 5 years if 95% confidence is desired, based on the uncertainty analysis with the original cross section variances, 1 year if 95% confidence level is desired, based on the uncertainty analysis with reduced variances for the critical cross sections.

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