CNEA/ANL COLLABORATION PROGRAM TO DEVELOP AN OPTIMIZED VERSION OF DART VALIDATION AND ASSESSMENT BY MEANS OF U_3Si_X AND U_3O_8-Al DISPERSED CNEA MINIPLATE IRRADIATION BEHAVIOR

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Abstract

The DART code is based upon a thermomechanical model that can predict swelling, recrystallization, fuel-meat interdiffusion and other issues related with MTR dispersed FE behavior under irradiation. As a part of a common effort to develop an optimized version of DART, a comparison between DART predictions and CNEA miniplates irradiation experimental data was made. The irradiation took place during 1981-82 for U3O8 miniplates and 1985-86 for U_3Si_x at Oak Ridge Research Reactor (ORR).

The microphotographs were studied by means of IMAWIN 3.0 Image Analysis Code and different fission gas bubbles distributions were obtained. Also it was possible to find and identify different morphologic zones. In both kinds of fuels, different phases were recognized, like particle peripheral zones with evidence of Al-U reaction, internal recrystallized zones and bubbles.

A very good agreement between code prediction and irradiation results was found. The few discrepancies are due to local, fabrication and irradiation uncertainties, as the presence of U_3Si phase in U_3Si_2 particles and effective burnup.

Introduction

This work concerns validation and assessment of DART[1] code, as a part of a CNEA and ANL collaboration program for the development of an optimized DART version. A comparison between U_3Si_x and U_3O_8 Al dispersed CNEA miniplate irradiation behavior and DART predictions, is made. For this purpose, IMAWIN 3.0[2], digital processing image code was applied to analyze several post-irradiation microphotographs, where different pore and phases areas were detected and measured.

DART code is based upon a thermomechanical model that can predict swelling, thermal conductivy evolution, recrystallization, U-Al reaction, aluminization depth and other issues related with MTR dispersed FE behavior under irradiation[3].

CNEA miniplates consist of two set, one concerning U_3Si and U_3Si_2[4] and another one concerning U_3O_8 and UAl_x[5], LEU Al dispersed fuels. These miniplates were irradiated at ORR in Oak Ridge National Laboratory, from 11/27/85 to 11/18/86 and from 6/20/81 to 10/82 in each case. Their respective behavior under irradiation was satisfactory.

IMAWIN is under development since 1994 at the Advanced Fuels Elaboration Division of the Nuclear Fuels Unit in Constituyentes Atomic Center, CNEA. It is capable to detect, recognize and measure different morphologic zones that are present in post-irradiation micrographs, metallographic inclusions and other image storage devices, like surface areas, borders, width and length of connected subsets, etc. IMAWIN 3.0 captures and transforms images into a digital form in appropriate scale, finds digital image contrast and prepares different data outputs like particle and pore size distribution using Johnson-Saltykov[6] methods, percentile area coverage for each zone and other issues.

General Considerations

Volume swelling was obtained by immersion method. The meat swelling found was solely due to fission product, particle swelling and aluminide formation. It is supposed that no swell took place in Al.

U_3Si_2-U_3Si miniplates behavior comparison

It is supposed that particle swelling first closes the fabrication porosity, and then a net meat swelling occurred. Under these suppositions, with meat swelling data, it is possible to evaluate the 'effective' particle swelling as:

'Effective' particle swelling = (meat swelling + porosity)/ Fuel volume fraction

The word 'effective' stands also for taking into account Al missing after aluminide reaction. As it has been already seen, aluminide formation could origin a big swelling in particle but not in meat.

Miniplate	RA311	RA313	RA315	RA316	RA319	RA320	RA321
Fuel	**U_3Si**	**U_3Si_2**	**U_3Si_2**	**U_3Si_2**	**U_3Si_2**	**U_3Si_2**	**U_3Si_2**
Volume fraction (%)	**36**	**44**	**44**	**44**	**44**	**44**	**44**
Miniplate thickness (µm)	**1524**	**1524**	**1524**	**1524**	**1270**	**1270**	**1270**
Meat thickness (µm)	**760**	**760**	**760**	**760**	**510**	**510**	**510**
Porosity (%)	**7.75**	**7.56**	**8.01**	**7.96**	**8.76**	**8.66**	**9.11**
Meat and Sheath Al-type	**6061**	**6061**	**6061**	**6061**	**6061**	**6061**	**6061**
Uranium Density (g/cm^3)	**4.81**	**4.87**	**4.85**	**4.85**	**4.82**	**4.83**	**4.81**

TABLE 1: U_3Si_x miniplates fabrication data

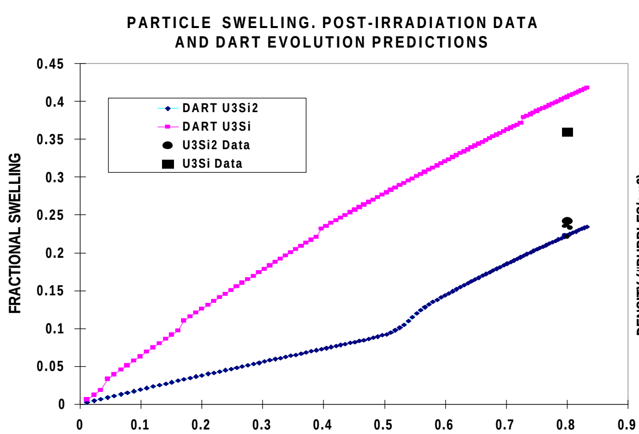


FIGURE 1

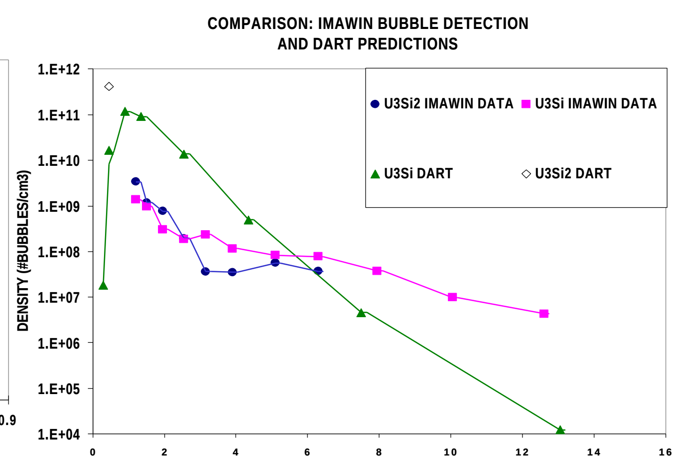


FIGURE 2

For DART simulation, it was employed a 20 radial node partition, a particle average diameter of 100µm, a boundary and center particle temperature of 373 K and 383 K respectively. The fission rate f' diminishes proportionally with burnup. It reached 80% atU^{235} after 273 days. It was calculated an exponential decay, which integrated media, coincides with that from data.

DART predicts recrystallization at $3.5*10^{21}$ fiss./cm3 fission density, corresponding to 50% burnup approximately. Once recrystallization happens, nucleation increases and so happens with grain corner bubbles population. There is an increase of particle swelling rate.

In the miniplates micrographs it is clearly seen aluminide formation in boundary particles (figs.3 and 4). DART aluminide depth prediction (3.25µm for U_3Si_2 and 3.66µm for U_3Si) is in concordance with measurements. DART bubble distribution predictions are shown in figures. 5 and 6 for different regions: recrystallized U_3Si_2 zone and amorphous U_3Si zone.

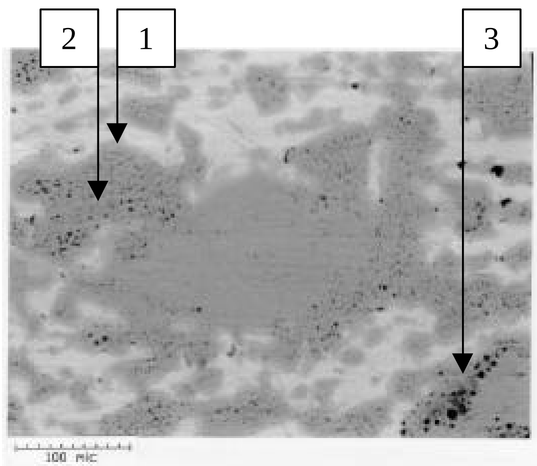


Figure 3. Irradiated U_3Si_2 Microphotograph. Miniplate RA321.

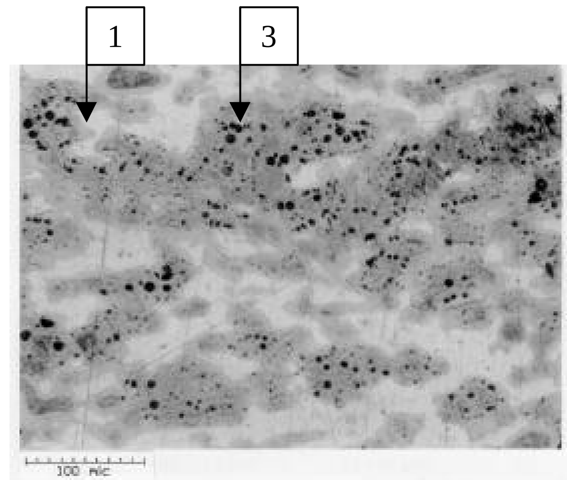


Figure 4. Irradiated U_3Si Microphograph. Miniplate RA311

It is observed aluminide formation around fuel particles (1,light grey) with no bubble presence. Recrystallized zones are present (2, little black dots). Amorphous structure zones present greater bubble size than in crystalline ones (3, black spots). IMAWIN 3.0 bubble population detection is in concordance with DART predictions.

- <u>Aluminide region</u>: DART predicts a small bubble distribution (diameter less than 0.01 µm). This is consistent with bubble free aluminide zone observation (figure 3 and 4, quote 1) (bubble radii dimensions are beyond photo resolution).
- <u>Recrystallized U_3Si_2</u>: DART predicts a bimodal bubble distribution (figure 6), with the second peak due to bubbles pinned at recrystallized grain corners. It would have an average diameter of 0.5µm. Although it is also beyond photo resolution, DART does not take into account the diameter spread of the distribution, centered at 0.5 µm. So it is possible greater diameter bubble population. In U_3Si_2 micrograph (fig.2, q2) it is shown zones associated with this kind of morphology.
- <u>Amorphous U_3Si</u>: DART predicts a greater diameter bubble population (1-12µm) (fig.5) than in U_3Si_2 case. This phase can be seen in fig. 4, q3, and also in fig. 3, q3, revealing coexistence of U_3Si phase in U_3Si_2 fuel miniplates.

In fig. 2, a comparison between IMAWIN 3.0 detection and DART predictions for amorphous U_3Si and recrystallized U_3Si_2 grain corner bubble peak is shown.

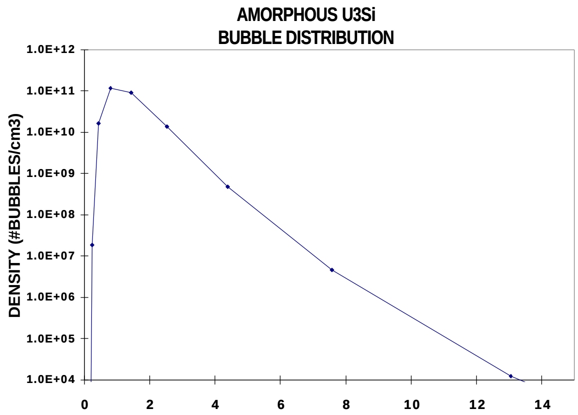
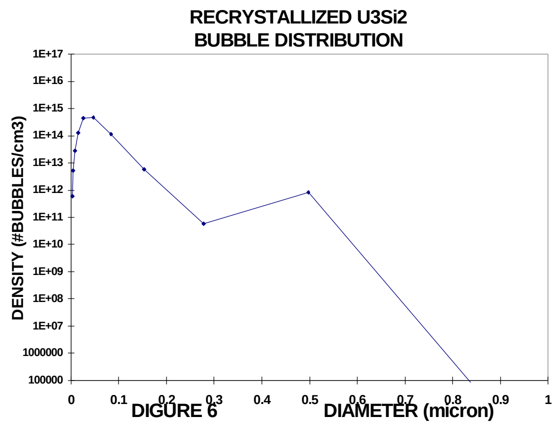


FIGURE 5



DIGURE 6

In U_3Si_2 case it was observed a bubble distribution associated with recrystallized grain corner bubbles (1-3μm). There was found also another peak at 5 μm, probably due to not closed as-fabricated porosity. The U_3Si-like morphology zones are probably due to phase coexistence during fabrication process.

In U_3Si case, it is observed a great diameter bubble distribution (up to 12μm). It is in qualitative agreement with DART predictions. The discrepancies are mainly due to photo resolution. Besides, IMAWIN 3.0, due to contrast resolution, while recognizing great bubbles looses small ones and in some cases, it is not able to distinguish between two bubbles separately, recognizing only one. This lack of precision produces an overestimation of great bubbles and an underestimation of small bubble populations. Another mention is that the breakaway swelling would start in U3Si at a similar fission density that the sample had reached. This phenomenon causes great bubbles interconnection and a huge particle swelling and it is not modeled by DART. The code only predicts U3Si behavior above this fission density.

U_3O_8 miniplates behavior comparison

Miniplate	RA209	RA218	RA219	RA222
Fuel	**U_3O_8**	**U_3O_8**	**U_3O_8**	**U_3O_8**
Volume fraction (%)	**35.2**	**41.6**	**35.1**	**44.6**
Miniplate thickness (μm)	**1520**	**1270**	**1530**	**1530**
Meat thickness (μm)	**900**	**520**	**740**	**740**
Porosity (%)	**6.17**	**9.04**	**7.00**	**10.04**
Meat Al-type	**99.5%wt. Al**	**99.5%wt. Al**	**99.5%wt. Al**	**99.5%wt. Al**
Sheath Al-type	**6061**	**6061**	**6061**	**6061**
Uranium Density (g/cm^3)	**2.47**	**2.91**	**2.46**	**3.12**

TABLE 2: U_3O_8 miniplates fabrication data

DART U_3O_8-Al reaction and irradiation sintering models were already presented[7]. Both phenomena contribute negatively to swelling. For DART simulation, it was employed a 20 radial node partition and a temperature of 373K at border and 383 K at center of particle. This subset of miniplates reached a final burnup of 87% at. U^{235}, after 352 days of full power. The average particle diameter was 80 μm.

DART swelling prediction and Al volume fraction evolution show a good agreement with data (figs. 7 and 8). However, swelling prediction is dependent on porosity sintering model employed by DART, so it is very sensitive to initial porosity uncertainties.

Post-irradiation microphotograph concerning U_3O_8 miniplates exhibit a more complex structure than in U_3Si_x case. Although U-Al reaction is widely generalized -large zones of aluminide phase are present (figure 9, q3)-, Al volume fraction always was under 10%. At this point, DART predicts softening and the beginning of general interconnection bubble phenomenon.

U_4O_9 globular unreacted recrystallized-like zones appear in microphotographs (fig. 9, q1). This phase may be due to fuel lamination or an intermediate U-Al reaction. Bubble distribution is similar to that found in former U_3Si_2 miniplate micrograph analysis.

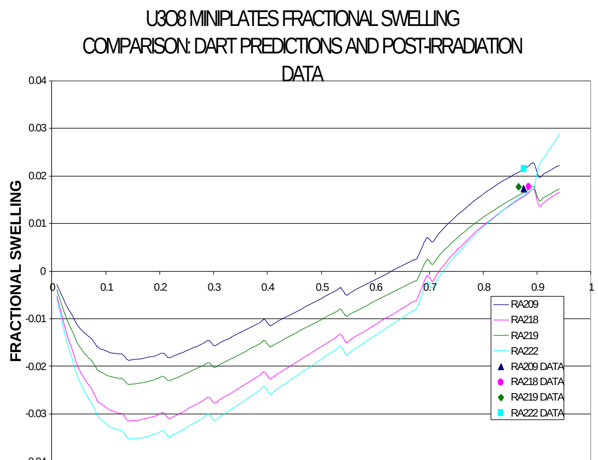


FIGURE 7 BURNUP (%at U235)

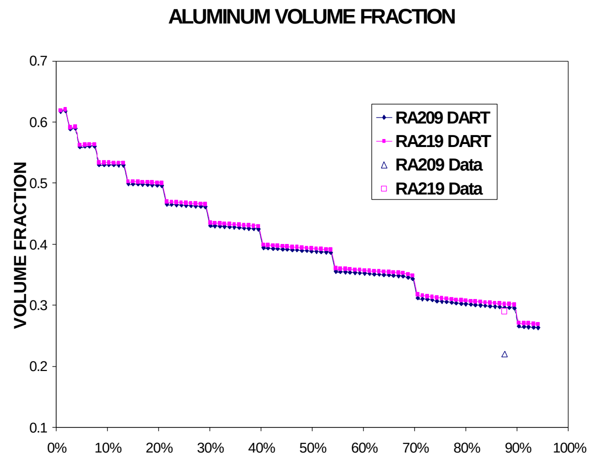
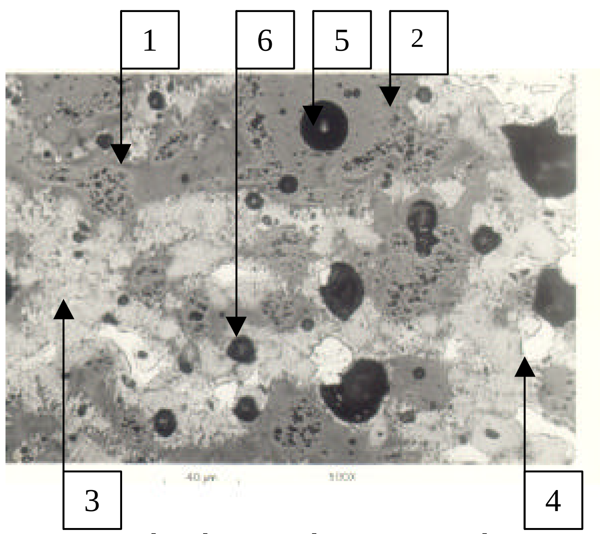


FIGURE 8 BURNUP (%at.U235)



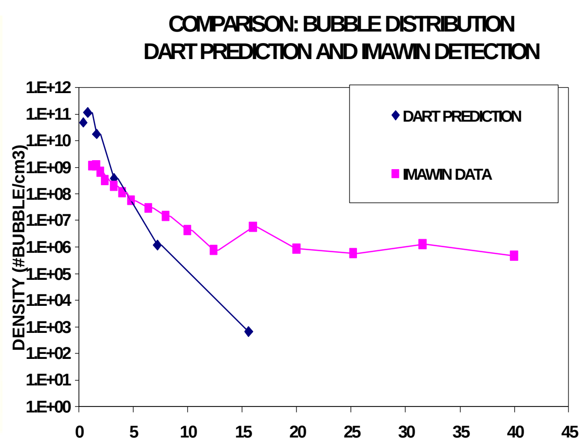


FIGURE 10 DIAMETER (micron)

FIGURE 9. Irradiated U_3o_8 Fuel. RA209 Miniplate. Burnup = 87%. It is observed U_4O_9 phase, in globular nucleus with bubbles (1, dark grey with black spots) of about 1-4 µm, a surrounding U_3O_8 unreacted area (2, dark grey), a wide aluminide reacted area (3, light grey) with bubbles up to 10µm (6, black), islets of no reacted Al (4, white) and big spherical porosity area (5, black)

For aluminide region DART predicts a bubble distribution centered at diameter = 1 µm and spreading to 10µm, several order of magnitude below. While the peak is not observed, there are present some bubbles up to 10µm (fig. 9, q6). Huge spherical bubbles were also observed (fig. 10). They could be due to as-fabricated porosity evolution and/or to an early stage of breakaway swelling

Conclusions

U_3Si_x case

- A good agreement between DART prediction and IMAWIN 3.0 samples measure was found. Prediction of swelling, U-Al interdiffusion depth and bubble distribution, are in concordance to observation for both silicide miniplate fuels. Nevertheless, for bubble distribution it would be necessary to have more accurate measurements as SEM micrographs. The observed discrepancies can be due to measurement uncertainties and to parameters used during simulation, as final burnup and as-fabricated porosity.
- CNEA U_3Si_2 miniplates photos show U_3Si-like morphologic zones. It is an evidence of this kind of phase presence.
- Aluminide formations at particle boundary acts as additional constrain for particle swelling. In U_3Si_2 case, because there is no recrystallization for aluminide, its swelling rate is lower than recrystallized fuel. In U_3Si case, aluminide is a crystalline phase, and its swelling rate is lower than that of amorphous fuel.
- The nodal radial partition as well as the number of bubbles classes follows from a precision-to-time computing ratio. If the simulation is carried out with few radial zones, aluminide formation is not predicted, and consequently a greater particle swelling is obtained. In this comparison it was employed a 20 radial mesh.
- U_3Si_2 fuel behavior is quite different from amorphous U_3Si. This last one shows a much greater swelling than crystalline fuel.

U_3O_8 case:

- It is also observed a good agreement between DART swelling and U-Al reaction predictions, and IMAWIN 3.0 detection. Initial pore uncertainties has a strong influence in swelling, via porosity sintering model.
- DART microstructure predictions, concerning bubbles found in U_4O_9 globular recrystallized zone, follows the trend showed by IMAWIN 3.0 stereology analysis.
- For aluminide zone, bubble distribution peak calculated by DART is not observed in CNEA miniplates. Huge bubbles present in samples may possibly be due to initial porosity evolution or to generalized interconnection.

DART predicts Al dispersed fuel behavior (rod, tube and plate geometry). It has models for bubble population distribution, mechanical behavior, bubble swelling, thermal conductivity, aluminide reaction and radiation-induced recrystallization. IMAWIN 3.0 captures, detects and measures micrograph zones of diverse morphology. In the common zone of application, defined by scope of optical media, it was performed a comparison between DART predictions and IMAWIN 3.0 detections. The outcome of this comparison, taking into account the performance of the different models included in DART, is satisfactory

Program for the development of a new and optimized DART version

DART was conceived as a mechanistic model for the assessment of dispersion fuel behavior for oxide, silicide, and other new dispersant phases (vhd alloys). However, due to its evolution as an R&D tool, it was not developed as a user-friendly code. Besides, each simulation done to study the effect of changing a particular parameter or operating condition, demands the iteration of a process consisting in input preparation, DART run, extraction of calculated quantities from program output file and plotting. Each step of the calculation process has an extension of a couple of minutes to many hours, depending on complexity of the problem. For an analysis covering a multitude of parameters and/or operating conditions, this is a very long and tedious process. In addition, different versions of DART exist for oxide, silicide, and for vhd candidate alloy fuels.

As a part of SISTERLAB agreement, ANL and CNEA have proposed several topics for mutual collaboration. One of them is related with modeling. It consists in a full revision of DART models and version codes and the inclusion of new models in the framework of the development of a unique parallel architecture version for DART code. The aim pursued is to

1. Enhance DART I/O by means of a complete reworking; in order to increase its availability and usefulness in the international community. The conversion of DART into a parallel architecture version is an ideal place to implement such improved interfaces.
2. Afford the opportunity to develop an interface whereby the user can monitor the evolution of various calculated quantities "in situ." In addition, it will provide the possibility for the user of changing values of various parameters and/or operating conditions during the course of a run. The user/code dialog will become highly optimized and the analysis procedure will be more efficient.
3. Parallelize a variety of calculations performed as a function of operating conditions and fuel morphology, like

- Evolution of the fission-gas bubble size distribution and meat thermal conductivity.
- Fuel-meat matrix interaction
- Evolution of fuel microstructure
- Stress/strain analysis,

and other issues. These processes will be parallelized providing for a much more efficient calculation.

4. Allow the opportunity to merge all different versions of DART into a single code.
5. Facilitate the development of new models such as

- Superplasticity
- Elastoplastic feedback
- Improved models for the calculation of fuel deformation and fuel microstructure evolution

6. Provide an opportunity for a rigorous inspection and overhaul of DART bringing to the user and developer of the international community a very valuable benchmark.
7. Form the basis of a code for the analysis of dispersion fuel during transient (and/or accident) conditions.

The conversion of DART to parallel architecture will facilitate its potential development.

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