# Study of uranium americium oxide behaviour under high helium production conditions at the Jules Horowitz reactor

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## Abstract

*The reaction chain $^{241}Am$ (n,γ) $^{242}Am$ (d,β) $^{242}Cm$ (d,α) $^{238}Pu$ results in a high He production in transmutation targets with significant minor actinide content, which are planned in several Generation IV reactor designs. For example total helium production is envisioned to reach up to 9 mg/cm$^3$ in 20% MA-bearing blankets in a SFR at EOL. Currently the DIAMINO/MARIOS [4] programmes at OSIRIS and HFR are under way to investigate the swelling and gas release of small $Am_{0.15}U_{0.85}O_{2-x}$ samples depending on porosity and temperature of the samples. This paper describes a follow-up experiment, which is studied at the JHR with the aim to additionally isolate the effects of burn-up and total helium production on the sample behaviour to provide data for the modelling of helium in oxide fuels and more specifically data for the design of the minor-actinide-bearing blanket at the SFR. It is shown that by modifying the uranium enrichment, the americium content (in a proportion which makes it possible to preserve the fuel microstructure) and the neutron flux level, helium production and burn-up can be manipulated separately. This approach may pave the way for a simulation of irradiation damages of fast reactor fuels by using experiments carried out in thermal neutron reactors.*

## Introduction

In order to close the nuclear fuel cycle several concepts envisage high MA content targets in either fast reactor blankets or as ADS fuel. One of the problems with high MA fuel, especially Am-rich fuel, is that it is known to have a very high He production compared to MOX fuel (~15 × MOX LWR for 10% Am content), but there is few data available about fuel behaviour under such conditions. Additionally MTR have to be utilised for testing whenever possible because there are currently few sufficiently intense fast neutron sources in operation and this is not likely to change soon. For americium oxide the DIAMINO/MARIOS [4] irradiation programmes at OSIRIS and HFR are under way to investigate the swelling and gas release of small $Am_{0.15}U_{0.85}O_{2-x}$ samples depending on porosity and temperature of the samples. This paper presents investigations about feasibility and options for a follow-up programme at JHR with the aim to provide data for modelling fuel swelling behaviour in fast reactors under high gas production conditions.

## Concept

DIAMINO/MARIOS [4] is essentially focused on an experimental matrix of sample porosity (8% and 13%) vs. temperature (four steps from 600°C to 1 200°C) with a He production of 3-4.5 g/cm³. Adding a He production vs. burn-up matrix for a selected sample temperature may be beneficial for a better modelling of fuel behaviour, because these are the main drivers for behaviour at a given temperature. As JHR is scheduled to take over from OSIRIS in 2016 the experimental set-up of DIAMINO may need to be ported to JHR, preferably with limited changes. In a previous conference paper [5] the possibility of covering a wide range of He production and fission rates in a thermal spectrum was demonstrated by varying uranium enrichment and neutron flux. While this allows covering many expected fast spectrum cases, it is generally not possible to match both the end state and time behaviour of the fuel burn-up closely. For screening experiments the focus has to be on the final compositions.

### *Effect of fast/thermal neutron spectrum*

Because fast neutrons carry less than 2.8% of the energy of fission products, the neutron spectrum has only an indirect impact by the way of different fission and capture rates. The actual processes producing the He and fission to reach equivalent values for the fast and thermal spectrum need not necessarily be the same as long as the different chemical composition does not change the fuel behaviour and the sample is homogeneous enough. Homogeneous enough in this case means that the isotopes of interest are homogeneously distributed to each other to well below the mean path length of fission products (~10 μm). This rules out macro-dispersed inert matrix fuels and might give troubles with MOX-type fuels in future experiments.

The main contribution of the He production comes from a single reaction chain [$^{241}$Am (n,γ) $^{242}$Am (d,β) $^{242}$Cm (d,α) $^{238}$Pu] with only one important neutron interaction and so the difference is basically given by the difference in capture cross-sections of $^{241}$Am between thermal and fast spectrum. Unfortunately for fission the case is rather more complex. The relative contributions of three main fissile isotopes $^{235}$U, $^{239}$Pu and $^{242m}$Am depend strongly on the spectrum because of both different breed rates and different relative fission cross-sections. Also, the power ramp-up at beginning of irradiation due to fissile isotope breeding is far more pronounced in a short duration thermal spectrum experiment but can be mitigated somewhat by uranium enrichment.

The effect of the higher self-shielding in the thermal spectrum is reduced by the small sample size (diameter 4.5 mm, thickness 1.5 mm), which is also necessary to reduce the temperature gradient in the sample. Figure 1 shows the neutron spectra for the SFR blanket and the JHR reflector. The JHR reference spectrum was taken from an irradiation position next to the core (C313) resulting in a sizeable fast neutron fraction. Due to the far higher thermal cross-sections this fast fraction has negligible effect on the reaction rates but would allow increasing the reaction rates further with additional moderation if necessary (not the case for oxide samples because of the high temperature gradient).


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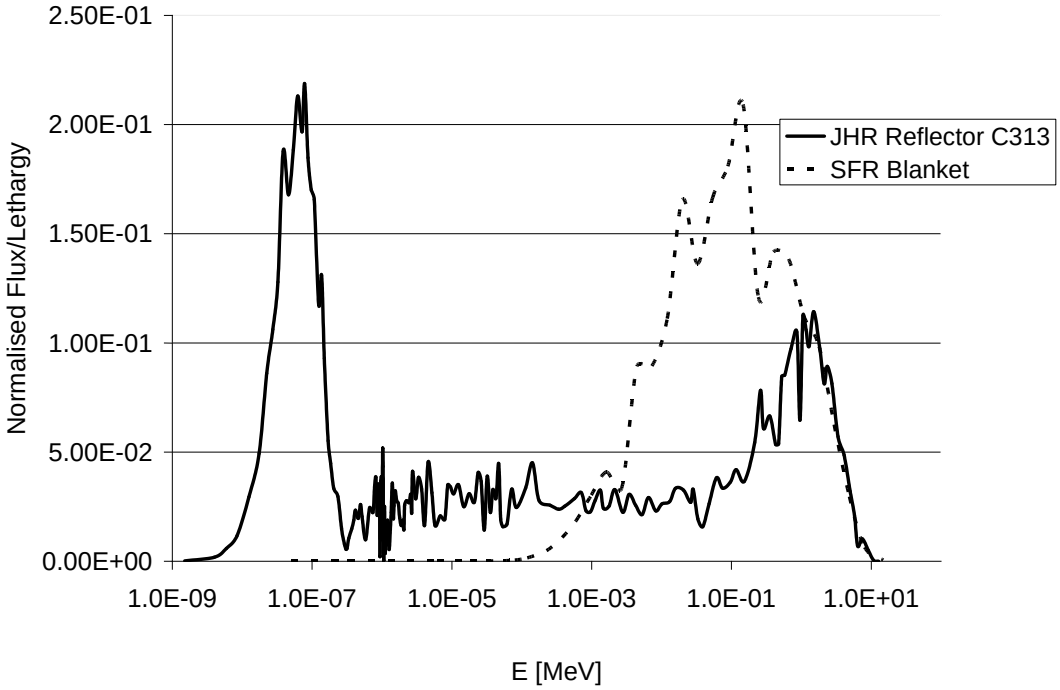
**Figure 1: Neutron spectrum per lethargy**




## Geometry

Currently two fixed positions in the JHR reflector are considered for the experiment: C313 next to the core (distance casing – centre position 7 cm) and P422 (distance casing – centre position 26 cm). As both have an inner diameter of 10 cm but the outer diameter of the experimental device (PHAETON-type) is only 3.31 cm, there is plenty of room to adjust the neutron flux by either positioning or shielding. In the central tube of the experimental device are 6-8 closed steel capsules (diameter 1.6 cm, height 6.7 cm) containing six sample discs each (diameter 0.45 cm, height 0.15 cm) with Mo support for gamma heating and thermal conductivity. This results in six to eight measurement points per reactor irradiation position used.

## Methodology

Like in the preliminary study [5] the depletion code DARWIN-2.2 [3] with JEF-2.2 library was used to handle the JHR burn-up calculation and the SFR reference sample. As a reference sample serves $Am_{0.15}U_{0.85}O_{2-x}$, as used in the DIAMINO/MARIOS experiment. Starting from the reference sample U enrichment, neutron flux level and Am content were modified in the JHR spectrum to discover the range of possible He content/burn-up results after 208 efpd and 416 efpd (1 and 2 years of reactor operation). Table 1 shows reference case for the reactor spectra with Figure 1 giving the different spectra. The spectra and self-shielding estimates are based on TRIPOLI-4 [2] calculations for JHR and ERANOS calculations for SFR by V. Brun-Magaud and L. Buiron [1]. The depletion calculations were done in one-hundred 45-day steps at constant power and self-shielding for the SFR case and 16 cycles of 26 days operation and 20 days cool-down for JHR.

**Figure 2: Positions in the JHR reflector envisaged for the experiment**

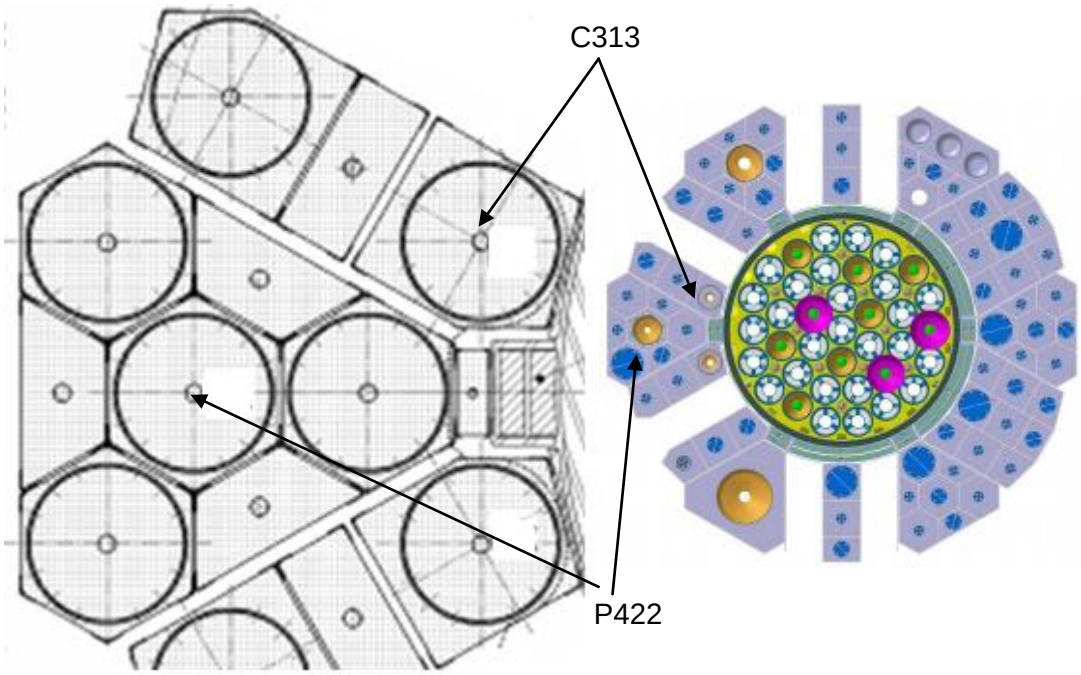



**Figure 3: Axial cut through TRIPOLI model of DIAMINO focused on a capsule**

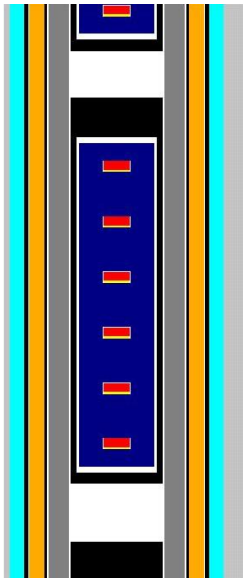


**Figure 4: Horizontal cut through the device with sample in the centre**

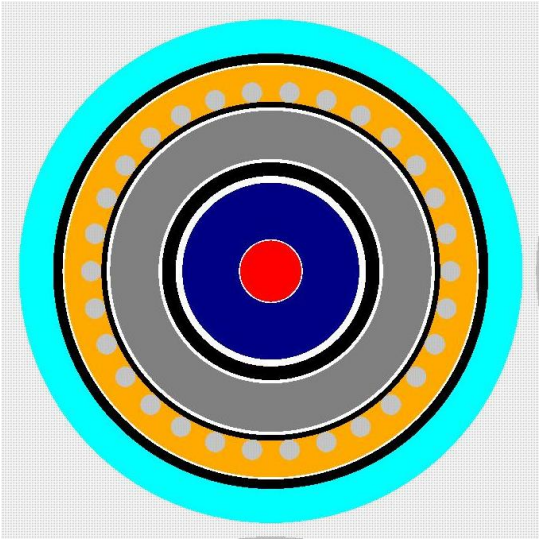


**Table 1: Reference case for each spectrum**

| | JHR P422 | JHR C313 | SFR blanket |
|---|---|---|---|
| Max total flux [n/s/cm²] | $3.43 \times 10^{13}$ | $4.78 \times 10^{14}$ | $7.59 \times 10^{14}$ |
| Energy groups | 172 | 172 | 33. |
| Reference sample | $Am_{0.15}U_{0.85}O_{2-x}$ | | |
| Density [g/cm³] | 10.3 | | |
| U mass composition | 0.25% $^{235}U$, 99.75% $^{238}U$ | | |

## Results

The originally considered variation range for the parameters were 5-20% Am, <20% U enrichment and >3% maximum flux but it turned out that the main thermal limit is the temperature gradient in the sample disc. A maximum allowable temperature gradient of 150°C limits the peak power to 650 W/cm$^3$, which in first order approximation limits the burn-up for 200 efpd to about 2.3% HM depletion (Figure 5). From the neutron physics point of view it could reach close to three times that burn-up. Figure 6 shows pronounced power peaks after the first 80 days of irradiation in the high flux cases due to $^{242m}Am$ (90% initial $^{241}Am$ depletion after about 250 efpd), which can be flattened by lower flux and higher uranium enrichment.

As a result not more than about 50% of the maximum possible flux can be used and this makes the use of the irradiation position C313 questionable unless the higher gamma heating is wanted for a high temperature experiment.

## Conclusion

While a DIAMINO-like experiment in JHR could be used for a parametric study of He content vs. burn-up in MA-bearing oxide fuel and would cover most of the expected range of SFR blanket scenarios, the low thermal conductivity of the oxide sample limits the maximum power level of the experiment. To increase the range of the burn-up it is either necessary to use even thinner samples (1.5 mm currently) or to increase the irradiation time. Increasing irradiation duration could have an additional benefit by checking for any kinetic effects on fuel behaviour, especially because SFR MABB aim for very long in core times, but it is really questionable from the experimental reactor operations point of view.

To get a limited data set to model He vs. burn-up behaviour a skeletal $3 \times 3$ matrix with seven points (one row, one column and the diagonal) is needed for a given sample temperature and porosity and this could be accommodated in a single irradiation position. Doing it for all the points in the MARIOS/DAIMINO matrix of four specific sample temperatures for two sample porosities (plus two capsules checking for kinetic effects) would need at least seven PHAETON devices (and as many reflector positions) for up to two years. Although parts of the programme could be done sequentially, if the full matrix is required, it would probably be beneficial to investigate an experimental rig, which allows for denser sample packing than PHAETON.

**Figure 5: Accessible range for 1 and 2 years duration with power below 650 W/cm$^3$**

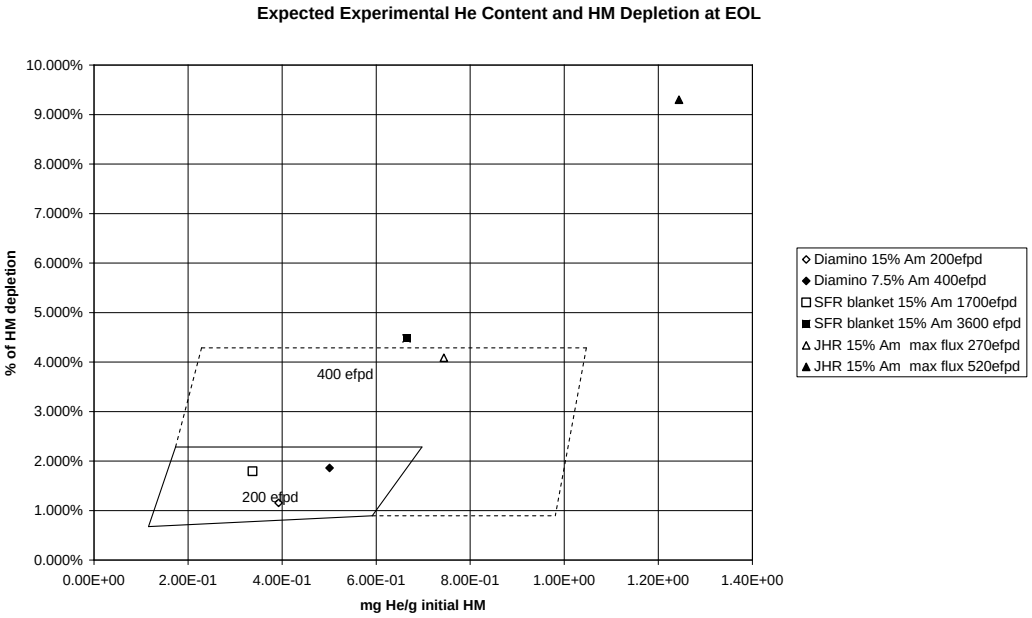
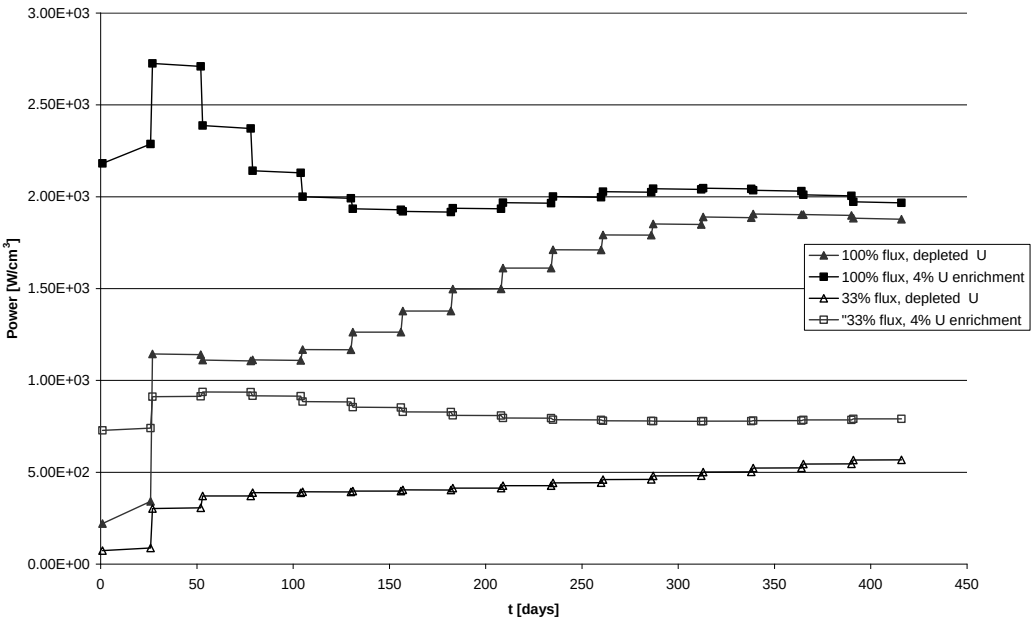



**Figure 6: Power development during irradiation in select samples**



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