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S. Maillard, G. Martin, C. Sabathier-Devals. Why a steady void size distribution in irradiated UO<sub>2</sub>? A modeling approach.. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2016, 374 (1), pp.58-66. 10.1016/j.nimb.2015.09.068 . cea-02509670

**HAL Id: cea-02509670**

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Submitted on 17 Mar 2020

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# Why a steady void size distribution in irradiated $\text{UO}_2$ ? A modeling approach.

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## Abstract

In  $\text{UO}_2$  pellets irradiated in reactor, Xe nano-bubbles nucleate, grow, coarsen and finally reach a quasi steady state size distribution (transmission electron microscope observations typically report a concentration around  $10^{-4} \text{ nm}^{-3}$  and a radius around 0.5 nm). This phenomenon is often considered as a consequence of radiation enhanced diffusion, precipitation of gas atoms and ballistic mixing. However, 4 MeV Au ion irradiation of  $\text{UO}_2$  thin foils at room temperature yields a nano-void population whose size distribution reaches a similar steady state, although quasi no foreign atoms are implanted nor significant cation vacancy diffusion expected at such temperature and ion energy. Atomistic simulations performed at low temperature support the assumption of heterogeneous nucleation: 25 keV sub-cascades produce defect aggregates that grow through sub-cascade overlapping. In this work a semi-empirical model is proposed to extend these results to the simulation of the size distribution evolution of a representative defect aggregates population in a fraction of a material grain under a cascade overlap regime. To account for the damage accumulation when cascades overlap, this model is based on simple rules inferred from the atomistic simulation results. It satisfactorily reproduces the TEM observations of nano-voids size and concentration, which paves the way for the introduction of a more realistic damage term in rate theory models.

*Keywords:* Fission gas bubbles,  $\text{UO}_2$ , Irradiation effects, Object Kinetic Monte Carlo, Irradiation cascade.

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## 1. Introduction

Fission gases produced in irradiated nuclear fuel materials such as  $\text{UO}_2$  may have detrimental effects on the fuel rod integrity due to excessive loading of the cladding, either when gas release increases the rod pressure, or when high temperature events enhance the pellet swelling. For this reason, extensive studies of fission gas behavior in  $\text{UO}_2$  under irradiation were developed over decades. They progressively achieved a clearer description of the phenomena, but a global understanding of the controlling mechanisms is still in progress. This paper is focused on the buildup of the cavity or bubble quasi steady state size distribution arising from the in pile irradiation of the external and colder zone of a fuel pellet, but also in other situations as will be seen below. The issue will be addressed through the modeling of the cavity growth process under ion irradiation as observed in a transmission electron microscope (TEM). This represents a first step in solving the more complex question of in-pile generated bubbles.

In fact, as reported for example in the review of [4], TEM observations of in-pile irradiated  $\text{UO}_2$  [26, 2] show bubbles appearing in bulk  $\text{UO}_2$  at fission densities around  $10^{-2} \text{ nm}^{-3}$ , rapidly coarsening and growing until their size distribution finally reaches a quasi steady state. The average radius  $\bar{R}$  and concentration  $\bar{C}$  lay in the ranges of  $\bar{R} \sim 0.5 \text{ nm}$ ,  $\bar{C} \sim 10^{-4} \text{ nm}^{-3}$  respectively. Further studies [7] exploring a wider range of temperature and burn-up show that the cavity size distribution evolves slowly with fission density or temperature while staying in the same range (for this reason we will generally omit the word “quasi” in front of “steady state”). To allow deeper investigation of the material, mock-up experiments were undertaken, such as in-situ thin foils irradiation with accelerated ions. For example in [15],  $\text{UO}_2$  foils implanted with 390 keV Xe ions at 600 °C also show a steady state cavity distribution moreover very similar to that of in-pile irradiated samples. Additionally this low energy ion irradiation experiment does not support the often claimed hypothesis of bubble “heterogeneous nucleation” in the wake of fission fragments, which reactivates the question of the bubble nucleation mechanism. Other investigations gave important information on this issue:

- Similar experiments with non gaseous ions (irradiation with 300 keV Cs ions at RT [21]) yielded the same cavity evolution. This suggests that the previously observed cavities may not contain gas and raises questions on the reality of the “homogeneous nucleation” alternative mechanism supposing bubbles nucleate when two gas atoms collide during the diffusion process. XANES analysis of samples similarly implanted with Xe ions proves the cavities are practically devoid of gas [1]. This conclusion was confirmed by irradiation with 4 MeV Au ions of  $\text{UO}_2$  foils too thin to allow significant Au implantation: the same cavity pattern has been observed. This clearly indicates that the origin of the cavity nucleation is to be first sought in the damage process and not in the implanted foreign atoms. Moreover, these results keep raising questions concerning the cavity growth mechanism: growth requires cation vacancies gathering but

45 in these conditions, no diffusion is expected (see discussion), so that the  
 46 gathering mechanism is questionable.

- 47 – Various classical molecular dynamics (CMD) simulations of cascades in  
 48  $\text{UO}_2$  shed some light on both the issues of cavity nucleation and growth  
 49 [11, 10, 12, 27, 28]. A global picture of the damage process can be drawn  
 50 [10, 4, 23]. An ion beam produces a displacement cascade that finally  
 51 splits into independent “sub-cascades” starting at an energy of  $\sim 25$  keV,  
 52 each producing a disordered and very hot zone of volume around a few  
 53 hundreds of  $\text{nm}^3$ . This zone finally anneals out in a damaged crystalline  
 54 zone comprising approximately one hundred Frenkel pairs (O and U iso-  
 55 lated or clustered vacancies and interstitial atoms); larger clusters such as  
 56 one cavity of 10 to 20 vacancies and one or two loops of 10-20 interstitial  
 57 atoms occupy the center and the periphery respectively of the previously  
 58 disordered zone. In addition, the central cavity volume grows in propor-  
 59 tion to the number of sub-cascade overlaps in the simulation box ([10] Fig.  
 60 7).

61 The global analysis (e.g. [4]) of these results suggests that the irradiation effect  
 62 results from successive independent sub-cascades in which cavities and loops  
 63 directly nucleate without requiring foreign atoms and that these defect clus-  
 64 ters can grow through sub-cascade overlaps in the absence of cation migration.  
 65 Nevertheless, the last point concerning the cavity radius evolution during sub-  
 66 cascade overlapping is not consistent with the experimental results: in the CMD  
 67 simulations, the cavity average radius does not level off after 36 overlaps and  
 68 exceeds the experimental saturation value of  $\sim 0.5$  nm. This discrepancy may  
 69 come from the fact that the simulation box is too small for overlapping simu-  
 70 lation: the same material zone is systematically overlapped, while in reality, a  
 71 sub-cascade can connect and anneal zones of the material with different damage  
 72 characteristics (for ex. a vacancy-rich zone and an interstitial atom rich zone)  
 73 which finally should impact the growth rate of the defect clusters. An appropri-  
 74 ate way to study this growth rate would be intensive CMD simulations in a box  
 75 large enough to house several independent disordered zones. As the computing  
 76 cost for such a simulation is high and as an analytical formulation of the dam-  
 77 age is eventually sought for fuel performance assessment codes, an alternative  
 78 modeling strategy is proposed. The rest of this paper will then present and  
 79 assess a semi-empirical model of the sub-cascade overlapping.

## 80 2. The model

81 The basic idea of this sub-cascade overlap model is that when a new sub-cascade  
 82 impacts an already damaged zone, the existing damage is first annealed (be-  
 83 cause of the high temperature generated by the sub-cascade), and the damage  
 84 surviving to the annealing process is then added to that created by the new sub-  
 85 cascade. In the model, the space is discretized in cubic voxels, each schematically  
 86 representing a “site” or  $\text{UO}_2$  pattern (of volume  $0.043 \text{ nm}^3$ ), no detail is consid-  
 87 ered below this pattern so that U and O atoms are not considered separately:  
 88 all defects are supposed stoichiometric. This might be a crude approximation  
 89 for the smaller sizes, as for example non-stoichiometric (thus charged) defect  
 90 aggregates appear as a result of CMD simulations (e.g [10]) although the inter-  
 91 atomic potential does not allow the charge transfer liable to reduce the energy  
 92 cost of such non-stoichiometric clusters. Nevertheless, at room temperature and  
 93 above, the migration of charge and oxygen vacancies or interstitial atoms should  
 94 be fast enough to enable rapid relaxation of local departure from the exact sto-  
 95 ichiometry. At last, the proposed model is aimed at giving a general picture  
 96 of the irradiation damage accumulation; considering the very high sub-cascade  
 97 energy compared to the ionization and binding energies of defects, the main  
 98 characteristics of the primary damage (total number of O or U defects, volume  
 99 of aggregates) should not be very much affected by the possibility or not of  
 100 ionization and departure from stoichiometry.

101 In this respect the damage is composed of sets of Schottky ( $V_U(V_O)_2$ ) and anti-  
 102 Schottky ( $U_i(O_i)_2$ ) defects, respectively stated as “vacancies” or “interstitials  
 103 atoms” and accordingly coded by a number  $\xi_i = \pm 1$  at the defect position in  
 104 space (labeled by  $i$ );  $\xi_i = 0$  for undamaged voxels. Sub-cascades are succes-  
 105 sively generated at random positions, for each occurrence the resulting damage  
 106 is evaluated. Any sub-cascade generates a cubic disordered zone of volume  $V^s$   
 107 ( $n^s$  voxels) in which the  $\xi_i$  are updated according to the CMD results for sub-  
 108 cascade simulations ([10] Fig. 6) in a simplified way. The primary damage is  
 109 the same for all sub-cascades: one cavity at the center, one loop at the periph-  
 110 ery and some tens of isolated “vacancies” (Schottky) and “interstitial atoms”  
 111 (anti-Schottky). In the case of an initially undamaged piece of material, a total  
 112 number of  $n^p$  pairs of Schottky - anti-Schottky defects are generated, among  
 113 which  $n^p - n^c$  vacancies and  $n^p - n^c$  interstitial atoms are isolated and ran-  
 114 domly dispersed in the volume  $V^s$  (the vacancy preferentially occupying the  
 115 center of the disordered zone while the interstitial atoms lying more in the pe-  
 116 riphery). The remaining  $n^c$  vacancies are clustered: a cubic cavity of volume  
 117  $V^c$  ( $n^c$  vacancies with  $\xi_i = -1$ ) is created at the center. Symmetrically the  
 118  $n^c$  interstitial atoms cluster in a square shaped loop inserted near one of the  
 119 faces of the zone ( $n^c$  interstitials for which  $\xi_i = 1$ )<sup>1</sup>. Figure 2 shows the first  
 120 occurrences of the simulated damage process (isolated defects are omitted) and

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<sup>1</sup>The sizes of the cavity and loop could be slightly different. For simplicity they were supposed equal (to  $n^c$ ) in this version of the model.

121 the model parameters are summarized in Table 1. When the impact zone of the  
 122 sub-cascade is already damaged, the annealing step is first simulated supposing  
 123 that a defect annihilates if there is a defect of opposite sign in the disordered  
 124 zone. The surviving net damage  $\delta n$  in the algebraic sum of the defects initially  
 125 present in the zone:  $\delta n = \sum_{i \in V^s} \xi_i$ . This yields a loop (of size  $\delta n$ ) if  $\delta n > 0$ , a  
 126 cavity of size  $-\delta n$  in the opposite case. Secondly, this net damage is added to  
 127 the new damage described above, finally yielding a cavity of  $n^c + |\delta n|$  vacancies  
 128 and a loop of  $n^c$  interstitials if  $\delta n < 0$  or a cavity of  $n^c$  vacancies and a loop of  
 129  $n^c + |\delta n|$  interstitials if  $\delta n > 0$ . The  $2(n^p - n^c)$  isolated defects are added at ran-  
 130 dom positions in the volume  $V^s$  (the vacancies being closer to the center of the  
 131 disordered zone). Finally, as interstitial atoms are supposed to be very mobile  
 132 at room temperature [25, 24], a thermal annealing step is processed between two  
 133 successive sub-cascades following an object kinetic Monte Carlo (OKMC) pro-  
 134 cedure which interstitial atoms diffuse and can be irreversibly trapped by other  
 135 defects. Considering  $\Delta t_b$  as the average time period between two successive  
 136 sub-cascades, the program creates for each sub-cascade (at date  $t$ ) a list of all  
 137 the random diffusion events [8] (here anti-Schottky defects diffusion) occurring  
 138 in the time interval  $[t, t + \Delta t_b]$ . These events are successively activated (each  
 139 corresponding anti-Schottky is moved to its calculated final position) before a  
 140 new random sub-cascade is created at  $t + \Delta t_b$ .

### 141 3. An application of the model

142 The model has been applied to the irradiation of a  $\text{UO}_2$  foil with 4 MeV Au ions  
 143 at room temperature [20].

#### 144 3.1. Model parameters

145 Most of the model parameters were derived from studies implying various sim-  
 146 ulation techniques.

147 The sub-cascade energy was set to  $E_s = 25$  keV according to the analysis in  
 148 [4, 23] based both on CMD and BCA calculation. This is somehow simplified  
 149 as it does not simultaneously consider the U and O atoms nor variability of  
 150 this energy from a cascade event to another; a deeper analysis should lead to a  
 151 refined value of this energy, which in turn would slightly impact the number of  
 152 point defects generated by an average sub-cascade.

153 The primary damage characteristics for a 25 keV sub-cascade were extrapolated  
 154 on the basis of CMD simulations. They comprise the total number of pairs of  
 155 defects ( $n^p = 50$ ), the size of the larger cavity or loop ( $n^c = 3$  Schottky volumes)  
 156 [11], and the size of the disordered zone ( $n^s = 3375$  Schottky volumes at 300 K)  
 157 [13].

158 For the OKMC model, the anti-Schottky migration energy was estimated to  
 159  $E_m = 0.7$  eV and the diffusion pre-factor ( $D_0 = 1.22 \text{ nm}^2/\text{ps}$ ) is based on a  
 160 frequency close to the Debye frequency ([24, 9] which give orders of magnitude  
 161 similar to [25]).

## 162 3.2. Experiment characteristics

163 The larger part of the ballistic energy effectively deposited in the foil and re-  
 164 sponsible for the sub-cascades is that lost by the ion beam and is to the first  
 165 order proportionnal to the thickness  $h$ . But for low values of  $h$  a significant  
 166 part of the displaced atoms eventually leave the material through sputtering  
 167 and reduce the cascade yield. The effectively deposited ballistic energy  $E_b$  as a  
 168 function of the thickness (Figure 1 and Table 1) has been estimated on the basis  
 169 of ten SRIM [31] simulations of a 4 MeV Au beam in foils of thickness ranging  
 170 from 10 to 100 nm. Technically this energy has been computed as the differ-  
 171 ence between the ballistic loss of the beam and the total energy carried by both  
 172 the “sputtered” and “transmitted recoil ions” as described in the corresponding  
 173 SRIM output files.

174 The thickness  $h$  of the foil is thought to be in the range 10 – 50 nm on the  
 175 basis of EELS measurements of similar samples; the simulation results will help  
 176 fitting this parameter as will be seen below.

177 For irradiation with ions transferring a ballistic energy  $E_b$  to the material, the  
 178 fluency  $F$  in a foil of thickness  $h$  is related to the number  $i$  of sub-cascades of  
 179 energy  $E_s$  in the simulation cell of volume  $V^t$  through the differential equation  
 180  $dF \frac{V^t E_b}{h E_s} = di$ .

181 In the same idea, the time interval between two successive sub-cascades in the  
 182 simulation cell is  $\Delta t_b = 1 / \left( \frac{F E_b}{h E_s} V^t \right)$ .

## 183 3.3. Results

184 Several analysis of the model simulation were done, each with a value of the  
 185 thickness  $h$  ranging from 10 to 100 nm. The value  $h = 20$  nm was selected as  
 186 allowing the best fit for the cavity concentration and size distribution on the  
 187 basis of the different versions of Figures 4 and 6 (only the version with  $h = 20$  nm  
 188 is displayed here).  $h$  is the only fitted parameter in the process, all the other  
 189 ones are determined on the basis of measurements or simulations.

190 Figure 3 shows the simulated distribution of size (here the number of Schottky  
 191 or anti-Schottky defects) for cavities and loops at 3000 steps (all the results  
 192 are actually averages on 10 successive time-steps in order to smooth the time  
 193 evolution). At smaller sizes, the cavity and loop distributions strongly differ:  
 194 while wandering by diffusion, the interstitial atoms eventually encounter either  
 195 a cavity and annihilate (the cavity shrinks) or a loop (which grows). The larger  
 196 size region of the distribution graph is less concerned because large clusters  
 197 represent a negligible trapping cross section.

198 Figure 4 is an analysis of the void size (here radius) distribution for a fluency  
 199 of  $0.76 \text{ nm}^{-2}$ . The purple squarish graph represents the experimental data [20],  
 200 the light blue one is the raw histogram of the simulation results. Each smoothed  
 201 graph presents the simulated size distribution that should be observed in a TEM  
 202 image assuming <sup>1</sup>various values  $R_{TEM}$  for the optical resolution for cavities  
 203 (which is not very well known) and <sup>2</sup>a  $0.025 \text{ nm}$  ( $\sim 2$  pixels) uncertainty in the

204 cavity radius (see caption). A resolution of  $R_{TEM} \sim 0.35 - 0.40$  nm is consistent  
 205 with the experimental observations as can be seen in the next two figures.

206 Figure 5 displays the same comparison on a large range of fluency (between 0.076  
 207 and  $1. \text{ nm}^{-2}$ ). The agreement is best for the larger fluencies ( $F > 0.22 \text{ nm}^{-2}$ ),  
 208 provided the snapshot for  $F = 0.57 \text{ nm}^{-2}$  is considered as an outlier. For smaller  
 209 fluencies ( $F < 0.17 \text{ nm}^{-2}$ ), the model does not work so well: the total cavity  
 210 concentration is generally overestimated and the average size is overestimated or  
 211 underestimated without a clear tendency. This might be explained by the small  
 212 number of cavities that are created either in the experiment or in the simula-  
 213 tion, which produces statistical fluctuations of both simulated and experimental  
 214 quantities.

215 Figure 6 displays a realistic evolution of the total cavity concentration for various  
 216 threshold values  $R_{TEM}$ , also confirming the best value of  $R_{TEM}$  lies in the range  
 217  $0.35 - 0.40$  nm.

218 In conclusion, for a foil thickness  $h = 20$  nm, the model presented here re-  
 219 produces in a satisfactory way the experimental data and provides a plausible  
 220 explanation for the nucleation and growth of nano-cavities under ion beam ir-  
 221 radiation in the absence of thermal or irradiation-induced diffusion.

## 222 4. Discussion

223 We will first briefly discuss the possibility for the cavities to grow by diffusion  
 224 and secondly address the perspectives of applying this kind of model to the  
 225 modeling of fission gases behaviour.

226 As stated in the introduction, a brief analysis of the diffusion processes shows  
 227 that neither of the thermal or irradiation induced diffusion mechanisms of the  
 228 cation can explain the gathering of the cation vacancies surviving to the cascades  
 229 during the 400 s of the experiment:

230 – According to [14, 29], the order of magnitude of the thermal diffusiv-  
 231 ity would be  $D_{thermal} \sim 10^{-39} \text{ nm}^2/\text{s}$ , yielding a diffusion length around  
 232  $10^{-18}$  nm.

233 – The irradiation induced diffusivity comprises of a ballistic a non-ballistic  
 234 contribution. According to [12], the ballistic term writes:

$$D_{ballistic} = \frac{1}{2} \frac{\dot{F}}{h} E_b R_0^2 \Omega = 2 \times 10^{-4} \text{ nm}^2/\text{s}$$

235 where  $R_0^2 = 6.5 \text{ nm}^2/\text{keV}$  is the ratio between the square displacement and  
 236 the ballistic energy of a beam (as deduced from Figure 5.b of [12]). The  
 237 related diffusion length is around 0.3 nm.

238 – A model for the electronic (thermal spike) contribution linking the elec-  
 239 tronic energy loss of the ion and the diffusivity is proposed in [3] (eq. (12))



and (18)). The model parameters are taken in Table 5 of [3], except for values specific to this application: the range  $\mu = 20$  nm (order of magnitude of the foil thickness) and the electronic energy per ion  $E = 54$  keV (electronic energy loss calculated with SRIM for a foil thickness of 20 nm, cf. Fig 1).

The diffusion distances corresponding to the various possible diffusion mechanisms above are thus insufficient to explain the so big observed cavities. The mechanism proposed in this paper appears an appropriate alternative explanation.

Let us now consider the problem of fission gases, which appears to be somehow superimposed to the cavity production issue.

According to our simulation results, the TEM detects only a small part (1.7%) of the defect clusters, which represents 10% of the total vacancy volume (“porosity”). This means that if this model was in some way to be applied also to in-pile irradiation at low temperature (immobile cation vacancies), it is quite possible that the TEM observed cavities are devoid of Xe atoms because these atoms are likely to be trapped by the very small and abundant cavities as stated through XANES experiments [1].

The model also suggests ideas to explain similarities, and also differences, of the steady state distribution in many situations as stated in the introduction. In all these situations, the ion beams or fission fragments create similar 25 keV sub-cascades that have the same primary damage impact thus possibly yielding the same steady state distribution. Moreover, if the sub-cascade production is the only phenomenon affecting the micro-structure of the material (e. g. at very low temperature where no migration at all is expected) the evolution of the latter should be a function of the damaged volume fraction only; low temperature experiments would help validate this idea. In the opposite case (e.g. when the material is annealed between sub-cascades), other parameters not directly related to sub-cascades (such as defect diffusivity, or gas solubility) could account for variations in the material’s behavior and the micro-structure evolution may be accounted for by various time scales, depending on which defects are mobile at the considered temperature.

In the case of rare gas incorporation (through fission or ion implantation), the model needs to be adapted because gas atoms interact with the defects, moreover (cation) vacancies also diffuse at higher temperature relevant to many in-pile irradiation situations. Although several options are open for modeling sub-cascades interaction with cavities and loops in the vicinity of gas atoms, such a model may provide an interesting alternative to mechanisms often invoked in the literature to explain the steady state bubble distribution during in-pile irradiation.

- For instance, the interpretation of Russel or Veshchunov [19, 30] for the steady state distribution relies on an analysis of the nodal lines in the diagram (bubble gas content) VS (bubble-volume) obtained in the framework

284 of rate theory equations; the steady state occurs at the intersection of such  
 285 nodal lines at a stable critical point. In their analysis the presence of gas  
 286 appears essential to the steady state regime as it allows for two lines inter-  
 287 section, which would not occur in this framework if only self point-defects  
 288 were considered (corresponding to the bubble volume only). The interpre-  
 289 tation of the present article does not require gas atoms in principle and  
 290 might be relevant also for the cases studied by these authors. Assessing  
 291 the relevance of the various interpretations in an irradiation simulation  
 292 would be of high interest.

- 293 – Furthermore, the bubble steady state of certain models of fission gas be-  
 294 havior [18, 17] is grounded on a gas re-resolution mechanism either “ho-  
 295 mogeneous” (ballistic) or “heterogeneous” (induced by thermal spikes).  
 296 None of these mechanisms appear to be well understood yet: for exam-  
 297 ple <sup>1</sup>)the ballistic term as calculated in [16] has been recently reevaluated  
 298 and shown to be overestimated of a factor as large as 50 [22]; <sup>2</sup>)the het-  
 299 erogeneous term has been evaluated by various CMD approaches whose  
 300 conclusions do not agree [6, 5]. Taking into account the mechanism de-  
 301 scribed here and the fact that the “bubbles” could be devoid of gas atoms,  
 302 in addition to a better account of point defects and small clusters for in-  
 303 pile irradiation situations might also help re-assess the relevance of the  
 304 various re-resolution models .

## 305 5. Conclusion

306 A model for damage accumulation in  $\text{UO}_2$  under irradiation is proposed to  
 307 account for the fact that, during irradiation of a thin foil at room temperature  
 308 in the ballistic regime, nanometer sized cavities appear and reach a quasi steady  
 309 state size distribution although no gas is implanted and no cation diffusion is  
 310 expected. The basic ideas of the model are:

- 311 – any displacement cascade is split into equivalent sub-cascades of  $\sim 25$  keV,  
 312 each producing a highly disordered region in the material that eventually  
 313 imperfectly recrystallizes, leaving isolated point defects, a small cavity and  
 314 a small loop;
- 315 – the overlapping of a new sub-cascade on an already damaged zone first  
 316 anneals the defects included in the zone and secondly accumulates the  
 317 damage surviving to the annealing with that associated to the new sub-  
 318 cascade;
- 319 – in addition to the sub-cascade driven processes, the material’s micro-  
 320 structure evolves under thermal migration of the interstitial atoms; this  
 321 process is simulated through an Object Kinetic Monte Carlo algorithm.

322 The simulated evolution of the cavity size distribution compares favorably with  
 323 the TEM in-situ observations at various fluencies in the case of an irradiation

324 with 4 MeV Au ions which is encouraging since the only fitted parameter of  
325 our model is the foil thickness. In particular, the simulated steady state size  
326 distribution is similar to the observations at the same fluency. The cavity nu-  
327 cleation mechanism and growth is consistent with a heterogeneous nucleation  
328 mechanism for fission gas bubbles.

329 The quasi steady state cavity distribution is not very sensitive to the experi-  
330 mental conditions (namely the beam energy and flux, and to a lesser extend the  
331 temperature). This could be accounted for by the fact that the displacement cas-  
332 cades split in any situation in 25 keV sub-cascades which are actually responsible  
333 for the primary damage in the material. By the way, if the mechanism proposed  
334 here is confirmed to be relevant in the case of fission product irradiation (which  
335 remains to be done), it could contribute to explaining the quasi steady size dis-  
336 tribution during in-pile irradiation in an alternative or complementary way as  
337 invoked in various fission gas codes, such as irradiation-induced resolution or  
338 stable nodes in the diagram (bubble gas content) VS (bubble volume).

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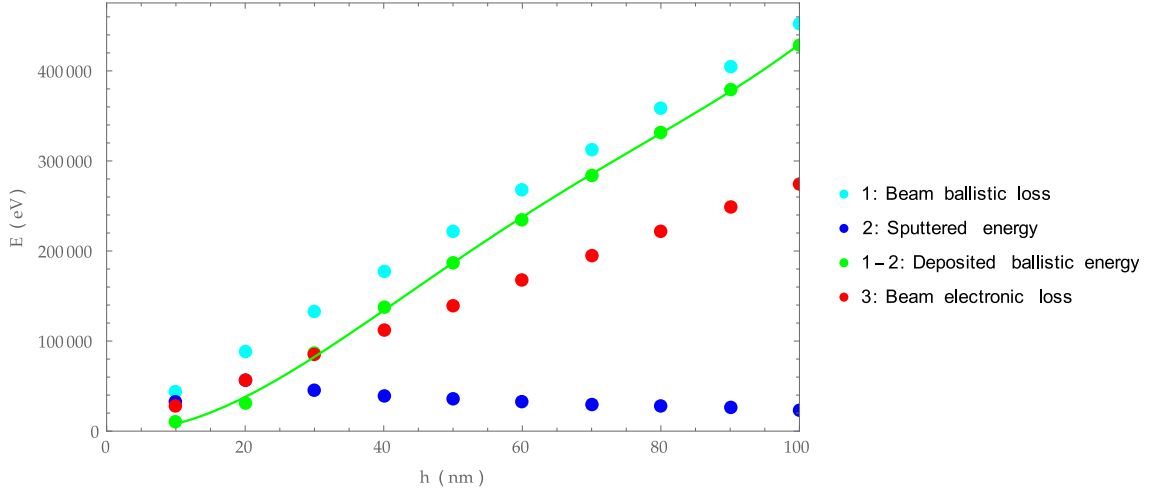


Figure 1: Deposited ballistic energy (difference between the beam ballistic loss and the sputtered energy) as a function of the foil thickness.

<b>Experiment parameters</b>		
$h$	20 nm	Foil thickness
$E_b$	31 keV	Ballistic energy transferred to the foil (taking account of the sputtering): $E_b$ (eV) = $3321.17 - 1146.41h + 184.607h^2 - 2.22491h^3 + 0.00919919h^4$ , $h$ (nm) (Fig 1)
$E_s$	25 keV	Sub-cascade energy
$F$		Fluency
$\Delta t_b$	0.06 s	Average time between 2 sub-cascades in the simulation cell
$T$	300 K	Temperature
<b>Sub-cascade model parameters</b>		
$n^s$	3375	Number of voxels for the disordered zone
$n^c$	3	Number of voxels for the clusters (cavity and loop) nucleated in the sub-cascade
$n^p$	47	Number of pairs (Schottky/anti-Schottky) of isolated defects (1 voxel) per sub-cascade
$N^t$	$10^6$	Number of voxels for the simulation cell
<b>UO<sub>2</sub> related physical properties</b>		
$E^m$	0.7 eV	Migration energy for anti-Schottky defect [24, 25]
$D_0$	$1.22 \times 10^{12}$ nm <sup>2</sup> /s	Diffusivity coefficient at room temperature, $T = 300$ K ( $D = D_0 e^{-\frac{E^m}{kT}}$ ) [24]
$V$	0.0427 nm <sup>3</sup>	Volume of the Schottky defect
$a$	0.555 nm	Size of the Schottky defect considered as a cube: $V = a^3$
$V^s$	150 nm <sup>3</sup>	Volume of the disordered zone $V^s = n^s V$
$V^c$	0.176 nm <sup>3</sup>	Volume of the cluster $V^c = n^c V$
$V^t$	43961 nm <sup>3</sup>	Volume of the simulation cell $V^t = N^t V$

Table 1: Parameters for the simulation (implantation of 4 MeV Au atoms).

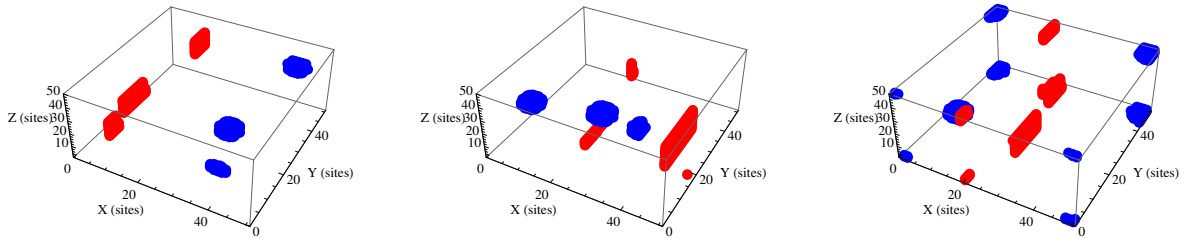


Figure 2: Example of micro-structure after 2, 5 and 10 sub-cascades, the isolated defects are omitted. For example , periodic boundary conditions can be seen in the first image where a loop and a cavity are split two ways.

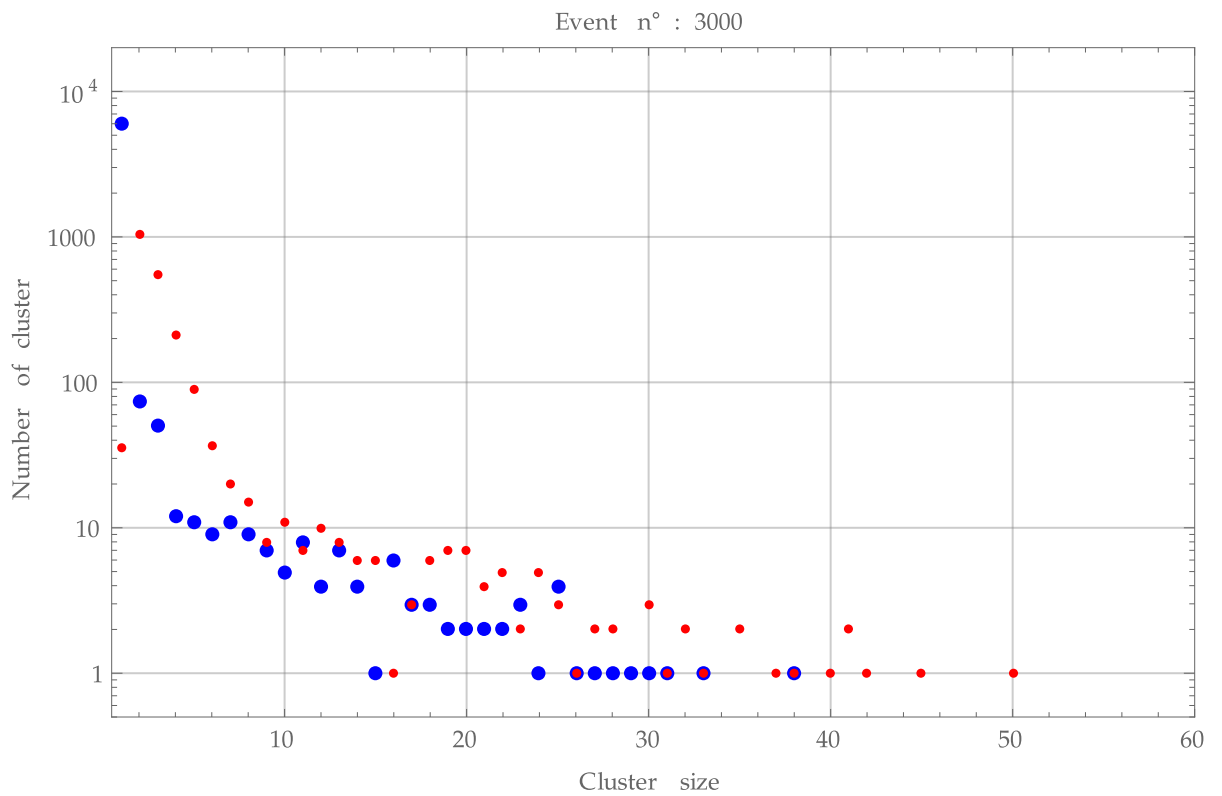


Figure 3: Cluster size distribution after 3000 iterations ( $F \sim 1 \text{ nm}^{-2}$ ). The blue and red dots correspond to cavities and loops.



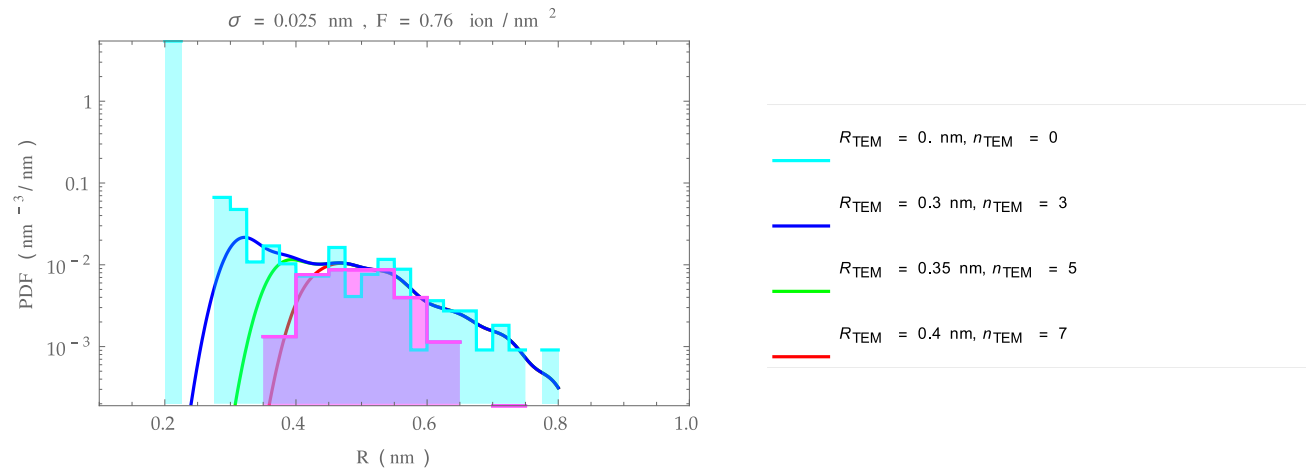


Figure 4: Impact of the TEM sensitivity on the measured size distribution. The purple curve is the measured distribution (TEM observations) at a fluency  $F \sim 0.76 \text{ nm}^{-2}$ . The light blue curve is the as simulated histogram of the distribution. The other curves (blue, green, red) describe the simulated distribution with various thresholds  $R_{\text{TEM}}$  for the TEM resolution and smoothed with a rms of 0.025 nm (corresponding to the camera resolution for the cavity limit).

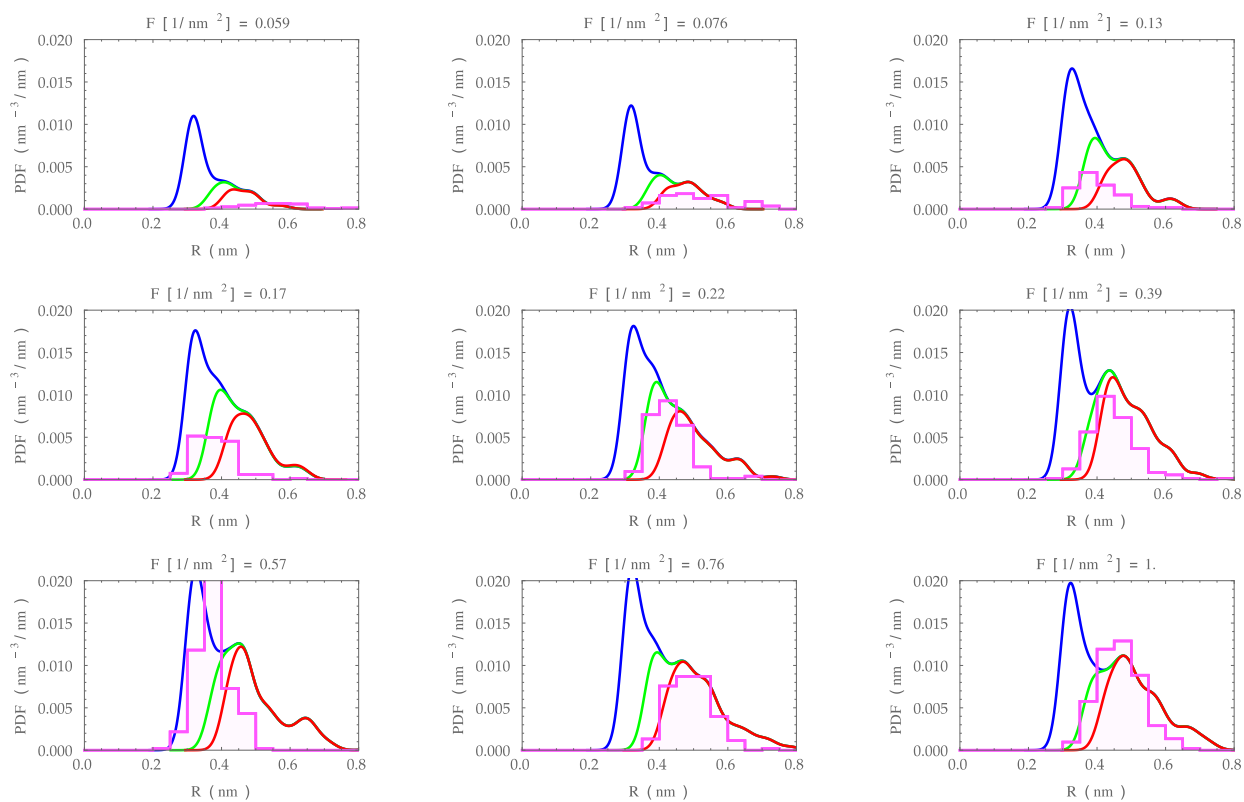


Figure 5: Snapshots at various fluencies of the simulated (with  $R_{TEM} = 0.30, 0.35, 0.40$  nm) and measured size distributions (see Figure 4 for colors).

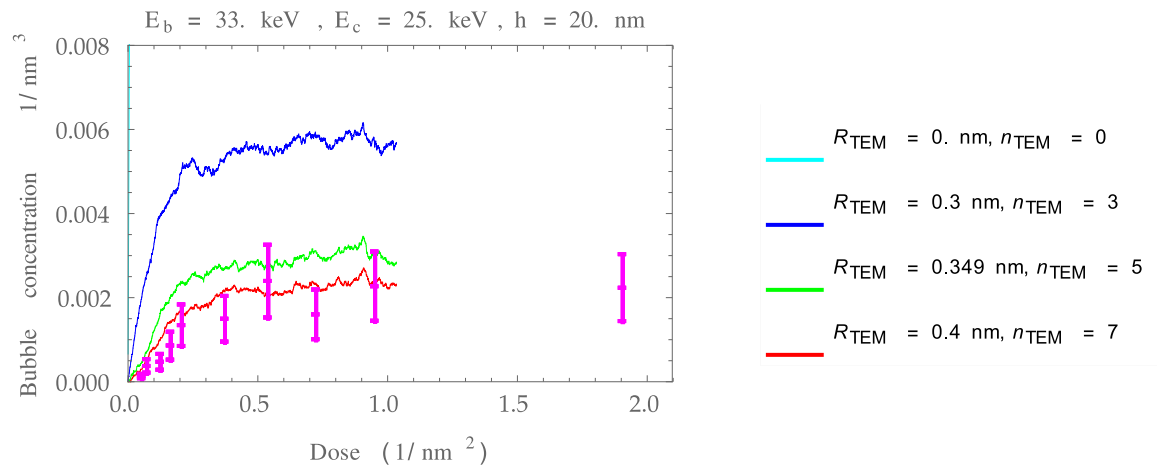


Figure 6: Evolution of the concentration of the cavities larger than some threshold values  $R_{TEM} = 0., 0.30, 0.35, 0.40 \text{ nm}$ .